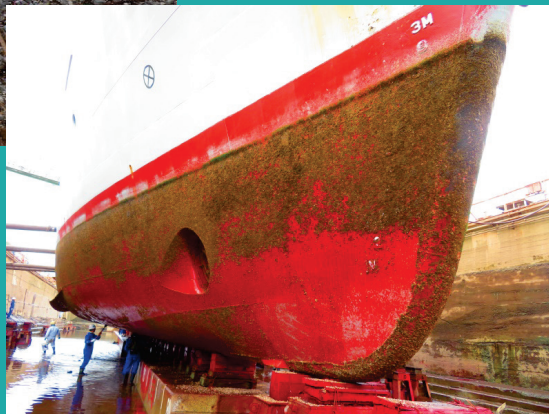
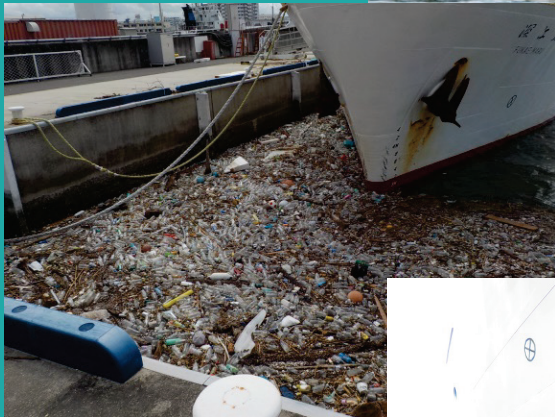
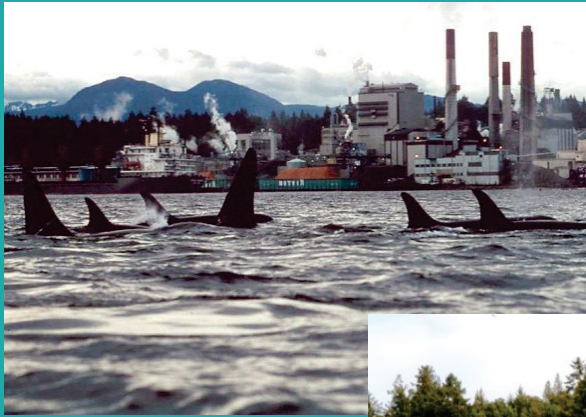


ISBN 978-1-927797-38-9
ISSN 1198-273X

PICES SCIENTIFIC REPORT

No. 56, 2020



NORTH PACIFIC MARINE SCIENCE ORGANIZATION



Report of Working Group 31 on Emerging Topics in Marine Pollution

PICES Scientific Report No. 56
2020

Report of Working Group 31 on
Emerging Topics in Marine Pollution

Edited by
Juan José Alava, Olga N. Lukyanova, Peter S. Ross and Won Joon Shim



February 2020

North Pacific Marine Science Organization (PICES)
P.O. Box 6000, Sidney, BC, V8L 4B2, Canada
www.pices.int

PICES Scientific Reports

Published since 1993, the PICES Scientific Report series includes final reports of PICES expert groups, proceedings of PICES workshops, data reports and reports of planning activities. Formal peer reviews of the scientific content of these publications are not generally conducted.

PICES Scientific Reports can be found at: <https://meetings.pices.int/publications/scientific-reports>

This report was developed under the guidance of the PICES Science Board and its Marine Environmental Quality Committee. The views expressed in this report are those of participating scientists under their responsibilities.

Front cover

From top: Killer whales (*Orcinus orca*) offshore from a pulp mill, Strait of Georgia (Photo credit: Dr. Lance Barrett-Lennard, Ocean Wise/Vancouver Aquarium); Steller sea lion (*Eumetopias jubatus*) entangled in netting gear in Baynes Sound, Strait of Georgia (Photo credit: courtesy of Denman Island resident, Association of Denman Island Marine Stewards-ADIMS); marine debris accumulation after the passing of a typhoon, Osaka Bay (Photo credit: Dr. Hideaki Maki, NIES); Kobe University T/S *Fukae-maru* undergoing antifouling procedure (Photo credit: Dr. Yohiji Yano, Kobe University).

This document should be cited as follows:

Alava, J.J., Lukyanova, O.N., Ross, P.S. and Shim, W.J. (Eds.) 2020. Report of Working Group 31 on Emerging Topics in Marine Pollution. PICES Sci. Rep. No. 56, 161 pp.

Table of Contents

In Memoriam.....	vii
Executive Summary	ix
Preamble.....	1
1 Canada	
1.1 Temporal trends for PCBs and PBDEs in harbour seals (<i>Phoca vitulina</i>) from British Columbia, Canada: 1984–2014 <i>Juan José Alava, Marie Noel and Peter S. Ross</i>	7
1.2 Trends of brominated flame retardants in marine birds from the Pacific coast of Canada: 1990–2015 <i>John E. Elliott, Aroha A. Miller, Kyle H. Elliott and Sandi Lee</i>	10
1.3 Recent trends in perfluoralkyl substances in marine birds from the Pacific coast of Canada: (1990–2011) <i>Aroha A. Miller, John E. Elliott, Kyle H. Elliott and Sandi Lee</i>	15
1.4 Temporal trends for mercury in aquatic bird eggs from the Pacific coast of Canada (1968–2015) <i>Kyle H. Elliott, John E. Elliott, Aroha A. Miller and Sandi Lee</i>	20
1.5 Historical mercury trends in sediments from coastal British Columbia, Canada <i>Juan José Alava and Marie Noel</i>	25
1.6 Temporal trends of marine contamination by copper in British Columbia, Canada: The legacy of the Britannia Mine <i>Juan José Alava, Karin Bodtker, Jennifer Chapman, Amber Dearden and Aroha Miller</i>	28
1.7 Assessment of marine debris pollution in the marine-coastal region of British Columbia, Canada <i>Kate Le Souef and Juan José Alava</i>	33
1.8 Spatial assessment of microplastic pollution in the marine-coastal region of British Columbia, Canada <i>Juan José Alava, Alejandra Díaz, Ellika Crichton, Esther Gies, Marie Noel and Peter S. Ross</i>	36
2 China	
2.1 Polychlorinated biphenyls (PCBs) in the marine environment of the China coast <i>Peng Zhang, Ying Bian, Chuanli Huo and Guangshui Na</i>	39
2.2 Organochlorine pesticides (OCPs) in the marine environment of the China coast	42
2.3 Polybrominated diphenyl ether (PBDE) flame retardants in the marine environment of the China coast <i>Xindong Ma, Chuanlin Huo and Guangshui Na</i>	45

2.4	Perfluorinated compounds (PFCs) in the marine environment of the China coast <i>Yao Yao, Ruijing Li, Chuanlin Huo and Guangshui Na</i>	48
2.5	Polycyclic aromatic hydrocarbons (PAHs) in the marine environment of the China coast <i>Zhen Wang, Chuanlin Huo and Guangshui Na</i>	51
3	Japan	
3.1	Temporal trends of PCB concentrations in mussels around the Japanese coast <i>Kazuhiko Mochida and Hiroyuki Tanaka</i>	54
3.2	Temporal and spatial trends of PCBs in coastal-marine sediments and seawater in Japan <i>Kazuhiko Mochida and Hiroyuki Tanaka</i>	56
3.3	Current status of marine pollution by antifouling biocides in Japan <i>Kazuhiko Mochida</i>	59
3.4	Distribution and temporal trends of mercury in sediments from coastal seas around Japan <i>Hideaki Maki</i>	62
4	Republic of Korea	
4.1	Accumulation and temporal trends of polychlorinated biphenyls (PCBs), organochlorine pesticides (OCPs) and polybrominated diphenyl ethers (PBDEs) in finless porpoises (<i>Neophocaena asiaeorientalis</i>) in the Republic of Korea <i>Hyo-Bang Moon</i>	66
4.2	Accumulation and temporal trends of polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/PCDFs) and dioxin-like polychlorinated biphenyls (DL-PCBs) in finless porpoises in the Republic of Korea <i>Hyo-Bang Moon</i>	69
4.3	Contamination status and temporal trends of polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/PCDFs) in sediments and bivalves from coastal areas in the Republic of Korea <i>Hyo-Bang Moon and Minkyu Choi</i>	72
4.4	Contamination status and temporal trends of polycyclic aromatic hydrocarbons (PAHs) in sediments and bivalves from coastal areas in the Republic of Korea <i>Hyo-Bang Moon and Minkyu Choi</i>	75
4.5	Temporal trends of antifouling biocides (Butyltins) in rock shells (<i>Thais clavigera</i>) and finless porpoises (<i>Neophocaena asiaeorientalis</i>) in the Republic of Korea <i>Minkyu Choi and Hyo-Bang Moon</i>	78
4.6	Contamination status and temporal trends of antifouling biocides (TBT, Diuron and Irgarol) in coastal areas in the Republic of Korea <i>Minkyu Choi, Byoung Seok Yoon and Won Joon Shim</i>	82
4.7	Contamination status and temporal trends of mercury (Hg) in mussels (<i>Mytilus edulis</i>) and oysters (<i>Crassostrea gigas</i>) from coastal areas in the Republic of Korea <i>Dong-Woon Hwang and Yong-Woo Lee</i>	85
4.8	Contamination status and temporal trends of mercury (Hg) in sediments from coastal areas in the Republic of Korea <i>Dong-Woon Hwang and Yong-Woo Lee</i>	88
4.9	Contamination status and temporal trends of lead (Pb) in mussels (<i>Mytilus edulis</i>) and oysters (<i>Crassostrea gigas</i>) from coastal areas in the Republic of Korea <i>Dong-Woon Hwang and Yong-Woo Lee</i>	90

4.10	Contamination status and temporal trends of lead (Pb) in sediments from coastal areas in the Republic of Korea <i>Dong-Woon Hwang and Yong-Woo Lee</i>	92
4.11	Contamination status and temporal trends of marine debris at beaches in the Republic of Korea <i>Sunwook Hong and Won Joon Shim</i>	94
5	Russia	
5.1	The human impact on the mercury accumulation in modern sediments of Amur Bay (the Sea of Japan) <i>Kirill Aksentov</i>	96
5.2	Inter-annual dynamics of Pb concentrations in bottom sediments of Peter the Great Bay (the Sea of Japan) around Vladivostok, Russia <i>Alexander V. Sevastianov, Tatyana S. Lishavskaya and Tatyana A. Belan</i>	99
5.3	Organochlorine pesticides in tissues of marine mammals from the western part of the Bering Sea, Russia <i>Olga Lukyanova and Vasiliy Tsygankov</i>	101
5.4	Microplastic monitoring in the coastal area of the Peter the Great Bay (Sea of Japan), Russia <i>Yana Blinovskaya and Nikolai Kozlovskii</i>	103
6	United States of America	
6.1	Temporal trends in polycyclic aromatic hydrocarbons (PAHs) in blue mussels (<i>Mytilus</i> spp.) from marine waters of Washington State <i>Jennifer Lanksbury, Alan Mearns, James West and Gina Ylitalo</i>	106
6.2	NOAA Marine Debris Monitoring and Assessment Project: Four years of effort in the U.S. Pacific States <i>Sherry Lippiatt and Carlie Herring</i>	109
	Summary and Recommendations	117
	Appendix 1 WG 31 Terms of Reference	118
	Appendix 2 WG 31 Membership	119
	Appendix 3 WG 31's Contribution to the North Pacific Ecosystem Status Report	122
	Appendix 4 Journal Publications.....	125
	Appendix 5 Topic Session/Workshop Summaries and Meeting Reports from Past Annual Meetings	131

In Memoriam



Dr. Olga Nikolaevna Lukyanova

This report is dedicated to the memory of Dr. Olga Nikolaevna Lukyanova who passed away on January 19, 2019 after losing a battle with cancer.

Executive Summary

Pollution can adversely affect the health and abundance of marine biota, especially in densely populated coastal areas of PICES member countries. The downstream socio-economic and public health consequences can be significant, with numerous examples of consumption advisories, commercial fishery closures, and commercial trade interdictions and diminished aboriginal access to food resources around the North Pacific Ocean. The protection of ecosystem health and services requires an ability to detect emerging pollutant issues before serious adverse impacts arise. Regulations, policies and other management actions resulting from marine pollution research in the past have led to dramatic declines in environmental concentrations of a number of harmful pollutants, subsequently improving the health of marine biota.

The use, release, and/or disposal of approximately 100,000 chemicals on the global market has direct implications for the health of the world's oceans since they represent the ultimate 'sinks' for anthropogenic pollutants. While risk assessments and/or technical documentation provide decision makers in national governments with basic chemical, physical, and toxicological information in support of national regulations, there is very little information on the fate and effects of chemicals in the marine environment (Ross, 2014). The diversity of contaminants entering the ocean from human activities makes it necessary to summarize and prioritize contaminants by broad category. This underscores the distinct need for research, and for information sharing, in the area of marine pollution in the North Pacific.

Following the PICES Study Group on *Marine Pollutants* (SG-MP) recommendations, a new Working Group on *Emerging Topics in Marine Pollution* was established in 2014 to build on the foundation of past and current activities regarding marine pollution in the North Pacific. This Working Group, known as WG 31, was very active and was able to fulfill its terms of reference by convening Topic Sessions at every Annual Meeting during its 3-year term, including one at PICES-2017, and by publishing three special issues on selected papers from its Topic Sessions on "Microplastics in the Ocean", "Indicators of Marine Pollution in the North Pacific Ocean" and "Source, Transport and Fate of Hydrocarbons in the Marine Environment" in the peer-reviewed journal *Archives of Environmental Contamination and Toxicology* (TOR1). WG 31 fulfilled TOR2 by making a notable contribution on pollution status and temporal trend reports to the next PICES North Pacific Ecosystem Status Report (NPESR). A third in a series, NPESR3 is the first to discuss pollutants. WG 31 also collaborated closely with other organizations, such as GESAMP, NOWPAP and ICES, who were active participants at its meetings and who acted as co-sponsors of its sessions and workshop (TOR3).

In addition, WG 31 helped to address the question identified in the FUTURE Science Plan "How do human activities affect coastal ecosystems and how are societies affected by changes in these ecosystems?" by recognizing that:

- Pollution can adversely affect the health and abundance of marine biota, especially in densely-populated coastal areas;

- There are socio-economic consequences of coastal pollution, with consumption advisories, fishery closures, trade interdictions and diminished aboriginal access to food resources;
- Regulations, policies and other management actions resulting from marine pollution research in the past have led to declines in the concentrations of a number of harmful pollutants, improving the health of marine biota.

In this context, the Working Group on *Emerging Topics in Marine Pollution* recommends PICES:

- To continue the concerted and long-term characterization of emerging pollutants or pollution priorities in the North Pacific Ocean to determine the dominant anthropogenic pressures in coastal marine ecosystems and how they are changing and affecting societies depending upon marine resources;
- To keep track and ensure the compilation of pollutant data and monitoring programs conducted by its six member countries;
- To continue documenting the impacts of emerging pollutants in the North Pacific Ocean, especially in coastal environments, and in a changing ocean environment in the face of climate change forcing, including increasing sea surface temperature, sea bottom temperature, deoxygenation/hypoxia, ocean acidification, sea level rise and changes in primary production affecting nearshore and coastal ecosystems and their interactions with offshore and terrestrial systems;
- To engage and enhance collaboration with other expert groups of PICES to document the importance of marine pollution relative to multiple anthropogenic stressors and associated interactions and cumulative effects;
- To develop approaches to pollutant indicators that account for climate variability and change to understand the consequences of projected coastal ecosystem changes as well as taking into account the predictability and uncertainty of forecasted changes by characterizing changing pollution risks as climate changes;
- To prioritize pollutant sources of contaminants of emerging concern (*e.g.*, microplastics, pharmaceuticals and personal care products) and types in support of source control, regulations and best management practices.
- To enact and enforce regulations that prevent chemicals from being produced, sold and/or used commercially unless these substances are tested for and found not to exhibit bioaccumulation, toxicity and/or other harmful effects in the marine environment.

Reference

Ross, P.S. (Ed.) 2014. Report of the Study Group on Marine Pollutants. PICES Sci. Rep. No. 46, 49 pp.

Preamble

Ocean contamination by anthropogenic pollutants, including persistent organic pollutants (POPs), metals, antifouling biocides, and marine debris is an issue of serious concern because these contaminants are ubiquitous in the environment, detected at relatively high concentrations, and driven by trans-Pacific air pollution from Asia to the Pacific coast of North America (Wilkening *et al.*, 2000). Long-range atmospheric transport of POPs from temperate, subtropical and tropical areas also plays an important role in delivering these contaminants to oceanic and coastal regions of the North Pacific (Iwata *et al.*, 1993, 1994; Wania and Mackay, 1996; Wania and Dugani, 2003; Gouin *et al.*, 2004).

POPs are “a set of organic compounds that: a) possess toxic characteristics; b) are persistent; c) are liable to bioaccumulate; d) are prone to long-range atmospheric transport and deposition; and e) can result in adverse environmental and human health effects at locations near and far from their sources” (UNEP, 2002). POPs are regulated by the Stockholm Convention (UNEP, 2010). These compounds can be categorized as legacy (classic) POPs such as polychlorinated biphenyls (PCBs), dichloro-diphenyl-trichloroethanes (DDTs) and hexachlorocyclohexanes (HCHs), as well as emerging POPs such as polybrominated diphenyl ethers (PBDEs) and perfluorinated chemicals (PFCs), which were recently categorized as POPs by the Stockholm Convention on POPs (UNEP, 2010). The toxicity of POPs on human health and wildlife is of major concern because these compounds bioaccumulate and cause toxic effects, affecting the health and reproduction in marine organisms. Although some POPs, including PCBs and DDTs, were banned in developed and industrialized countries during the 1970s, some organochlorine pesticides (OCPs) are still used in developing countries to control malaria vectors and crop pests (*i.e.*, DDT) and, along with emerging POPs such as PBDE flame retardants and PFCs, continue to contaminate food webs. The highest concentrations of PCBs and DDTs still tend to be reported from locations in temperate countries where usage was very intense or where POPs were manufactured, stored or underwent ocean disposal (*e.g.*, Palos Verdes in Southern California Bight, Blasius and Goodmanlowe, 2008). POPs are still being found in relatively high concentrations in apex predators from the North Pacific; therefore, continued monitoring and assessment of POPs is critical for countries within PICES.

Although metals occur naturally in the environment, they can become a concern to marine organisms in localized areas such as urban and industrial centers where concentrations are elevated due to anthropogenic emissions. Anthropogenic sources of metals include mines, metal refineries/smelters, fossil fuel combustion, waste incineration, pesticides and wood preservatives, and release from domestic and industrial waste. A particular metal of concern in the global environment is mercury (Hg), including the highly toxic organic forms such as methylmercury (CH_3Hg^+) found in marine ecosystems. Mercury contamination in the global ocean is especially worrisome due to its toxicity, ubiquitous nature, and potential to accumulate (as methylmercury) in aquatic food webs. Mercury is emitted from both natural and anthropogenic sources. The latter is dominated by small-scale gold mining operations (37%) and coal-fired power plants (24%) which contaminate remote oceanic regions and marine biota, including economically important fisheries (UNEP, 2013; Lamborg *et al.*, 2014). Anthropogenic emissions and releases have doubled the amount of mercury in the top 100 meters of the world’s oceans

in the last 100 years (UNEP, 2013). The total amount of anthropogenic mercury present in the global oceans has been estimated to be 290 million moles, of which approximately 66.67% reside in waters shallower than 1000 m (Lamborg *et al.*, 2014). Despite regulatory measures, recent estimates still suggest future increases in global anthropogenic mercury emissions leading to a continued increase of Hg concentrations in the global ocean (Mason *et al.*, 2012; UNEP, 2013). The North Pacific is one of the major sinks for mercury emissions, and methylmercury bioaccumulation in apex predators has been documented in sentinel species from the northeastern Pacific (Peterson *et al.*, 2015; Noël *et al.*, 2016).

The Minamata Convention on Mercury (*i.e.*, adopted on October 10, 2013 in Kumamoto, Japan) is a global treaty that aims to protect human health and the environment from the adverse effects of mercury. The major highlights of the Convention include a ban on new mercury mines, the phasing out of existing ones, control measures on air emissions, and international regulation of the informal sector for artisanal and small-scale gold mining (UNEP, 2016). As for POPs, continued implementation of pollution programs to track and monitor mercury levels and trends in sediments and bioaccumulation (methylmercury) in marine biota from the North Pacific is of paramount importance in understanding the behavior of this pollutant in the face of global environmental change. Mercury has been detected in sediments from Korea, Russia and Canada, raising concerns about chronic contamination of the marine environment and metal pollution risk for seafoods in these countries. Similarly, copper (Cu) and lead (Pb) are of concern due to recent cases involving the release of these chemicals and measurements showing moderate concentrations in some countries from the North Pacific (*e.g.*, Cu in Canada and Pb in Korea and Russia).

Hydrocarbon pollution from oil spills and increasing maritime tanker traffic in the North Pacific (*e.g.*, eastern Asia and northeastern Pacific coastal–marine regions) cannot be ruled out as potential threats for marine organisms and their habitats. Such incidents might include direct impacts of oil spills from coastal-based infrastructural operations (*i.e.*, terminal and pipeline spills) and chemical spills from tanker accidents. A classic example of oil spill and hydrocarbon effects in a marine species can be seen in sea otters. During events involving chemical or oil spills, the pollutants of greatest concern include volatile organic compounds (VOCs) and polycyclic aromatic hydrocarbons (PAHs) (Ross and Desforges, 2014). While acute catastrophic oil spills are among the more obvious sources of chemical pollution at sea and along fragile coastal areas, chronic spills of hydrocarbons from multiple sources (*e.g.*, runoff from impervious surfaces on urbanized areas and parking lots, small boats, fishing vessels, marinas and fuel stations) represent an ongoing and largely uncharacterized threat for marine mammals (Harris *et al.*, 2011). For instance, the volume of used motor oil spilled annually into Canadian waters is equivalent to seven times the amount released by the Exxon Valdez tanker in 1989 (de Villiers, 1999). The lesson learned from the Exxon Valdez ecological disaster in Alaska is a poignant reminder of the pollution risks by oil spills and the concerted emergency plans and mitigation actions required to minimize the potential impacts of chemical pollution in the North Pacific.

Organometallic biocides, including tributyltin (TBT) and other organotin (OT) compounds, are used mainly as antifouling paints on ship hulls and readily bioaccumulate in marine mammals and humans (Tanabe, 1999; Linley-Adams, 1999; Antizar-Ladislao, 2008). TBT compounds have been associated with immunotoxicity and hepatotoxicity in sea otters, bottlenose dolphins and Dall's porpoises (Kannan *et al.*, 1997, 1998; Nakata *et al.*, 2002; Murata *et al.*, 2008). Because of their toxic and bioaccumulative nature, these substances have been banned in several countries from the North Pacific, including Korea, Japan and Canada, but new substitutes (*i.e.*, OT-free antifouling biocides) such as Diuron and Irgarol are now being used and have already been found in the coastal marine environments of Korea and Japan since the banning of TBT. Despite TBT compounds being prohibited in Canada since January 1, 2003

under the *Pest Control Products Act* (PCPA; Environment Canada and Health Canada, 2009), two tributyltin pesticide active ingredients have since been registered under the PCPA and are found in six end-use products (Environment Canada and Health Canada, 2009).

Marine debris, notably plastic, is one of the major threats for marine life and oceans. Marine debris and plastics can lead to entanglement and ingestion, leach toxic chemicals, and provide vectors for POPs and invasive species (Moore, 2008; GESAMP, 2010). It is known that 26 species of cetaceans can ingest plastic bags, fishing line and other plastic (Moore, 2008). Of particular concern are microplastics, defined as particles < 5 mm, which can be deliberately manufactured (plastic resin pellets and powder) or generated as breakdown by-products of larger debris and macroplastic (*e.g.*, clothing, ropes, bags, bottles) (Moore, 2008; GESAMP, 2010). Most plastics are derived from land-based sources, including household and industrial water, aquaculture, shipping and tourism.

In situ observations as well as global microplastic transportation models have shown higher microplastic abundance in the North Pacific and its marginal seas than in other oceans (Desforges *et al.*, 2014; Lusher, 2015; Van Sebille *et al.*, 2015; Lebreton *et al.*, 2018). While further research to assess the impact of plastics or microplastics in the North Pacific is needed, research efforts to understand and assess the abundance, distribution, exposure sources and bioaccumulation potential of microplastics in the marine environment and organisms are currently underway in Canada, China, Japan, Korea, Russia and USA (Doyle *et al.*, 2011; Desforges *et al.*, 2014, 2015; Song *et al.*, 2014; Cluzard *et al.* 2015; Isobe *et al.*, 2015; Kozlovskii *et al.*, 2017; Peng *et al.*, 2017; Cai *et al.*, 2018; Kazmiruk *et al.*, 2018; Collicutt *et al.*, 2019; Covernton *et al.*, 2019; Pan *et al.*, 2019; Alava, 2020; Blinovskaya *et al.*, 2020). Additional efforts must be undertaken by PICES member countries to monitor and track plastic pollution in the North Pacific.

The following sections describe the activities of Working Group 31 members, and colleagues, through their respective country reports on the status and trends of four target pollutant classes: POPs, metals/elements of concern, hydrocarbons, and marine debris and microplastics. The report completes with five Appendices: Working Group 31's terms of reference, membership, a review of its contribution to NPESR3, a list of papers in three special issues in a peer-reviewed journal, and topic session/workshop summaries and meeting reports from PICES Annual Meetings.

References

- Alava, J.J. 2020. Modelling the bioaccumulation and biomagnification potential of microplastics in a cetacean foodweb of the Northeastern Pacific: A prospective tool to assess the risk exposure to plastic particles. *Frontiers in Marine Science* **7**: 566101, doi: 10.3389/fmars.2020.566101.
- Antizar-Ladislao, B. 2008. Environmental levels, toxicity and human exposure to tributyltin (TBT)-contaminated marine environment. A review. *Environment International* **34**: 292–308.
- Blasius, M.E. and Goodmanlowe, G.D. 2008. Contaminants still high in to-level carnivores in the Southern California Bight: Levels of DDT and PCBs in resident and transient pinnipeds. *Marine Pollution Bulletin* **56**: 1973–1982.
- Blinovskaya, Y., Zakharenko, A., Golokhvast, K., Chernysh, O. and Zubtsova, I. 2020. Microplastic in the coastal sea waters of Russian Far East. *IOP Conference Series: Earth and Environmental Science* **459**: 052068. doi:10.1088/1755-1315/459/5/052068.

- Cai, M., He, H., Liu, M., Li, S., Tang, G., Wang, W., Huang, P., Wei, G., Lin, Y., Chen, B. and Hu, J. 2018. Lost but can't be neglected: huge quantities of small microplastics hide in the South China Sea. *Science of the Total Environment*, **633**: 1206–1216.
- Cluzard, M., Kazmiruk, T.N., Kazmiruk, V.D. and Bendell, L.I. 2015. Intertidal concentrations of microplastics and their influence on ammonium cycling as related to the shellfish industry. *Archives of Environmental Contamination and Toxicology* **69**: 310–319.
- Collicutt, B., Juanes, F. and Dudas, S.E. 2019. Microplastics in juvenile Chinook salmon and their nearshore environments on the east coast of Vancouver Island. *Environmental Pollution* **244**: 135–142.
- Covernton, G.A., Collicutt, B., Gurney-Smith, H.J., Pearce, C.M., Dower, J.F., Ross, P.S. and Dudas, S.E. 2019. Microplastics in bivalves and their habitat in relation to shellfish aquaculture proximity in coastal British Columbia, Canada. *Aquaculture Environment Interactions* **11**: 357–374.
- Desforges, J.P.W., Galbraith M., Dangerfield, N. and Ross, P.S. 2014. Widespread distribution of microplastics in subsurface seawater in the NE Pacific Ocean. *Marine Pollution Bulletin* **79**: 94–99.
- Desforges, J.P.W., Galbraith, M. and Ross, P.S. 2015. Ingestion of microplastics by zooplankton in the Northeast Pacific Ocean. *Archives of Environmental Contamination and Toxicology* **69**: 320–330.
- de Villiers, M. 1999. Water. Stoddart Publishing Co. Toronto, Canada.
- Doyle, M.J., Watson, W., Bowlin, N.M. and Sheavly, S.B. 2011. Plastic particles in coastal pelagic ecosystems of the Northeast Pacific ocean. *Marine Environmental Research* **71**: 41–52.
- Environment Canada and Health Canada. 2009. Proposed risk management approach for non-pesticidal organotin compounds (Organotins) (Non-Pesticidal Organotins). Government of Canada. 15 pp.
- GESAMP (Joint Group of Experts on the Scientific Aspects of Marine Environmental Protection). 2010. Proceedings of the GESAMP International Workshop on plastic particles as a vector in transporting persistent, bio-accumulating and toxic substances in the oceans *in*: GESAMP Report Study No. 82 *edited by* T. Bowmer and P.J. Kershaw, 68 pp.
- Gouin, T., Mackay, D., Jones, K.C., Harner, T. and Meijer, S.N. 2004. Evidence for the “grasshopper” effect and fractionation during long-range atmospheric transport of organic contaminants. *Environmental Pollution* **128**: 139–148.
- Harris, K.A, Yunker, M.B, Dangerfield, N. and Ross, P.S. 2011. Sediment-associated aliphatic and aromatic hydrocarbons in coastal British Columbia, Canada: Concentrations, composition, and associated risks to protected sea otters. *Environmental Pollution* **159**: 2665–2674.
- Isobe, A., Uchida, K., Tokai, T. and Iwasaki, S. 2015. East Asian seas: A hot spot of pelagic microplastics. *Marine Pollution Bulletin* **101**: 618–623.
- Iwata, H., Tanabe, S. Sakai, N. and Tatsukawa, R. 1993. Distribution of persistent organochlorines in the oceanic air and surface seawater and the role of ocean on their global transport and fate. *Environmental Science and Technology* **27**: 1080–1098.
- Iwata, H., Tanabe, S., Sakai, N. Nishimura, A. and Tatsukawa, R. 1994. Geographical distribution of persistent organochlorines in air, water and sediments from Asia and Oceania, and their implications for global redistribution from lower latitudes. *Environmental Pollution* **85**: 15–33.
- Kannan, K., Senthilkumar, K., Loganathan, B.G., Takahashi, S., Odell, D.K. and Tanabe, S. 1997. Elevated accumulation of tributyltin and its breakdown products in bottlenose dolphins (*Tursiops truncatus*) found stranded along the U.S. Atlantic and Gulf coasts. *Environmental Science and Technology* **31**: 296–301.

- Kannan, K., Guruge, K.S., Thomas, N.J., Tanabe, S. and Giesy, J.P. 1998. Butyltin residues in southern sea otters (*Enhydra lutris nereis*) found dead along California coastal waters. *Environmental Science and Technology* **32**: 1169–1175.
- Kazmiruk, T.N., Kasmiruk, V.D. and Bendell, L.I. 2018. Abundance and distribution of microplastics within surface sediments of a key shellfish growing region of Canada. *PloS ONE* **13**: e0196005, <https://doi.org/10.1371/journal.pone.0196005>.
- Kozlovskii, N.V., Hong, S.H., Song, Y.K. and Kachur, A.N. 2017. Distribution of beached marine litter and floating microplastics in the Minonosok Inlet of the Possiet Bay of the Peter the Great Gulf, pp. 235–239 in: Resources, Environment and Regional Sustainable Development in Northeast Asia, Proceedings of the III International Conference, October 10–14, 2016, Vladivostok, Russia.
- Lamborg, C.H., Hammerschmidt, C.R., Bowman, K.L., Swarr, G.J., Munson, K.M., Ohnemus, D.C., Lam, P.J., Heimbürger, L.E., Rijkenberg, M.J.A. and Saito, M.A. 2014. A global ocean inventory of anthropogenic mercury based on water column measurements. *Nature* **512**: 65–68.
- Lebreton, L., Slat, B., Ferrari, F., Sainte-Rose, B., Aitken, J., Marthouse, R., Hajbane, S., Cunsolo, S., Schwarz, A., Levivier, A. and Noble, K., 2018. Evidence that the Great Pacific Garbage Patch is rapidly accumulating plastic. *Scientific Reports* **8**: 1–15.
- Linley-Adams, G. 1999. The accumulation and impact of organotins on marine mammals, seabirds and fish for human consumption. Report for World Wildlife Fund -UK, WWF-UK Project No 98054, 26 pp.
- Lusher, A.L. 2015. Microplastics in the marine environment: Distribution, interaction, and effects, pp. 245–307 in: *Marine Anthropogenic Litter* edited by M. Bergmann, L. Gutow and M. Klages, Springer, New York.
- Mason, R.P., Choi, A.L., Fitzgerald, W.F., Hammerschmidt, C.R., Lamborg, C.H., Soerensen, A.L. and Sunderland, E.M. 2012 Mercury biogeochemical cycling in the ocean and policy implications. *Environmental Research* **119**: 101–117.
- Moore, C.J. 2008. Synthetic polymers in the marine environment: A rapidly increasing, long-term threat. *Environmental Research* **108**: 131–139.
- Murata, S., Takahashi, S., Agusa, T., Thomas, N.J., Kannan, K. and Tanabe, S. 2008. Contamination status and accumulation profiles of organotins in sea otters (*Enhydra lutris*) found dead along the coasts of California, Washington, Alaska (USA), and Kamchatka (Russia). *Marine Pollution Bulletin* **56**: 641–649.
- Nakata, H., Sakakibara, A., Kanoh, M., Kudo, S., Watanabe, H., Nagai, N., Miyazaki, N., Asano, Y. and Tanabe, S. 2002. Evaluation of mitogen-induced responses in marine mammal and human lymphocytes by in-vitro exposure of butyltins and non-ortho coplanar PCBs. *Environmental Pollution* **120**: 245–253.
- Noël, M., Jeffries, S., Lambourn, D.M., Telmer, K., Macdonald, R. and Ross, P.S. 2016. Mercury accumulation in harbour seals from the Northeastern Pacific Ocean: The role of transplacental transfer, lactation, age and location. *Archives of Environmental Contamination and Toxicology* **70**: 56–66.
- Pan, Z., Liu, Q., Sun, Y., Sun, X. and Lin, H. 2019. Environmental implications of microplastic pollution in the Northwestern Pacific Ocean. *Marine Pollution Bulletin* **146**: 215–224.
- Peng, G., Zhu, B., Yang, D., Su, L., Shi, H. and Li, D. 2017. Microplastics in sediments of the Changjiang Estuary, China. *Environmental Pollution* **225**: 283–290.
- Peterson, S.H., Ackerman, J.T. and Costa, D.P. 2015. Marine foraging ecology influences mercury bioaccumulation in deep diving northern elephant seals. *Proceedings of the Royal Society B* **282**: 20150710, <https://doi.org/10.1098/rspb.2015.0710>.
- Ross, P.S. (Ed.) 2014. Report of the Study Group on Marine Pollutants. PICES Sci. Rep. No. 46, 49 pp.

Preamble

- Ross, P.S. and Desforges, J.P. 2014. Towards a framework for organizing a forward-looking pollution workshop for SARA-listed marine mammals in February 2015. Report prepared for Fisheries and Ocean Canada, Fisheries Management–Species at Risk, March 18, 2014. Ocean Pollution Research Program, Vancouver Aquarium Marine Science Centre, 31 pp.
- Song, Y.K., Hong, S.H., Jang, M., Kang, J.H., Kwon, O.Y., Han, G.M. and Shim, W.J. 2014. Large accumulation of micro-sized synthetic polymer particles in the sea surface microlayer. *Environmental Science and Technology* **48**: 9014–9021.
- Tanabe, S. 1999. Butyltin contamination in marine mammals – A review. *Marine Pollution Bulletin* **39**: 62–72.
- UNEP (United Nations Environment Programme). 2002. Protecting the environment from Persistent Organic Pollutants and other persistent toxic substances. UNEP’s Action in the Framework of the Global Environment Facility. UNEP/GEF. Geneva, Switzerland, 42 pp.
- UNEP. 2010. The Nine New POPs: an introduction to the nine chemicals added to the Stockholm Convention by the Conference of the Parties at its fourth meeting. United Nations Environment Program Secretariat of the Stockholm Convention on Persistent Organic Pollutants.
- UNEP. 2013. Global Mercury Assessment 2013: Sources, Emissions, Releases and Environmental Transport. UNEP Chemicals Branch, Geneva, Switzerland.
- UNEP 2016. Minamata Convention on Mercury. <http://www.mercuryconvention.org/Convention>.
- Van Sebille, E., Wilcox, C., Lebreton, L., Maximenko, N., Hardesty, B.D., van Franeker, J.A., Eriksen, M., Siegel, D., Galgani, F. and Law, K.L. 2015. A global inventory of small floating plastic debris. *Environmental Research Letters* **10**: 214006.
- Wania, F. and Dugani, C. 2003. Assessing the long-range transport potential of Polybrominated Diphenyl Ethers: A comparisons of four multimedia models. *Environmental Toxicology and Chemistry* **22**: 1252–1261.
- Wania, F. and Mackay, D. 1996. Tracking the distribution of persistent organic pollutants. *Environmental Science and Technology* **30**: 390A–296A.
- Wilkening, K.E., Barrie, L.A. and Engle, M. 2000. Trans-Pacific air pollution. *Science* **290**: 65–66.

1 Canada

1.1 Temporal trends for PCBs and PBDEs in harbour seals (*Phoca vitulina*) from British Columbia, Canada: 1984–2014

Juan José Alava^{1,2}, Marie Noel² and Peter S. Ross²

¹Institute for the Oceans and Fisheries, University of British Columbia, Vancouver, British Columbia, Canada

²Ocean Pollution Research Program, Coastal and Ocean Research Institute, Ocean Wise Conservation Association, Vancouver, British Columbia, Canada

Polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs) are two classes of persistent organic pollutants (POPs) that have been measured continuously in sediments and top predators of the coastal–marine region of British Columbia (BC), Canada (Rayne *et al.*, 2004; Grant *et al.*, 2011; Alava *et al.*, 2012, 2016; Ross *et al.*, 2013; Alava, 2019). While PCBs were banned in Canada in 1979, PBDEs have only recently been phased out in Canada and USA, due to their persistence, bioaccumulative and toxic nature (Ross *et al.*, 2009). The Strait of Georgia is adjacent to Vancouver in BC and is considered the receiving environment for pollutants from industries, agriculture, forestry, and other local human activities (Grant *et al.*, 2011; Alava, 2019). Killer whales (*Orcinus orca*) in this region are among the most PCB-contaminated organisms in the world, raising concerns for their long-term survival and recovery (Hickie *et al.*, 2007). While PBDEs were readily detected at levels of concern in migratory Steller sea lions (Alava *et al.*, 2012), harbour seals (*Phoca vitulina*) from the Strait of Georgia serve as sentinels of coastal food web contamination by POPs because of their non-migratory behavior, high trophic position in the food web and long lives (Ross *et al.*, 2013). Here, we report on the status of temporal trends in PCBs and PBDEs based on the research conducted with harbour seals in BC.

PCB and PBDE trends in harbour seals

PCBs and PBDEs were measured in blubber biopsies collected from live-captured harbour seal pups at two locations in BC (Hornby Island and the Fraser River estuary in the Strait of Georgia) in 2003 (Ross *et al.*, 2013). Estimated contaminant data in harbour seals for 1984, 1990, 1993, 1997, 2009 and 2014 are also presented in this report. Estimated temporal concentration data for PCBs and PBDEs in harbour seals in the Strait of Georgia from 1984 to 2014 reveal a history of accumulation of these POPs in this species (Fig. 1.1.1). Harbour seals exhibited declining concentrations of PCBs with 81% reduction from 1984 to 2009. PBDEs showed an increase in concentrations of 99% from 1984 to 2003 but then reached a steady state until 2009 when concentrations dropped, reflecting the withdrawal of penta- and octa-formulations from the market in 2004 (Ross *et al.*, 2009). Temporal predictions (1984–2014) in terms of the estimated total mass (kg) of PCBs and PBDEs in this species show a similar trend, with the total load of PBDEs in seals dropping by 2009 (Ross *et al.*, 2013; Fig. 1.1.2). Harbour seals were estimated

to contain a total of 1.70 kg of PCBs in 2009, a significant decline (73%) from the estimate total of 6.30 kg in 1997 (Fig. 1.1.2). These seals also contained an estimated 0.63 kg of PBDEs in 2009, 31% lower than the total mass of PBDEs (0.90 kg) in the peak year (2003). While the harbour seal data indicate that PCB levels have declined and PBDEs reached steady state in the region, recent food-web bioaccumulation modelling work highlighted that long-lived and higher trophic level species, including resident killer whale populations, still exceed the total PCB risk-based toxic effect concentration of 1.3 mg/kg lipid for harbour seals (Mos *et al.*, 2010) and the PBDE-endocrine disruption threshold of 1.5 mg/kg reported for grey seals (Hall *et al.*, 2003) in several critical habitats of BC (Alava *et al.*, 2016). In general, the contaminant data suggest that past and current regulations and source controls have markedly reduced inputs of these pollutants to the Strait of Georgia, consequently reducing the associated health risks to marine wildlife. However, monitoring and further research should continue tracking the trends of POPs in the BC marine environment as recycling and disposal at sea operations in critical habitats in combination with climate change may conspire against the recovery of endangered marine mammal species in this region (Alava *et al.*, 2018; Alava, 2019).

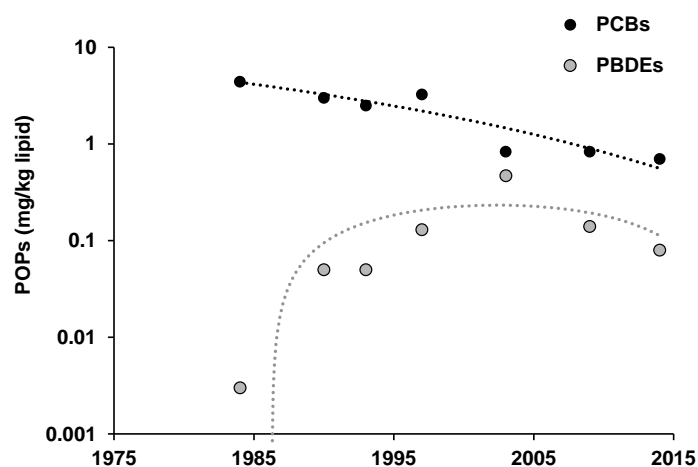


Fig. 1.1.1 Estimated temporal trends in PCB and PBDE concentrations (log scale in mg/kg lipid) in harbour seals from the Strait of Georgia (BC, Canada), based on the trends observed in harbour seals from Puget Sound, WA, USA (Ross *et al.*, 2013).

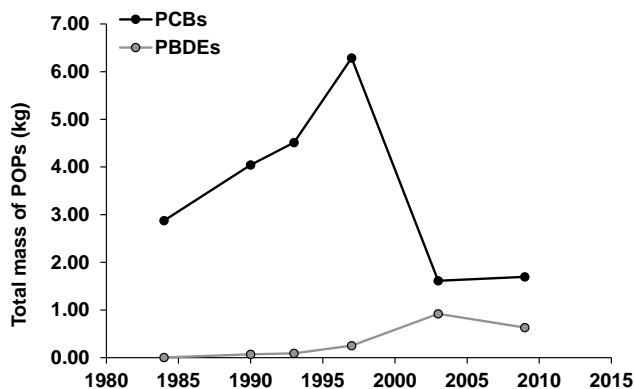


Fig. 1.1.2 Total mass of PCBs and PBDEs (kg) estimated for the free-ranging population of harbour seals inhabiting the Strait of Georgia. Adapted from Ross *et al.* (2013).

References

- Alava, J.J. 2019. Legacy and emerging pollutants in marine mammals' habitat from British Columbia: Management perspectives for sensitive marine ecosystems, pp. 87–114 *in*: Stewarding the Sound: The Challenge of Managing Sensitive Ecosystems *edited by* L.I. Bendell, P. Gallagher, L. Wood and S. McKeachie, CRC Press/Taylor and Francis Group. 148 pp.
<https://www.crcpress.com/Stewarding-the-Sound-The-Challenge-of-Managing-Sensitive-Coastal-Ecosystems/Bendell-Gallagher-McKeachie-Wood/p/book/9780367112035>.
- Alava, J.J., Lambourn, D., Olesiuk, P., Lance, M., Jeffries, S.J., Gobas, F.A. and Ross, P.S., 2012. PBDE flame retardants and PCBs in migrating Steller sea lions (*Eumetopias jubatus*) in the Strait of Georgia, British Columbia, Canada. *Chemosphere* **88**: 855–864.
- Alava, J.J., Ross, P.S. and Gobas, F.A. 2016. Food web bioaccumulation model for resident killer whales from the Northeastern Pacific Ocean as a tool for the derivation of PBDE-sediment quality guidelines. *Archives of Environmental Contamination and Toxicology* **70**: 155–168.
- Alava, J.J., Cisneros-Montemayor, A.M., Sumaila, R., and Cheung, W.W.L. 2018. Projected amplification of food web bioaccumulation of MeHg and PCBs under climate change in the Northeastern Pacific. *Scientific Reports* **8**: 13460 doi:10.1038/s41598-018-31824-5.
- Grant, P.B.C., Johannessen, S.C., Macdonald, R.W., Yunker, M., Sanborn, M., Dangerfield, N., Wright, C. and Ross, P.S. 2011. Environmental fractionation of PCBs and PBDEs during particle transport as recorded by sediments in coastal waters. *Environmental Toxicology and Chemistry* **30**: 1522–1532.
- Hall, A.J., Kalantzi, O.I. and Thomas, G.O. 2003. Polybrominated diphenyl ethers (PBDEs) in grey seals during their first year of life—are they thyroid hormone endocrine disruptors? *Environmental Pollution* **126**: 29–37.
- Hickie, B.E., Ross, P.S., Macdonald, R.W. and Ford, J.K.B. 2007. Killer whales (*Orcinus orca*) face protracted health risks associated with lifetime exposure to PCBs. *Environmental Science and Technology* **41**: 6613–6619.
- Mos, L., Cameron, M., Jeffries, S.J., Koop, B.F. and Ross, P.S. 2010. Risk-based analysis of PCB toxicity in harbor seals. *Integrated Environmental Assessment and Management* **6**: 631–640.
- Rayne, S., Ikonou, M.G., Ross, P.S., Ellis, G.M. and Barrett-Lennard, L.G. 2004. PBDEs, PBBs, and PCNs in three communities of free-ranging killer whales (*Orcinus orca*) from the Northeastern Pacific Ocean. *Environmental Science and Technology* **38**: 4293–4299.
- Ross, P.S., Couillard, C.M., Ikonou, M.G., Johannessen, S.C., Lebeuf, M., Macdonald, R.W. and Tomy, G.T. 2009. Large and growing environmental reservoirs of Deca-BDE present an emerging health risk for fish and marine mammals. *Marine Pollution Bulletin* **58**: 7–10.
- Ross, P.S., Noël, M., Lambourn, D., Dangerfield, N., Calambokidis, J. and Jeffries, S. 2013. Declining concentrations of persistent PCBs, PBDEs, PCDEs, and PCNs in harbor seals (*Phoca vitulina*) from the Salish Sea. *Progress in Oceanography* **115**: 160–170.

1.2 Trends of brominated flame retardants in marine birds from the Pacific coast of Canada: 1990–2015

John E. Elliott¹, Aroha A. Miller², Kyle H. Elliott³ and Sandi Lee¹

¹Environment and Climate Change Canada, Ecotoxicology and Wildlife Health Directorate, Pacific Wildlife Research Centre, Delta, British Columbia, Canada

²Applied Animal Science, University of British Columbia, Vancouver, British Columbia, Canada

³Department of Natural Resource Sciences, McGill University, Ste Anne-de-Bellevue Quebec, Canada

Flame retardant chemicals are produced and marketed for the purpose of reducing the flammability and therefore fire risk of various materials, including textiles, plastics, and construction materials (Alaee *et al.*, 2003). Several million kg of brominated flame retardants (BFRs) such as polybrominated diphenyl ethers (PBDEs) have been manufactured and applied. Because major PBDE congeners are highly persistent and bioaccumulative, they are now widespread contaminants of abiotic and biotic ecosystem components across the globe (Darnerud, 2003), including the west coast of North America (Elliott *et al.*, 2005; Ross, 2006) and other parts of the North Pacific region (*e.g.*, Kajiwara *et al.*, 2004; Moon *et al.*, 2010; Greig *et al.*, 2011). As a result of that contamination, some BFRs, including penta and octa PBDE formulations, are now regulated as Persistent Organic Pollutants (POPs) under the Stockholm Convention, as is hexabromocyclododecane (HBCDD). In addition to being persistent and bioaccumulative, certain BFRs are toxic, with effects, including endocrine disrupting endpoints, reported in studies of PBDEs and HBCDD in birds (Darnerud, 2008; Crump *et al.*, 2010; Eng *et al.*, 2012; Winter *et al.*, 2013).

Seabirds are highly efficient and effective indicators of the contamination of marine ecosystems by persistent chemicals (Gilbertson *et al.*, 1987; Elliott and Elliott, 2013), including BFRs at various global locations (Sellström *et al.*, 2003; Braune *et al.*, 2007; Lavoie *et al.*, 2010; Crosse *et al.*, 2012). Previously, we reported rapidly increasing concentrations of PBDEs over the period 1990 to 2002 in eggs of an estuarine and shoreline indicator species, the great blue heron (*Ardea herodias*) and a nearshore indicator, the double-crested cormorant (*Phalacrocorax auritus*) from the Salish Sea region of the Pacific coast of Canada (Elliott *et al.*, 2005). We later updated those trends with data up to 2011 and including HBCDD (Miller *et al.* 2015). We also expanded the trend assessment to the broader Pacific coast using a continental shelf species, the rhinoceros auklet (*Cerorhinca monocerata*), and an offshore pelagic indicator, the Leach's storm petrel (*Oceanodroma leucorhoa*) (Miller *et al.*, 2014). Here, we summarize that temporal trend data for the North Pacific using the auklet and storm petrel as representatives of the Pacific coast, and include new results from 2015 (Fig. 1.2.1).

Detailed methods are described in Miller *et al.* (2014, 2015), including basic biology of study species, study locations, egg collection, and analytic methods. Briefly, 15 eggs were collected at 4-year intervals from randomly selected nests at the monitoring colonies and transported to the laboratory. Five pools were prepared consisting of 3 whole eggs each, and analyzed by gas chromatography with mass-specific detection (GC/MSD), using appropriate quality assurance procedures. Σ PBDEs consisted of 14 congeners (17, 28, 47, 49, 66, 85, 99, 100, 138, BDE 154/BB 153, 153, 183, 190, 209), the dominant congeners being BDEs 47, 99 and 100. Egg material remaining after initial analyses was stored in the Environment Canada Wildlife Specimen Bank. Results dated earlier than 2002 are from retrospective

surveys of archived samples. Concentrations are presented on a lipid weight basis to allow comparisons between species due to inter-specific variation in egg lipid content. Natural log-transformed Σ PBDE and HBCDD concentrations were split relative to usage restrictions, circa 2000, and tested with linear regression. Doubling times were calculated from the slope of the regression line and assume first-order (exponential) increases. For purposes of this PICES report, we selected data for two species and locations: storm petrels from Hippa Island (53°26'N 132°59'W), located on the northwest side of Haida Gwaii in the Gulf of Alaska, and rhinoceros auklets from Cleland Island (49°10'N; 126°5'W) off the south-central west coast of Vancouver Island.

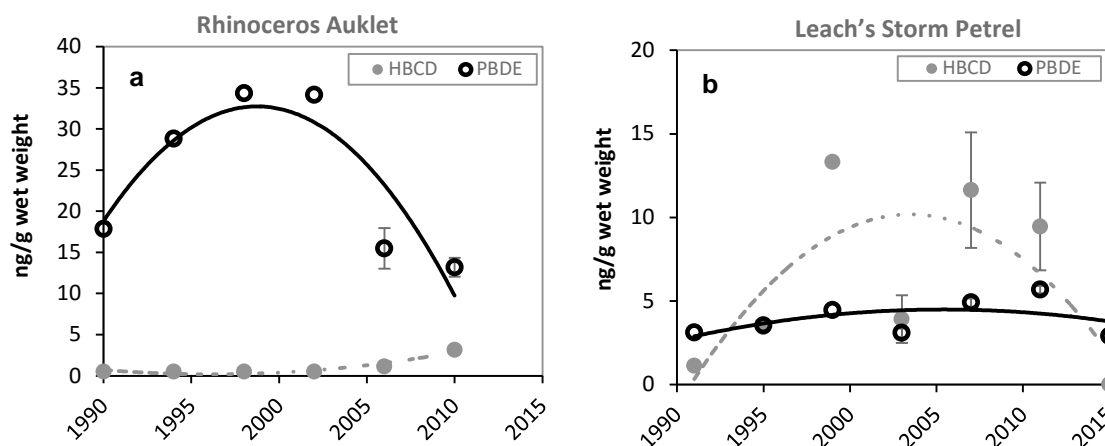


Fig. 1.2.1 HBCDD and Σ PBDE (ng/g lw) over time in a) rhinoceros auklet eggs from Cleland Island and b) Leach's storm petrel eggs from Hippa Island. Standard error of the mean is shown in years where multiple pooled samples were taken. Adapted from Miller *et al.* (2014).

Following implementation of voluntary restrictions by North American industry and subsequent phasing out of penta-BDE mixtures, Σ PBDE concentrations generally decreased in rhinoceros auklet eggs from the colony at Cleland Island (Fig. 1.2.1a). However, that trend was not apparent for storm petrel eggs collected at Hippa Island, although the doubling time did decline post-2000 (Fig. 1.2.1b). That particular trend in PBDEs in storm petrels from Hippa Island may be the result of that breeding population foraging farthest from North America and closest to Asia. Among the colonies monitored here, Hippa Island is the most distant from any influence of coastal industrial activity. As a result, the impact of regulatory changes on contaminant release could be delayed in birds breeding at remote sites, which could explain the lower exposure to PBDEs detected in Hippa Island storm petrels.

After 1995, HBCDD became the dominant congener among all PBDE and non-PBDE flame retardants measured in petrel eggs at Hippa Island. By 2010, HBCDD was the second dominant congener in auklet eggs from Cleland Island. However, HBCDD was below the limit of detection in auklet eggs at Cleland Island until 2002 and thus no regression could be conducted pre-2000 for HBCDD in auklet eggs.

The elevated HBCDD concentrations in storm petrel eggs from Hippa Island compared to auklet eggs from Cleland Island are likely an integration of factors such as diet and hence trophic level and particular food sources, seasonal movement, and even metabolism (Borgå *et al.*, 2005; Lavoie *et al.*, 2010). HBCDD was never produced in Canada but, as with other flame retardants, was imported into

the country as a treatment on a variety of commercial products (Environment Canada and Health Canada, 2011). This chemical was manufactured in Japan, the USA, the Netherlands, and China (UNEP, 2010). There are reports of elevated concentrations of HBCDD from sampling in coastal areas of Japan, China, and Europe (UNEP, 2010). Our data on stable isotopes in storm petrels, particularly $\delta^{13}\text{C}$, show they are the most pelagic of the seabird species monitored at breeding colonies on the Pacific coast of Canada, with some evidence that birds from Hippi Island forage more remotely than conspecifics monitored at other sites, which could explain the patterns of HBCDD contamination (Miller *et al.*, 2014). It is, however, still possible that the increasing exposure measured in seabird eggs from this monitoring program may be the result of contamination release from the local Canadian coastal environment, although long-range transport from production zones and deposition to the North American west coast environment is also possible (De Wit, 2002). At this point, these possible explanations are largely conjecture due to the lack of information on, for example, seasonal movements of storm petrels.

As we reported previously, there were no significant temporal trends in $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ in eggs of either species (Miller *et al.*, 2014). Thus, there was no indication of any temporal shifts in trophic level ($\delta^{15}\text{N}$) influencing ΣPBDE for individual species and sites, although spatial variation determined from carbon source ($\delta^{13}\text{C}$) had to be accounted for. Those stable isotope data indicate that diet was stable over time, and therefore, there was no detected effect of trophic level at this scale. Thus, the temporal trends in BFRs reported here are likely not caused by dietary factors and instead are almost certainly due to restrictions imposed on PBDE usage (Canada Gazette, 2006).

In conclusion, the initial voluntary curtailments of PBDE production and usage by industry in North America, and the following regulatory changes, appear to have achieved the desired outcome of reducing contaminant concentrations. Data presented here from seabird eggs collected from colonies on the Pacific coast of Canada over the period 1990 to 2015 are consistent with data showing declining levels of PBDEs in various matrices from elsewhere in North America and Europe (Crosse *et al.*, 2012; Ross *et al.*, 2013). However, the increasing trends in HBCDD in seabird eggs provide some basis for concern. The decision to consider that compound a POP under the Stockholm Convention should result in decreased usage and eventually declining contamination of globally affected ecosystems.

References

- Alaee, M., Arias, P., Sjödin, A. and Bergman, Å. 2003. An overview of commercially used brominated flame retardants, their applications, their use patterns in different countries/regions and possible modes of release. *Environment International* **29**: 683e689, [http://dx.doi.org/10.1016/S0160-4120\(03\)00121-1](http://dx.doi.org/10.1016/S0160-4120(03)00121-1).
- Borgå, K., Wolkers, H., Skaare, J.U., Hop, H., Muir, D.C. and Gabrielsen, G.W. 2005. Bioaccumulation of PCBs in Arctic seabirds: influence of dietary exposure and congener biotransformation. *Environmental Pollution* **134**: 397–409.
- Braune, B.M., Mallory, M.L., Grant Gilchrist, H., Letcher, R.J. and Drouillard, K.G. 2007. Levels and trends of organochlorines and brominated flame retardants in ivory gull eggs from the Canadian Arctic, 1976 to 2004. *The Science of the Total Environment* **378**: 403e417, <http://dx.doi.org/10.1016/j.scitotenv.2007.03.003>.
- Canada Gazette. 2006. Part I, Vol. 140, No. 50, Ottawa. <http://www.gazette.gc.ca/rp-pr/p1/2006/2006-12-16/pdf/g1-14050.pdf>.

- Crosse, J.D., Shore, R.F., Jones, K.C. and Pereira, M.G. 2012. Long term trends in PBDE concentrations in gannet (*Morus bassanus*) eggs from two UK colonies. *Environmental Pollution* **161**: 93e100, <http://dx.doi.org/10.1016/j.envpol.2011.10.003>.
- Crump, D., Egloff, C., Chiu, S., Letcher, R.J., Chu, S. and Kennedy, S.W. 2010. Pipping success, isomer-specific accumulation, and hepatic mRNA expression in chicken embryos exposed to HBCD. *Toxicological Sciences* **115**: 492e500, <http://dx.doi.org/10.1093/toxsci/kfq068>.
- Darnerud, P.O. 2003. Toxic effects of brominated flame retardants in man and in wildlife. *Environment International* **29**: 841e853, [http://dx.doi.org/10.1016/S0160-4120\(03\)00107-7](http://dx.doi.org/10.1016/S0160-4120(03)00107-7).
- Darnerud, P.O. 2008. Brominated flame retardants as possible endocrine disrupters. *International Journal of Andrology* **31**: 152e160, <http://dx.doi.org/10.1111/j.1365-2605.2008.00869.x>.
- De Wit, C.A. 2002. An overview of brominated flame retardants in the environment. *Chemosphere* **46**: 583–624.
- Elliott, J.E. and Elliott, K.H., 2013. Tracking marine pollution. *Science* **340**: 556e558, <http://dx.doi.org/10.1126/science.1235197>.
- Elliott, J.E., Wilson, L.K. and Wakeford, B. 2005. Polybrominated diphenyl ether trends in eggs of marine and freshwater birds from British Columbia, Canada, 1979-2002. *Environmental Science and Technology* **39**: 5584e5591, <http://dx.doi.org/10.1021/es050496q>.
- Eng, M.L., Elliott, J.E., MacDougall-Shackleton, S.A., Letcher, R.J. and Williams, T.D. 2012. Early exposure to 2,2',4,4',5-pentabromodiphenyl ether (BDE-99) affects mating behavior of zebra finches. *Toxicological Sciences* **127**: 269e276, <http://dx.doi.org/10.1093/toxsci/kfs076>.
- Environment Canada and Health Canada, 2011. <http://www.ec.gc.ca/ese-ees/7882C148-8AE4-4BA4-8555-668C49F91500/HBCD%20-%20FSAR%20-%20EN.pdf>.
- Gilbertson, M., Elliott, J.E. and Peakall, D.B. 1987. Seabirds as indicators of marine pollution, pp. 231–248 *in: The Uses of Birds edited by A.W. Diamond and F. Fillion*, ICBP Technical Publication No. 6, Cambridge.
- Greig, D.J., Ylitalo, G.M., Wheeler, E.A., Boyd, D., Gulland, F.M., Yanagida, G.K., Harvey, J.T. and Hall, A.J. 2011. Geography and stage of development affect persistent organic pollutants in stranded and wild-caught harbor seal pups from central California. *Science of the Total Environment* **409**: 3537–3547.
- Kajiwara, N., Ueno, D., Takahashi, A., Baba, N. and Tanabe, S. 2004. Polybrominated diphenyl ethers and organochlorines in archived northern fur seal samples from the Pacific coast of Japan, 1972-1998. *Environmental Science and Technology* **38**: 3804–3809.
- Lavoie, R.A., Champoux, L., Rail, J.-F. and Lean, D.R.S. 2010. Organochlorines, brominated flame retardants and mercury levels in six seabird species from the Gulf of St. Lawrence (Canada): relationships with feeding ecology, migration and molt. *Environmental Pollution* **158**: 2189e2199, <http://dx.doi.org/10.1016/j.envpol.2010.02.016>.
- Miller, A.A., Elliott, J.E., Elliott, K.H., Guigueno, M.F., Wilson, L.K., Lee, S. and Idrissi, A. 2014. Spatial and temporal trends in brominated flame retardants in seabirds from the Pacific coast of Canada. *Environmental Pollution* **195**: 48–55, Doi: 10.1016/j.envpol.2014.08.009.
- Miller, A.A., Elliott, J.E., Elliott, K.H., Guigueno, M.F., Wilson, L.K., Lee, S. and Idrissi, A. 2015. Brominated flame retardant trends in aquatic birds from the Salish Sea region of the west coast of North America, including a mini-review of recent trends in marine and estuarine birds. *Science of the Total Environment* **502**: 60–69.

- Moon, H.B., Kannan, K., Choi, M., Yu, J., Choi, H.G., An, Y.R., Choi, S.G., Park, J.Y. and Kim, Z.G. 2010. Chlorinated and brominated contaminants including PCBs and PBDEs in minke whales and common dolphins from Korean coastal waters. *Journal of Hazardous Materials* **179**: 735–741.
- Ross, P.S. 2006. Fireproof killer whales (*Orcinus orca*): flame-retardant chemicals and the conservation imperative in the charismatic icon of British Columbia, Canada. *Canadian Journal of Fisheries and Aquatic Sciences* **63**: 224–234.
- Ross, P.S., Noël, M., Lambourn, D., Dangerfield, N., Calambokidis, J. and Jeffries, S. 2013. Declining concentrations of persistent PCBs, PBDEs, PCDEs, and PCNs in harbor seals (*Phoca vitulina*) from the Salish Sea. *Progress in Oceanography* **115**: 160–170.
- Sellström, U., Bignert, A., Kierkegaard, A., Häggberg, L., de Wit, C., Olsson, M. and Jansson, B. 2003. Temporal trend studies on tetra- and pentabrominated diphenyl ethers and hexabromocyclododecane in guillemot egg from the Baltic Sea. *Environmental Science and Technology* **37**: 5496–5501.
- UNEP (United Nations Environment Programme). 2010. The nine new POPs: an introduction to the nine chemicals added to the Stockholm Convention by the Conference of the Parties at its fourth meeting. United Nations Environment Program Secretariat of the Stockholm Convention on Persistent Organic Pollutants.
- Winter, V., Williams, T.D. and Elliott, J.E. 2013. A three-generational study of in ovo exposure to PBDE-99 in the zebra finch. *Environmental Toxicology and Chemistry* **32**: 562e568, <http://dx.doi.org/10.1002/etc.2102>.

1.3 Recent trends in perfluoralkyl substances in marine birds from the Pacific coast of Canada: (1990–2011)

Aroha A. Miller¹, John E. Elliott^{2,1}, Kyle H. Elliott³ and Sandi Lee²

¹ Applied Animal Science, University of British Columbia, Vancouver, British Columbia, Canada

² Environment and Climate Change Canada, Ecotoxicology and Wildlife Health Directorate, Pacific Wildlife Research Centre, Delta, British Columbia, Canada

³ Department of Natural Resource Sciences, McGill University, Ste Anne-de-Bellevue Quebec, Canada

Perfluoralkyl substances (PFAS) such as sulfonates (PFSAs) and carboxylates (PFCAs) are used in various industrial and consumer products, *e.g.*, water and stain-proofing agents, grease and oil-resistant food packaging (Sinclair *et al.*, 2007), surfactants (Key *et al.*, 1998), and firefighting foams (Moody and Field, 2000). PFAS can bioaccumulate and biomagnify (Conder *et al.*, 2008; Ahrens, 2011) and are highly resistant to degradation (Key *et al.*, 1999). There is also concern about their toxicological effects (Houde *et al.*, 2006). Long-range dispersal of PFAS can also occur (Prevedouros *et al.*, 2006; Ahrens *et al.*, 2010). As such, these contaminants have been found in remote regions such as the Arctic (Houde *et al.*, 2006), across terrestrial, marine, and freshwater environments, and in virtually all matrices. They have been found across North America (Martin *et al.*, 2004a,b; Tittlemier *et al.*, 2007; Gebbink *et al.*, 2011; Braune and Letcher, 2013) including the west coast (Miller *et al.*, 2015). Consequently, various voluntary and regulatory restrictions on use, production, and manufacture of various PFAS have been implemented. In 2000, 3M, the major global producer of PFAS, phased out production of perfluorooctane sulfonic acid (PFOS), perfluorooctanoic acid (PFOA) and related products (USEPA, 2000). Further restrictions in the U.S., Canada, and Europe were implemented throughout the 2000s (USEPA, 2006; Environment Canada, 2010; EC Directive 2006/122/EC; Stockholm Convention). However, despite concerns in some countries, China began large-scale production of PFOS in 2003 (Ministry of Environmental Protection of the People's Republic of China, 2008; Han, 2009; Lim *et al.*, 2011; Wang *et al.*, 2016).

Marine birds are ideally situated for use as indicator species in examining environmental contaminants (Elliott and Elliott, 2013), and have been used to monitor PFAS from locations around the world (Holmström *et al.*, 2005; Verreault *et al.*, 2007; Flieger *et al.*, 2012; Braune and Letcher, 2013). We reported on temporal trends of PFAS (1990–2011) from eggs of two oceanic seabird species, a continental shelf species, the rhinoceros auklet (*Cercohinca monocerata*) and an offshore pelagic indicator, Leach's storm petrels (*Oceanodroma leucorhoa*); and two coastal bird species, a nearshore species, the double crested cormorant (*Phalacrocorax auritus*) and an estuarine/near shoreline species, great blue herons (*Ardea herodias*) from the Pacific coast of British Columbia, Canada (Miller *et al.*, 2015). Here, we summarize the temporal trend data for the North Pacific using auklets (1990–2010) from Cleland Island (49°10'N, 126°5'W), located off the south-central west coast of Vancouver Island, and Leach's storm petrel (1991–2011) from Hippa Island (53°26'N, 132°59'W), off the northwest side of Haida Gwaii in the Gulf of Alaska.

Detailed methods are described in Miller *et al.* (2015), including basic biology, study locations, collection, analytical methods, and statistical analyses. Briefly, 15 fresh eggs were collected from

randomly selected, individual nests every 4 years, and stored frozen until analyzed. Eggs were analyzed retrospectively as one pool of 15 eggs from 1990/1991 to 2006/2007 and subsequently, *i.e.*, in 2010/2011, as 3 pools of 5 eggs, and re-analyzed as 1 pool of 15 eggs, as per previous years. Compounds were analyzed by high-performance liquid chromatography/mass spectrometry (HPLC/MS) using appropriate quality assurance procedures.

Here, Σ PFSA includes perfluorobutane sulfonic acid (PFBS), perfluorohexane sulfonic acid (PFHxS), PFOS, and perfluorodecane sulfonic acid (PFDS). Σ PFCA includes perfluorohexanoic acid (PFHxA), perfluoroheptanoic acid (PFHpA), PFOA, perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDA), perfluoroundecanoic acid (PFUdA), perfluorododecanoic acid (PFDoA), perfluorotridecanoic acid (PFTrDA), and perfluorotetradecanoic acid (PFTeDA). Natural log-transformed Σ PFSA, Σ PFCA, and dominant compound concentrations were split based on the year 3M phased out PFOS, PFOA, and related compounds (*i.e.*, pre- and post-2000), and tested using linear regression. Doubling times were calculated using the slope of the regression line, and assume first order (exponential) increases.

The dominant PFSA compound in both species was PFOS (C₈), contributing >90% to Σ PFSA in auklets, and >97% for petrels. PFOS is typically the most commonly detected PFSA in wildlife (Houde *et al.*, 2006; Conder *et al.*, 2008; Braune and Letcher, 2013). PFDS (C₁₀) also showed quantifiable levels in all years for auklet eggs, although concentrations were far below that of PFOS. PFDS remained below the analytical limit of detection (LOD) in petrel eggs. Dominant PFCA compounds were PFUdA (C₁₁) and PFTrDA (C₁₃), both having long, odd-numbered carbon chains. Detection of long uneven-chain PFCAs at higher concentrations compared to even-chain PFCAs has been widely reported (Martin *et al.*, 2004b; Verreault *et al.*, 2007; Reiner *et al.*, 2011; Braune and Letcher, 2013). Other PFCA compounds found in quantifiable concentrations were PFNA, PFDA, PFDoA, and PFTeDA. Other compounds were either not found or were below LOD.

Temporal trends of Σ PFSA, and thus PFOS, have been inconsistent for both species (Fig. 1.3.1a). PFOS doubling time in auklet eggs post-2000 was 630.1 years, considerably slower than that seen pre-2000 (10.6 years). PFOS concentration increased over the entire period in petrel eggs, with a post-2000 doubling time of 14.4 years, considerably faster than pre-2000 (43 years), indicating that concentrations continue to increase in the oceanic environment with phase-outs having little effect thus far on contamination of petrels at this location. As petrels spend the non-breeding season in offshore pelagic environments, potentially closer to Asia, and stable isotope analysis, particularly $\delta^{13}\text{C}$, reveals them to be the most pelagic of the seabird species monitored in the Pacific coastal region of Canada (Miller *et al.*, 2015), their exposure to PFCs may be influenced less by regulatory actions in North America, and more by ongoing production in Asia.

Significant ($p < 0.01$) increasing Σ PFCA temporal trends were seen for both species. PFUdA showed significant ($p < 0.01$) increasing temporal trends in both species (Fig. 1.3.1b). PFTrDA showed a significant ($p < 0.01$) increasing trend in petrels at Hippa Island, and a general increasing trend over time in auklets. Despite restrictions implemented in North America for these compounds (USEPA, 2006; Environment Canada, 2010; Buck *et al.*, 2011), there has not been enough time to observe a related effect on Σ PFCA concentrations in petrels and auklets. As reported previously, no significant temporal trends were observed for stable isotope analyses ($\delta^{15}\text{N}$, $\delta^{13}\text{C}$) in auklets or petrels, with no trophic level transcendence over the examined period (Miller *et al.*, 2015). Thus, these data show auklet and petrel diet was stable over time and hence, trophic level changes have not influenced temporal PFAS trends.

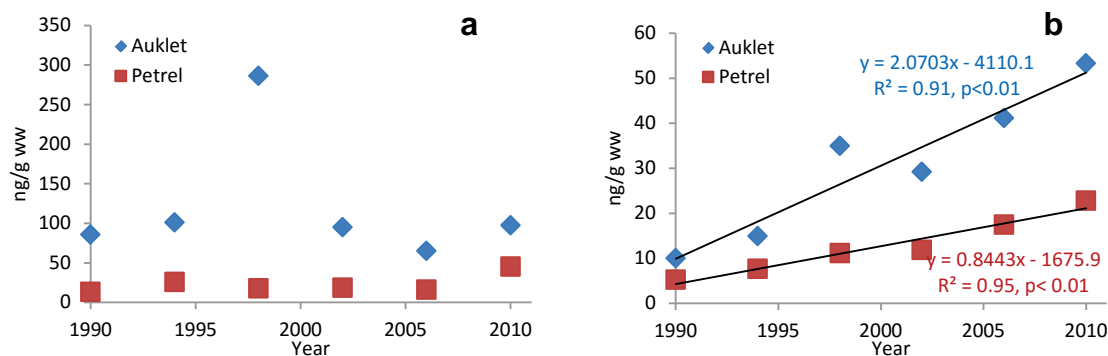


Fig. 1.3.1 Trends of perfluoralkyl substances in seabirds from British Columbia, Canada: a) PFOS concentrations (ng/g ww) and b) Σ PFCA (ng/g ww) in auklets and petrel eggs from Cleland Island and Hippa Island, respectively.

To conclude, restrictions on PFAS compound use and production in North America have not yet achieved the desired effect in terms of reducing the levels measured in wildlife. PFSA data presented here from seabird eggs collected on the Pacific coast of Canada between 1990 and 2011 display some consistency with trends in Canadian Arctic wildlife (Braune and Letcher, 2013) and European wildlife, where rapid increases were seen during the 1970s and 1980s before levelling off (Holmström *et al.*, 2005, 2010). PFCA data presented here are also consistent with trends in other wildlife, *e.g.*, herring gulls (*Larus argentatus*) from Norway (Verreault *et al.*, 2007) and coastal German regions (Fliedner *et al.*, 2012), as well as selected seabirds from the Canadian Arctic (Braune and Letcher, 2013). Increasing trends are an ongoing cause for concern. We plan to continue the long-term monitoring program described here along with associated research.

References

- Ahrens L. 2011. Polyfluoroalkyl compounds in the aquatic environment: A review of their occurrence and fate. *Journal of Environmental Monitoring* **13**: 20–31.
- Ahrens, L., Xie, Z. and Ebinghaus, R. 2010. Distribution of perfluoroalkyl compounds in seawater from Northern Europe, Atlantic Ocean, and Southern Ocean. *Chemosphere* **78**: 1011–1016.
- Braune, B.M. and Letcher, R.J. 2013. Perfluorinated sulfonate and carboxylate compounds in eggs of seabirds breeding in the Canadian Arctic: Temporal trends (1975–2011) and interspecies comparison. *Environmental Science and Technology* **47**: 616–624.
- Buck, R.C., Franklin, J., Berger, U., Conder, J.M., Cousins, I.T., de Voogt, P., Jensen, A.A., Kannan, K., Mabury, S.A. and van Leeuwen, S.P.J. 2011. Perfluoroalkyl and polyfluoroalkyl substances in the environment: Terminology, classification, and origins. *Integrated Environmental Assessment and Management* **7**: 513–541.
- Conder, J.M., Hoke, R.A., De Wolf, W., Russell, M.H. and Buck, R.C. 2008. Are PFCAs bioaccumulative? A critical review and comparison with regulatory criteria and persistent lipophilic compounds. *Environmental Science and Technology* **42**: 995–1003.
- Elliott, J.E. and Elliott, K.H. 2013. Tracking marine pollution. *Science* **340**: 556–558.

- Environment Canada. 2010. Environmental Performance Agreement: Perfluorinated carboxylic acids (PFCAs) and their precursors (2010-2015). <https://www.canada.ca/en/environment-climate-change/services/environmental-performance-agreements/results.html#X-2013092511492112> (accessed September 18, 2014).
- European Union. 2006. Directive 2006/122/EC of the European Parliament and of the Council of 12 December 2006. *Official Journal of the European Union* L372/32-34.
- Fliedner, A., Rüdell, H., Jüriling, H., Müller, J., Neugebauer, F. and Schröter- Kermani, C. 2012. Levels and trends of industrial chemicals (PCBs, PFCs, PBDEs) in archived herring gull eggs from German coastal regions. *Environmental Sciences Europe* **24**: 7, <https://doi.org/10.1186/2190-4715-24-7>.
- Gebbink, W.A., Letcher, R.J., Hebert, C.E. and Chip Weseloh, D.V. 2011. Twenty years of temporal change in perfluoroalkyl sulfonate and carboxylate contaminants in herring gull eggs from the Laurentian Great Lakes. *Journal of Environmental Monitoring* **13**: 3365–3372.
- Han, W. 2009. PFOS related actions in China. Proceedings, International Workshop on Managing Perfluorinated Chemicals and Transitioning to Safer Alternatives, Geneva, Switzerland, February 12–13, 2009.
- Holmström, K.E., Järnberg, U. and Bignert, A. 2005. Temporal trends of PFOS and PFOA in guillemot eggs from the Baltic Sea, 1968–2003. *Environmental Science and Technology* **39**: 80–84.
- Holmström, K.E., Johansson, A.-K., Bignert, A., Lindberg, P. and Berger, U. 2010. Temporal trends of perfluorinated surfactants in Swedish peregrine falcon eggs (*Falco peregrinus*), 1974–2007. *Environmental Science and Technology* **44**: 4083–4088.
- Houde, M., Martin, J., Letcher, R.J., Solomon, K.R. and Muir, D. 2006. Biological monitoring of polyfluoroalkyl substances: A review. *Environmental Science and Technology* **40**: 3463–3473.
- Key, B., Howell, R. and Criddle, C. 1998. Defluorination of organofluorine sulfur compounds by *Pseudomonas* sp. strain D2. *Environmental Science and Technology* **32**: 2283–2287.
- Key, B., Howell, R. and Criddle, C. 1999. Fluorinated organics in the biosphere. *Environmental Science and Technology* **31**: 2445–2454.
- Lim, T.C., Wang, B., Huang, J., Deng, S. and Yu, G. 2011. Emission inventory for PFOS in China: Review of past methodologies and suggestions. *The Scientific World Journal* **11**: 1963–1980.
- Martin, J.W., Whittle, D.M., Muir, D.C.G. and Mabury, S.A. 2004a. Perfluoroalkyl contaminants in a food web from Lake Ontario. *Environmental Science and Technology* **38**: 5379–5385.
- Martin, J.W., Smithwick, M.M., Braune, B.M., Hoekstra, P.F., Muir, D.C.G. and Mabury, S.A. 2004b. Identification of long-chain perfluorinated acids in biota from the Canadian Arctic. *Environmental Science and Technology* **38**: 373–380.
- Miller, A., Elliott, J.E., Elliott, K.H., Lee, S. and Cyr, F. 2015. Temporal trends of perfluoroalkyl substances (PFAS) in eggs of coastal and offshore birds: increasing PFAS levels associated with offshore bird species breeding on the Pacific coast of Canada and wintering near Asia. *Environmental Toxicology and Chemistry* **34**: 1799–1808, DOI: 10.1002/etc.2992.
- Ministry of Environmental Protection of the People’s Republic of China. 2008. Additional information on production and use of PFOS. Memorandum to Mr. Donald Cooper, Secretariat of the Stockholm Convention, Châtelaine, Switzerland. Beijing, China.
- Moody, C. and Field, J. 2000. Perfluorinated surfactants and the environmental implications of their use in fire-fighting foams. *Environmental Science and Technology* **34**: 3864–3870.

- Prevedouros, K., Cousins, I., Buck, R.C. and Korzeniowski, S.H. 2006. Sources, fate and transport of perfluorocarboxylates. *Environmental Science and Technology* **40**: 32–44.
- Reiner, J.L., O’Connell, S.G., Moors, A.J., Kucklick, J.R., Becker, P.R. and Keller, J.M. 2011. Spatial and temporal trends of perfluorinated compounds in beluga whales (*Delphinapterus leucas*) from Alaska. *Environmental Science and Technology* **45**: 8129–8136.
- Sinclair, E., Kim, S.K., Akinleye, H.B. and Kannan K. 2007. Quantitation of gasphase perfluoroalkyl surfactants and fluorotelomer alcohols released from nonstick cookware and microwave popcorn bags. *Environmental Science and Technology* **41**: 1180–1185.
- Stockholm Convention on Persistent Organic Pollutants (POPs). 2008. Perfluorooctane sulfonic acid, its salts and perfluorooctane sulfonyl fluoride.
<http://chm.pops.int/TheConvention/ThePOPs/TheNewPOPs/tabid/2511/Default.aspx>.
- Tittlemier, S.A., Pepper, K., Seymour, C., Moisey J., Bronson, R., Cao, X.-L. and Dabeka, R.W. 2007. Dietary exposure of Canadians to perfluorinated carboxylates and perfluorooctane sulfonate via consumption of meat, fish, fast foods, and food items prepared in their packaging. *Journal of Agricultural and Food Chemistry* **55**: 3203–3210.
- USEPA (U.S. Environmental Protection Agency). 2000. EPA and 3M announce phase out of PFOS.
https://archive.epa.gov/epapages/newsroom_archive/newsreleases/33aa946e6cb11f35852568e1005246b4.html (accessed August 14, 2014).
- USEPA. 2006. 2010/2015 PFOA Stewardship Program. <http://www.epa.gov/opptintr/pfoa/pubs/stewardship/> (accessed September 18, 2014).
- Verreault, J., Berger, U. and Gabrielsen, G.W. 2007. Trends of perfluorinated alkyl substances in herring gull eggs from two coastal colonies in northern Norway: 1983–2003. *Environmental Science and Technology* **41**: 6671–6677.
- Wang, T., Vestergren, R., Herzke, D., Yu, J. and Cousins, I.T. 2016. Levels, isomer profiles, and estimated riverine mass discharges of perfluoroalkyl acids and fluorinated alternatives at the mouths of Chinese rivers. *Environmental Science and Technology* **50**: 11,584–11,592, DOI: 10.1021/acs.est.6b03752.

1.4 Temporal trends for mercury in aquatic bird eggs from the Pacific coast of Canada (1968–2015)

Kyle H. Elliott¹, John E. Elliott^{2,3}, Aroha A. Miller³ and Sandi Lee²

¹Department of Natural Resource Sciences, McGill University, Ste Anne-de-Bellevue Quebec, Canada

²Environment and Climate Change Canada, Ecotoxicology and Wildlife Health Directorate, Pacific Wildlife Research Centre, Delta, British Columbia, Canada

³Applied Animal Science, University of British Columbia, Vancouver, British Columbia, Canada

Mercury is a naturally occurring element found in various chemical and physical forms (Suzuki *et al.*, 1991) that resist degradation (Vo *et al.*, 2011; Mason *et al.*, 2012; Driscoll *et al.*, 2013). Natural sources of mercury include volcanoes, forest fires, fossil fuels, petroleum, and cinnabar ore (Calvert, 2007). However, numerous anthropogenic activities also produce mercury *e.g.*, fossil fuel combustion, mining, smelting and solid waste combustion, fertilisers, and industrial wastewater disposal amongst others. Various consumer products contain mercury, *e.g.*, thermometers, some electrical switches, amalgam for dental restoration, and batteries. Mercury bioaccumulates (Clarkson, 1992) and methylmercury (MeHg⁺), the organic form of mercury, which is most toxic to animals and of most concern to human health, biomagnifies in foodwebs (Kidd *et al.*, 1995; Atwell *et al.*, 1998; Elliott 2005; Clayden *et al.*, 2013). Sulfate-reducing bacteria have been shown to be a controlling factor of mercury methylation in estuarine, anoxic and acidic sediments (Choi and Bartha, 1994). High mercury exposure can affect brain development (Doetzel, 2007), as methylmercury disturbs cell division and development. As a consequence, various regulations have been instigated around the world to decrease mercury use and reduce mercury levels in the environment. In 2013, Canada signed the Minamata Convention on Mercury, a global treaty ratified extensively by the majority of the international community (Minamata Mercury Convention; UNEP 2016), with the aim of “protecting human health and the environment from anthropogenic emissions and releases of mercury and mercury compounds” (Environment Canada, 2016).

To assess the effectiveness of the Minamata Convention, temporal and spatial trends must be monitored. Dietary variation, as assessed by $\delta^{15}\text{N}$ (trophic position) and $\delta^{13}\text{C}$, can confound such monitoring due to the biomagnifying properties of mercury. However, $\delta^{34}\text{S}$ may be a particularly useful dietary tracer for mercury as it varies little from source to consumer and is, therefore, unaffected by trophic level (Elliott and Elliott, 2016). As such, aquatic bird eggs are preferred for this biomonitoring because almost all mercury deposited in the eggs is in the form of methylmercury (Scheuhammer *et al.*, 2001; Ackerman *et al.*, 2013). In the past few decades, mercury levels in seabird eggs in the Canadian Arctic have increased after accounting for trophic level (Braune *et al.*, 2014), while mercury levels in seabirds from the Atlantic have varied interannually but with no clear trend (Burgess *et al.*, 2013). Leach’s storm petrel (*Oceanodroma leucorhoa*) eggs from both the Pacific and Atlantic oceans typically have the highest mercury levels of any seabird species used in biomonitoring in Canada (Elliott and Scheuhammer, 1997; Goodale *et al.*, 2008; Bond and Diamond, 2009; Braune *et al.*, 2014) despite feeding at the surface on invertebrates and small fish.

Recently, mercury trends in ancient murrelets (*Synthliboramphus antiquus*), rhinoceros auklets (*Cerorhinca monocerata*), Leach’s storm petrels (*Oceanodroma leucorhoa*), double-crested cormorants

(*Phalacrocorax auritus*), pelagic cormorants (*Phalacrocorax pelagicus*), and great blue herons (*Ardea herodias*) from a range of sites on the Pacific coast of Canada were published (Elliott and Elliott, 2016). Here, we summarize temporal trend data for mercury in the Pacific northwest of Canada, using 1) an offshore pelagic indicator, the Leach's storm petrel (1968–2015), collected from Cleland Island (49°10'N; 126°5'W) located off the west coast of Vancouver Island, and Hippa Island (53°26'N, 132°59'W), located on the northwest side of Haida Gwaii in the Gulf of Alaska; 2) two continental shelf indicators, the ancient murrelet (1968–2009) from Langara Island (54°12'N; 133°1'W) located on the north coast of Haida Gwaii, and the rhinoceros auklet (1970–2014) from Lucy Island (49°10'N; 126°5'W) located on the northwest mainland coast of British Columbia.

Mercury and stable isotope trends in Leach's storm petrel, ancient murrelet, and rhinoceros auklet

Detailed methods are described in Elliott and Elliott (2016). Briefly, for storm petrels and auklets, 15 fresh eggs were collected from each species every 4 years from randomly selected nests within colonies. For ancient murrelets, approximately 10 eggs were collected more opportunistically at Langara Island. In all cases, eggs were kept at ambient temperature until refrigerated for short-term storage and shipping. Once at the laboratory, eggs were opened and contents stored frozen until analyzed, which was done retrospectively. Samples were processed for mercury at Environment Canada laboratory facilities using an AMA-254 mercury analyzer and following standardized treatment protocols including quality assurance procedures (Braune *et al.*, 2014). Stable isotopes of $\delta^{15}\text{N}$, $\delta^{13}\text{C}$, and $\delta^{34}\text{S}$ were analyzed from the same egg samples at the Stable Isotope Facility at the University of California, Davis (<http://stableisotopefacility.ucdavis.edu>; described in Miller *et al.* (2014) and Blight *et al.* (2015)). $\delta^{13}\text{C}$ values were lipid-normalized as variation in lipid content can obscure variation in $\delta^{13}\text{C}$ (Drenick *et al.*, 2015). Statistical analyses were conducted in R (version 3.2.1) using general linear models with mercury (dry weight basis, log-transformed) as the dependent variable, and species, moisture, site, year, and isotope values as the independent variables were conducted (further details in Elliott and Elliott, 2016).

Temporal trends

Mercury levels declined with year in murrelets ($t_{56} = -4.06$, $P = 0.0002$), and displayed no trends in storm petrels ($t_{112} = -1.01$, $P = 0.31$; colony covariate: $t_{112} = -2.03$, $P = 0.048$) or auklets ($t_{57} = 1.02$, $P = 0.31$; Fig. 1.4.1). In the almost 50-year data set for Pacific seabird eggs presented here, Hg concentrations displayed some high inter-annual variation but with no clear temporal trends, except for cases where diet ($\delta^{34}\text{S}$) changed in tandem with Hg concentrations (Elliott and Elliott, 2016). Trends in mercury concentrations in marine mammals and fish from the Pacific also remained effectively stable over the recent 50-year period (Kraepiel *et al.*, 2003), also with the exception of cases where a dietary shift may have been a contributing variable (Drenick *et al.*, 2015). In eggs of seabirds from Atlantic Canada, no clear trends could be discerned from the high individual variability and after factoring dietary variation (Burgess *et al.*, 2013). In the present data set we inferred dietary variability from simultaneous measurement of sulfur rather than nitrogen isotopes (Elliott and Elliott, 2016); thus, we encourage researchers to consider more complex interpretation of food web structure as part of monitoring programs (*e.g.*, Hebert and Weseloh, 2006; Hebert *et al.*, 2006). We detected significant differences in mercury over time in both resident (cormorant; Elliott and Elliott, 2016) and migratory (murrelet) species. Because time trends of mercury changed in tandem with trends in isotopic signatures

(which indicate the bird's diet over the previous two weeks, when all species were resident around their breeding sites), we argue that the measured mercury time trends are representative of the situation at the local breeding grounds, as opposed to indicating mercury trends retained from exposure at wintering locations.

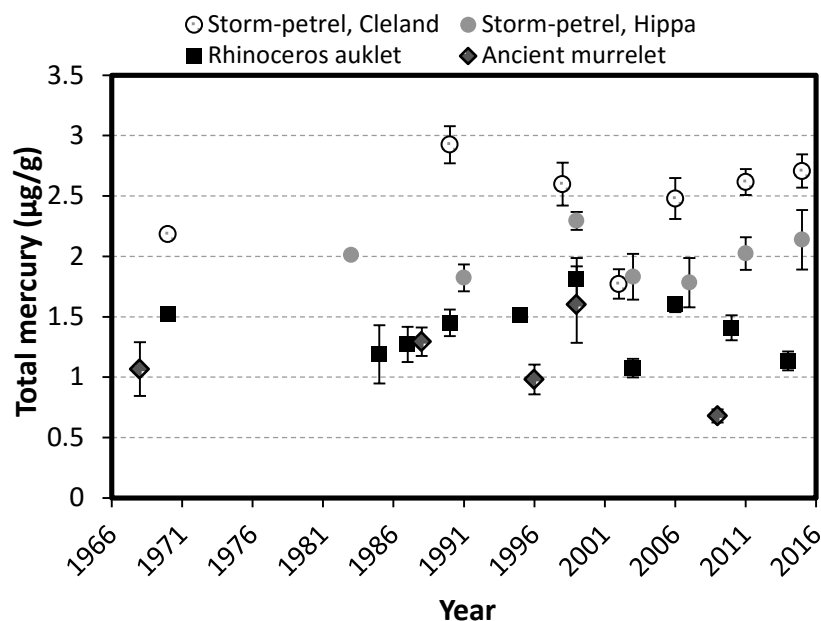


Fig. 1.4.1 Mercury (dry weight) trends between 1968 and 2015 in the eggs of three seabird species (including two storm petrel populations) from the Pacific coast of Canada.

In the oceans, as with other environments, Hg concentrations increased significantly during and since the Industrial Revolution. Mercury concentrations in feathers and eggshells increased over that period of time as acceleration of mercury contamination of global food chains became detectable in top marine predators (Vo *et al.*, 2011; Xu *et al.*, 2011). Although mercury deposition rates in the Pacific Ocean have increased 3- to 5-fold since the Industrial Revolution (Laurier *et al.*, 2004), levels in seabirds have increased by less than 2-fold over that period (Vo *et al.*, 2011), implying that there is a dampening effect of the food web on levels in top predators. More recently, while global mercury primary production and levels in the Atlantic have declined, mercury levels in Pacific surface water have increased because of coal burning in Asia (Hylander and Meili, 2004; Mason *et al.*, 2012; Driscoll *et al.*, 2013; Lamborg *et al.*, 2014). We have found no evidence of an increasing mercury trend in our Pacific seabird eggs concurrent with the surface water data. That may be because in top predators mercury accumulation is limited in large part by availability of sulfate rather than mercury (Elliott and Elliott, 2016). We plan to continue our monitoring and related research of mercury and other contaminants in seabirds from the Pacific coast of Canada and to report further on our results.

References

Ackerman, J.T., Herzog, M.P. and Schwarzbach, S.E. 2013. Methylmercury is the predominant form of mercury in bird eggs: a synthesis. *Environmental Science and Technology* **47**: 2052–2060.

- Atwell, L., Hobson, K.A. and Welch, H.E. 1998. Biomagnification and bioaccumulation of mercury in an arctic marine food web: insights from stable nitrogen isotope analysis. *Canadian Journal of Fisheries and Aquatic Sciences* **55**: 1114–1121.
- Blight, L.K., Hobson, K.A., Kyser, T.K. and Arcese, P. 2015. Changing gull diet in a changing world: A 150-year stable isotope ($\delta^{13}\text{C}$, $\delta^{15}\text{N}$) record from feathers collected in the Pacific Northwest of North America. *Global Change Biology* **21**: 1497–1507.
- Bond, A.L. and Diamond, A.W. 2009. Mercury concentrations in seabird tissues from Machias Seal Island, New Brunswick, Canada. *Science of the Total Environment* **407**: 4340–4347.
- Braune, B.M., Gaston, A.J., Hobson, K.A., Gilchrist, H.G. and Mallory, M.L. 2014. Changes in food web structure alter trends of mercury uptake at two seabird colonies in the Canadian Arctic. *Environmental Science and Technology* **48**: 13,246–13,252.
- Burgess, N.M., Bond, A.L., Hebert, C.E., Neugebauer, E. and Champoux, L. 2013. Mercury trends in herring gull (*Larus argentatus*) eggs from Atlantic Canada, 1972–2008: temporal change or dietary shift? *Environmental Pollution* **172**: 216–222.
- Calvert, J.B. 2007. <http://mysite.du.edu/~jcalvert/phys/mercury.htm#Prod>, created 2002, last revised 2007 (accessed February 11, 2017).
- Choi, S.C. and Bartha, R. 1994. Environmental factors affecting mercury methylation in estuarine sediments. *Bulletin of Environmental Contamination and Toxicology* **53**: 805–812.
- Clarkson, T.W. 1992. Mercury: Major issues in environmental health. *Environmental Health Perspectives* **100**: 31–38.
- Clayden, M.G., Kidd, K.A., Wyn, B., Kirk, J.L., Muir, D.C. and O’Driscoll, N.J. 2013. Mercury biomagnifications through food webs is affected by physical and chemical characteristics of lakes. *Environmental Science and Technology* **47**: 12,047–12,053.
- Doetzel, LM. 2007. An investigation of the factors affecting mercury accumulation in lake trout, *Salvelinus namaycush*, in Northern Canada. Unpublished PhD thesis, University of Saskatchewan. 141 pp.
- Drenick, P.E., Lamborg, C.H. and Horgan, M.J. 2015. Increase in mercury in Pacific yellowfin tuna. *Environmental Toxicology and Chemistry* **34**: 931–934.
- Driscoll, C.T., Mason, R.P., Chan, H.M., Jacob, D.J. and Pirrone, N. 2013. Mercury as a global pollutant: sources, pathways, and effects. *Environmental Science and Technology* **47**: 4967–4983.
- Elliott, J.E. 2005. Trace metals, stable isotope ratios, and trophic relations in seabirds from the North Pacific Ocean. *Environmental Toxicology and Chemistry* **24**: 3099–3105.
- Elliott, J.E. and Scheuhammer, A.M. 1997. Heavy metal and metallothionein concentrations in seabirds from the Pacific Coast of Canada. *Marine Pollution Bulletin* **34**: 794–801.
- Elliott, K.H. and Elliott, J.E. 2016. Origin of sulfur in diet drives spatial and temporal mercury trends in seabird eggs from Pacific Canada 1968–2015. *Environmental Science and Technology* **50**: 13,380–13,386, DOI: 10.1021/acs.est.6b05458.
- Environment Canada 2016. Minamata Convention on Mercury. <https://www.ec.gc.ca/international/default.asp?lang=En&n=5C03713D-1> (last updated June 2016; accessed February 10, 2017).
- Goodale, M.W., Evers, D.C., Mierzykowski, S.E., Bond, A.L., Burgess, N.M., Otorowski, C.I., Welch, L.J., Hall, C.S., Ellis, J.C., Allen, R.B. and Diamond, A.W. 2008. Marine foraging birds as bioindicators of mercury in the Gulf of Maine. *EcoHealth* **5**: 409–425.

- Hebert, C.E. and Weseloh, D.C. 2006. Adjusting for temporal change in trophic position results in reduced rates of contaminant decline. *Environmental Science and Technology* **40**: 5624–5628.
- Hebert, C.E., Arts, M.T. and Weseloh, D.C. 2006. Ecological tracers can quantify food web structure and change. *Environmental Science and Technology* **40**: 5618–5623.
- Hylander, L.D. and Meili, M. 2003. 500 years of mercury production: global annual inventory by region until 2000 and associated emissions. *Science of the Total Environment* **304**: 13–27.
- Kidd, K.A., Hesslein, R.H., Fudge, R.J. and Hallard, K.A. 1995. The influence of trophic level as measured by $\delta^{15}\text{N}$ on mercury concentrations in freshwater organisms, pp. 1011–1015 *in*: Mercury as a Global Pollutant *edited by* D.B. Porcella, J.W. Huckabee and B. Wheatley, Springer, The Netherlands.
- Kraepiel, A.M., Keller, K., Chin, H.B., Malcolm, E.G. and Morel, F.M. 2003. Sources and variations of mercury in tuna. *Environmental Science and Technology* **37**: 5551–5558.
- Lamborg, C.H., Hammerschmidt, C.R., Bowman, K.L., Swarr, G.J., Munson, K.M., Ohnemus, D.C., Lam, P.J., Heimbürger, L.E., Rijkenberg, M.J. and Saito, M.A. 2014. A global inventory of anthropogenic mercury based on water column measurements. *Nature* **512**: 65–68.
- Laurier, F.J.G., Mason, R.P., Gill, G.A. and Whalin, L. 2004. Mercury distributions in the North Pacific Ocean: 20 years of observations. *Marine Chemistry* **90**: 3–19.
- Mason, R.P., Choi, A.L., Fitzgerald, W.F., Hammerschmidt, C.R., Lamborg, C.H., Sorensen, A.L. and Sunderland, E.M. 2012. Mercury biogeochemical cycling in the ocean and policy implications. *Environmental Research* **119**: 101–117.
- Miller, A., Elliott, J.E., Elliott, K.H., Guigueno, M.F., Wilson, L.K., Lee, S. and Idrissi, A. 2014. Spatial and temporal trends in brominated flame retardants in seabirds from the Pacific coast of Canada. *Environmental Pollution* **195**: 48–55.
- Minamata Mercury Convention. <http://www.mercuryconvention.org/Countries/tabid/3428/Default.aspx> (accessed February 11, 2017).
- Scheuhammer, A.M., Perrault, J.A. and Bond, D.E. 2001. Mercury, methylmercury, and selenium concentrations in eggs of common loons (*Gavia immer*) from Canada. *Environmental Monitoring and Assessment* **72**: 79–94.
- Suzuki T., Imura N. and Clarkson T.W. 1991. Overview, 32 pp. *in*: Advances in Mercury Toxicology *edited by* T. Suzuki, N. Imura and T.W. Clarkson, Proceedings of a Rochester International Conference in Environmental Toxicology, Tokyo, Japan, August 1–3, 1990. Plenum Press, New York, 489 pp.
- UNEP (United Nations Environment Programme). 2016. Minamata Convention on Mercury. <http://www.mercuryconvention.org/Convention>.
- Vo, A.T.E., Bank, M.S., Shine, J.P. and Edwards, S.V. 2011. Temporal increase in organic mercury in an endangered pelagic seabird assessed by century-old museum specimens. *Proceedings of the National Academy of Sciences of the United States of America* **108**: 7466–7471.
- Xu, L.Q., Liu, X.D., Sun, L.G., Chen, Q.Q., Yan, H., Liu, Y., Luo, Y.H. and Huang, J.A. 2011. 700-year record of mercury in avian eggshells of Guangjin Island, South China Sea. *Environmental Pollution* **159**: 889–896.

1.5 Historical mercury trends in sediments from coastal British Columbia, Canada

Juan José Alava^{1,2} and Marie Noël¹

¹Institute for the Oceans and Fisheries, University of British Columbia, Vancouver, British Columbia, Canada

²Coastal and Ocean Research Institute, Vancouver Aquarium Marine Science Centre, Vancouver, BC, Canada

Mercury (Hg) continues to represent a health risk to marine wildlife, especially as atmospheric emissions of this toxic element from human activities in the Pacific Rim and worldwide continue (Mason *et al.*, 2012; UNEP, 2013; Lamborg *et al.*, 2014; Peterson *et al.*, 2015; Noël *et al.*, 2016; Alava *et al.*, 2018). In the Northeast Pacific, local sources of mercury in coastal British Columbia have included chlor-alkali plants, pulp mills, mine tailings, shipyards, and municipal outfalls. In this report, we review the status of Hg contamination in Strait of Georgia as a receiving environment in close proximity to urban centers and industries.

Mercury in sediment cores

Sediment box cores were collected in the Strait of Georgia, Vancouver Harbour, and Howe Sound between 1990 and 2001 (Johannessen *et al.*, 2005). Mercury determined in these sediment cores reveal a history of contamination that began in the 1860s and include episodic inputs during World War II and in the late 1960s (Fig. 1.5.1). A peak in mercury contamination in the period 1965 to 1970 can be attributed to discharges from a chlor-alkali plant in Howe Sound (Fig. 1.5.1). Surface sediment Hg concentrations at sites throughout the Strait of Georgia ranged from 60 to 420 ng/g dry weight (Fig. 1.5.2). There were no major detectable trends in sediment Hg concentrations in recent times, although Hg levels may be declining in specific locations (Johannessen *et al.*, 2005). The Canadian Interim Sediment Quality Guideline for mercury (ISQG: 130 ng/g dw) was exceeded at 23 points in time as estimated in the sediment cores (Fig. 1.5.1). Similarly, Hg in surficial sediments varied in recent times, but exceeded the ISQG at six points in time (Fig. 1.5.2). While more recent data in sediments is lacking, a recent study on mercury in the hair of harbour seals (*Phoca vitulina*) from British Columbia and Washington State (USA) found that total Hg (THg) concentrations were in the order of 5.3 ± 0.3 $\mu\text{g/g}$ dw in pups, 4.5 ± 0.5 $\mu\text{g/g}$ in juveniles and 8.3 ± 0.8 $\mu\text{g/g}$ in adults, with adult individuals exhibiting the highest levels (Noël *et al.*, 2016). This study concluded that both anthropogenic loadings and natural processes contributed to mercury in harbour seals. These concentrations are among the highest levels detected in marine mammals from the North Pacific, raising questions about the possible contribution of anthropogenic emissions to these mercury levels in seals in the Northeast Pacific Ocean. In addition, THg concentrations of 5.8–7.0 $\mu\text{g/g}$ in muscle of deep-diving and offshore-foraging northern elephant seals (*Mirounga angustirostris*) may indicate contamination of the mesopelagic environment of the North Pacific Ocean (Peterson *et al.*, 2015).

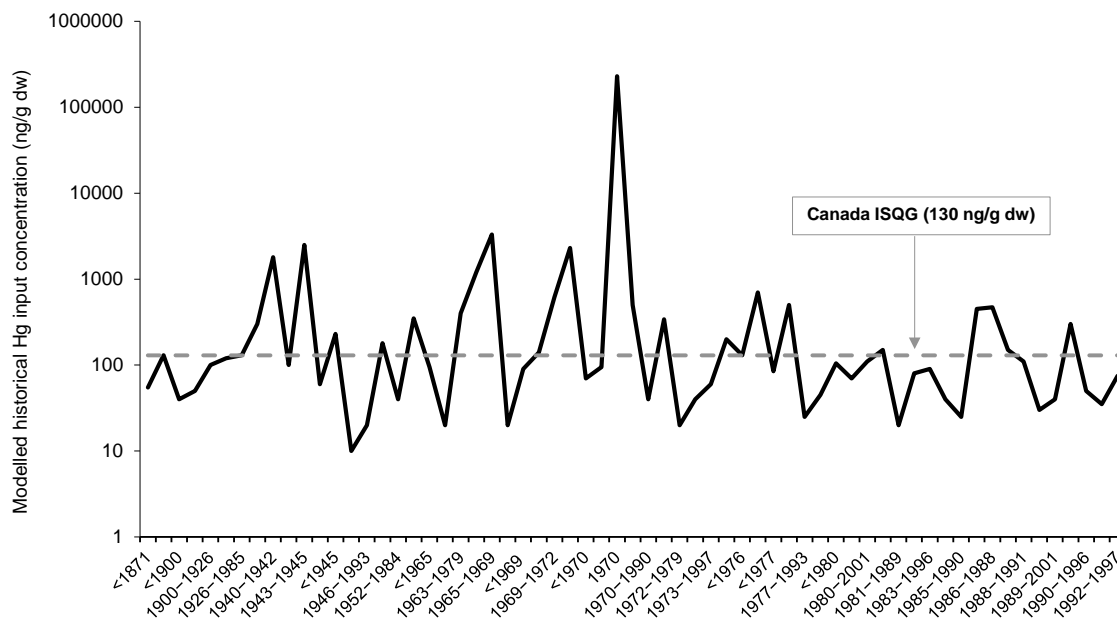


Fig. 1.5.1 Modeled mercury (Hg) depositional history (input concentration in ng/g dry weight; dw) between the period before 1871 and the year 2001 based on sediment cores collected from 17 sites in the Strait of Georgia. Adapted from Johannessen *et al.* (2005). The x-axis was set up as discrete (and variable) time periods in order of start date and not in a sequential timescale. The dashed line represents the Canadian Interim Sediment Quality Guideline (ISQG) for mercury in marine and estuarine sediments (CMME, 1999).

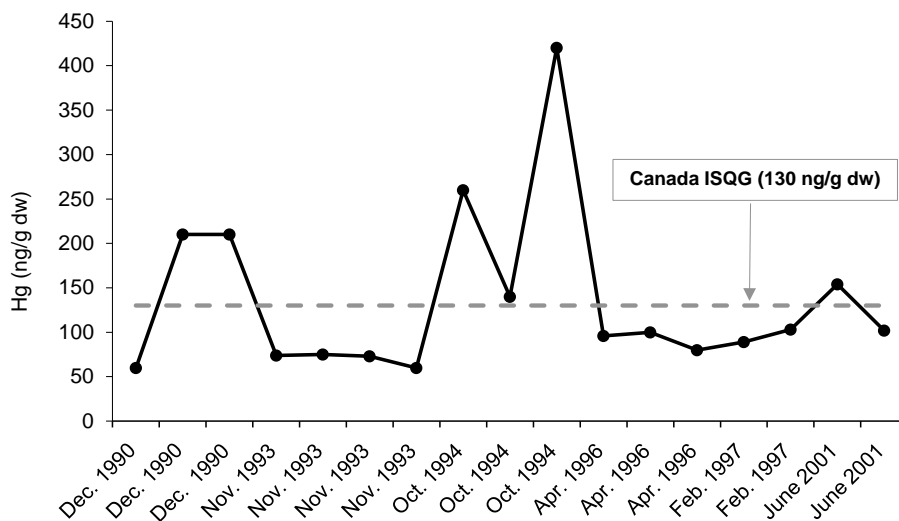


Fig. 1.5.2 Measured surface mercury (Hg ng/g dw) in sediment box cores collected in the Strait of Georgia, Vancouver Harbour, and Howe Sound from 1990 to 2001. The dashed line represents the Canada Interim Sediment Quality Guideline (ISQG) for mercury in marine and estuarine sediments (CCME, 1999).

The implementation of regulations for chlor-alkali plants and pulp and paper mills, as well as source control of other industrial and municipal activities (e.g., dental practices) have helped to reduce point source releases into the coastal environment in Canada. Emissions of mercury into the atmosphere through coal-fired generating stations represents a major source of concern requiring global action, something that was underscored in the 2013 Minamata Convention on Mercury.

References

- Alava, J.J., Cisneros-Montemayor, A.M., Sumaila, R., and Cheung, W.W.L. 2018. Projected amplification of food web bioaccumulation of MeHg and PCBs under climate change in the Northeastern Pacific. *Scientific Reports* **8**:13460, doi:10.1038/s41598-018-31824-5.
- CCME (Canadian Council of Ministers of the Environment). 1999. Canadian sediment quality guidelines for the protection of aquatic life: Mercury. Canadian Environmental Quality Guidelines, Winnipeg.
- Johannessen, S.C., Macdonald, R.W. and Magnus Eek, K. 2005. Historical trends in mercury sedimentation and mixing in the Strait of Georgia, Canada. *Environmental Science and Technology* **39**: 4361–4368.
- Lamborg, C.H., Hammerschmidt, C.R., Bowman, K.L., Swarr, G.J., Munson, K.M., Ohnemus, D.C., Lam, P.J., Heimbürger, L.E., Rijkenberg, M.J.A. and Saito, M.A. 2014. A global ocean inventory of anthropogenic mercury based on water column measurements. *Nature* **512**: 65–68.
- Mason, R.P., Choi, A.L., Fitzgerald, W.F., Hammerschmidt, C.R., Lamborg, C.H., Soerensen, A.L. and Sunderland, E.M. 2012 Mercury biogeochemical cycling in the ocean and policy implications. *Environmental Research* **119**: 101–117.
- Noël, M., Jeffries, S., Lambourn, D.M., Telmer, K., Macdonald, R. and Ross, P.S. 2016. Mercury accumulation in harbour seals from the Northeastern Pacific Ocean: The role of transplacental transfer, lactation, age and location. *Archives of Environmental Contamination and Toxicology* **70**: 56–66.
- Peterson, S.H., Ackerman, J.T. and Costa, D.P. 2015. Marine foraging ecology influences mercury bioaccumulation in deep diving northern elephant seals. *Proceedings of the Royal Society B* **282**: 20150710, <https://doi.org/10.1098/rspb.2015.0710>.
- UNEP (United Nations Environment Programme). 2013. Global Mercury Assessment 2013: Sources, Emissions, Releases and Environmental Transport. UNEP Chemicals Branch, Geneva, Switzerland.

1.6 Temporal trends of marine contamination by copper in British Columbia, Canada: The legacy of the Britannia Mine

Juan José Alava¹, Karin Bodtker², Jennifer Chapman², Amber Dearden² and Aroha Miller²

¹Institute for the Oceans and Fisheries, University of British Columbia, Vancouver, British Columbia, Canada

²Ocean Wise Research Institute, Ocean Wise Conservation Association, Vancouver, British Columbia, Canada

Throughout the 20th century, Britannia Mine in British Columbia, Canada, was a major source of copper and other metal pollutants into the marine environment of Átl'ka7tsem/Txwnéwu7ts/Howe Sound (Danon-Shaffer, 2002; Wilson *et al.*, 2005; Alava and Bodtker, 2017; Chapman and Dearden, 2020). The mine, which is now a museum (Britannia Mine Museum), is located in southwestern British Columbia, *i.e.*, 49°38'N, 123°12'W (Wilson *et al.*, 2005; O'Hara, 2007). The mine covered an area of 28 to 36.5 km² and consisted of a series of tunnels and some open-cast mining (Wilson *et al.*, 2005; O'Hara, 2007). The Britannia Mine Museum now occupies part of the mine site, but the mine itself has been closed for approximately 45 years. From 1898 until 1974, four to 40 million litres of metal-laden waters (acid mine drainage) were discharged daily into Átl'ka7tsem/Txwnéwu7ts/Howe Sound, depending on the time of year (Danon-Shaffer, 2002; Wilson *et al.*, 2005). Historically, the site was referred to as being one of the worst sources of water pollution involving metal contamination in North America (Warketin, 2000; O'Hara, 2007) and the worst point source of metal pollution in British Columbia (McCandless, 1995). Acid mine drainage from the Britannia site included contaminants such as copper, aluminum, iron, zinc, and manganese.

In the late 1990s, it became clear that ongoing contamination from the former Britannia Mine was an issue requiring attention. For example, copper concentrations in mine drainage were thousands of times greater than provincial water quality guidelines (WQG) of the time (Wilson *et al.*, 2005; O'Hara 2007; Government of BC, Ministry of Environment, 2016), and there were clear indicators of the effects in the shoreline community. In 1999, concentrations of dissolved copper in seawater detected in nearshore waters close to the mouth of Britannia Creek (Levings *et al.*, 2005) exceeded British Columbia WQG (Government of BC, Ministry of Environment, 2016) by approximately 20 times and was lethal to caged salmon and local mussels (Barry *et al.*, 2000). Research on the effects of metal contamination at Britannia prior to remediation has shown detrimental impacts on salmon fry (Barry *et al.*, 2000), mussels, algae, and invertebrates in Átl'ka7tsem/Txwnéwu7ts/Howe Sound (Levings *et al.*, 2005; Levings *et al.*, 2004). Primary production in Átl'ka7tsem/Txwnéwu7ts/Howe Sound was reduced (Levings *et al.*, 2004) – contaminated sediments in Britannia Creek were toxic to important food sources such as midge larvae and amphipods; sand dollar reproduction was inhibited (Levings *et al.*, 2004); and blue mussel growth was impaired (Grout and Levings, 2001).

Remediation efforts underway since 2001 have resulted in significant improvements in the nearshore environment. However, despite these efforts, metal contamination from past operations of the mine lingers at some sites (Alava and Bodtker, 2017; Chapman and Dearden, 2020). While the flow of contaminated water directly into Átl'ka7tsem/Txwnéwu7ts/Howe Sound has decreased, sampling of marine porewater in areas adjacent to the groundwater management system and in near-field reference sites indicate that metal contamination continues to occur above WQGs (Golder Associates, 2017,

2018). In 2017 and 2018, British Columbia WQGs were exceeded for several metals (*i.e.*, boron, cadmium, chromium, copper, lead, manganese, nickel and zinc) (Golder Associates, 2017, 2018). Additionally, in a single 2017 sampling event, mercury exceeded the chronic and national safety levels (Golder Associates, 2017, 2018). Copper is a contaminant of primary concern that has either been relatively stable or slightly declined in recent years (Fig. 1.6.1). Between 2003 and 2018, surveys of intertidal communities at the three sites nearest the mine showed signs of recovery. However, recovery is less pronounced at the most contaminated site (Chapman and Dearden, 2020). Continued presence of metals and their impacts suggest that the area is unlikely to return to its pre-industrial state; therefore, the closure plan targets acceptable environmental conditions with continued operation of some mitigation measures (*e.g.*, the groundwater management system) (Golder Associates, 2010).

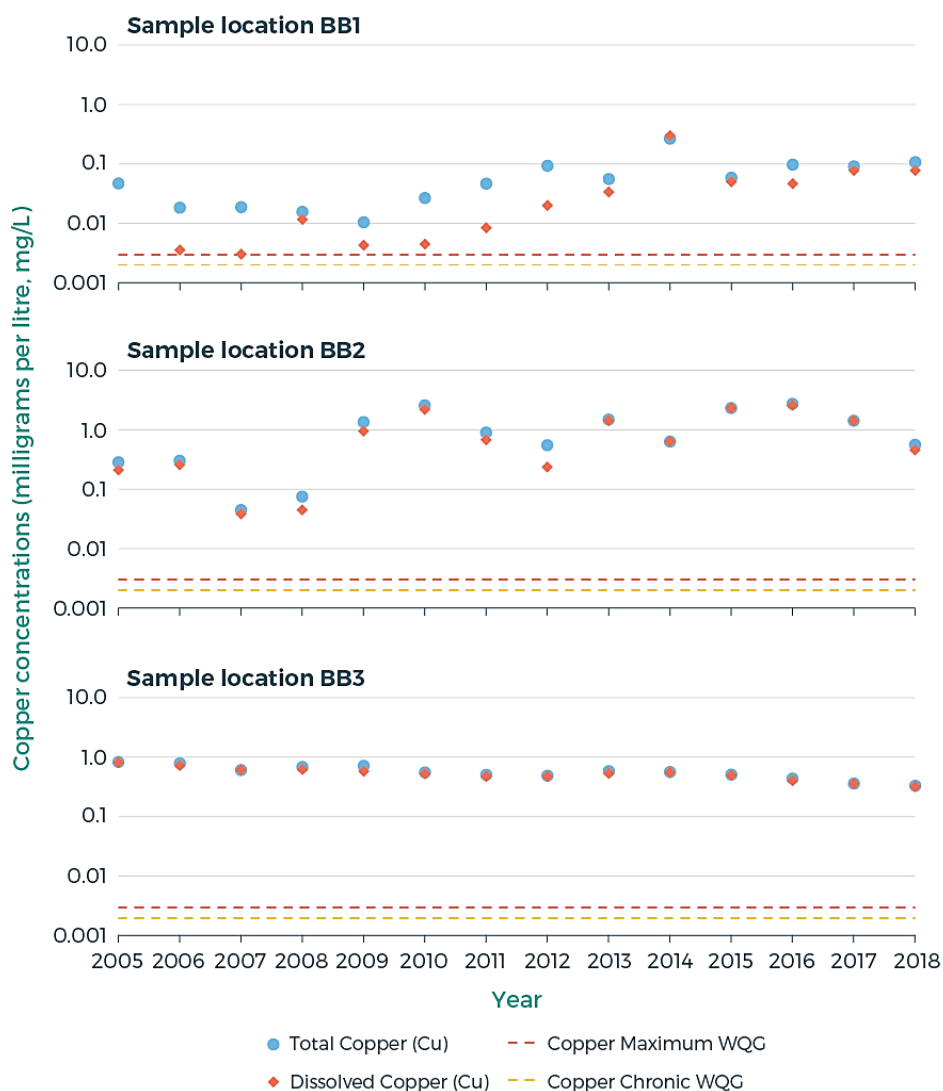


Fig. 1.6.1 Time series of total and dissolved copper concentrations (mg/L) in marine porewater for sampling locations near the groundwater management system (*i.e.*, BB-1, BB-2, BB-3). The chronic and maximum water quality guidelines (WQGs) (yellow and red dashed lines, respectively) are set for the protection of marine aquatic life in British Columbia (Golder Associates, 2017, 2018). Replicated from Chapman and Dearden (2020).

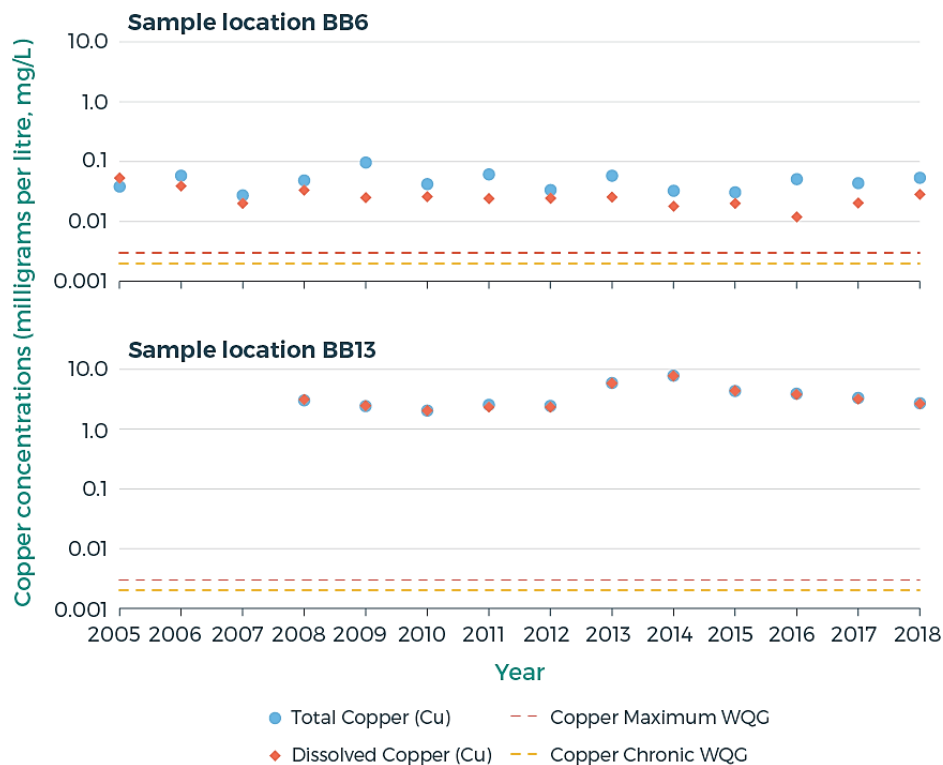


Fig. 1.6.1 Continued. Time series of total and dissolved copper concentrations (mg/L) in marine porewater for near-field reference sites (*i.e.*, BB-6 and BB-13).

Contamination by acid mine drainage in Átl'ka7tsem/Txwnéwu7ts/Howe Sound can and has had important implications for First Nation communities that rely on salmon and seafood from Átl'ka7tsem/Txwnéwu7ts/Howe Sound and the Squamish River Estuary (Alava and Bodtker, 2017; Chapman and Dearden, 2020). Salmon is not only a traditional food but holds strong spiritual and cultural significance. The idea that traditional foods can be contaminated and unhealthy is foreign to some elders who have continued to eat seafood despite warnings of contamination and closures of commercial fisheries (Alava and Bodtker, 2017). Both federal and provincial government regulations have played a role in the remediation and clean-up of the former Britannia Mine site. Several remediation orders were issued between the 1970s and late 1990s under the former *Waste Management Act*, replaced by the *Environmental Management Act* in 2004; both are provincial legislation (Danon-Shaffer, 2002). Since 2004, an environmental monitoring program and ecological risk assessment have been underway and remediation is on-going. Mussels, common to Átl'ka7tsem/Txwnéwu7ts/Howe Sound, are now naturally colonizing the shoreline except in a few localized areas, and rockweed, a common Átl'ka7tsem/Txwnéwu7ts/Howe Sound seaweed that was long absent along the shoreline for a kilometer on either side of Britannia Creek, is now also present, except at a few sites (Wernick *et al.*, 2010). In general, the environmental quality in the waters and marine ecosystems of Átl'ka7tsem/Txwnéwu7ts/Howe Sound has shown signs of improvement following concerted monitoring and remediation efforts (Miller *et al.*, 2020).

References

- Alava, J.J. and Bodtker, K. 2017. Metal contamination from the Britannia Mine site: lingering problems, ongoing remediation, pp. 144–155 *in: Ocean Watch: Howe Sound Edition edited by K. Bodtker*, Coastal Ocean Research Institute, Vancouver Aquarium Marine Science Centre, Vancouver, British Columbia, Canada, 365 pp.
- Barry, K.L., Grout, J.A., Levings, C.D., Nidle, B. and Piercey, G.E. 2000. Impacts of acid mine drainage on juvenile salmonids in an estuary near Britannia Beach in Howe Sound, British Columbia. *Canadian Journal of Fisheries and Aquatic Sciences* **57**: 2032–2043.
- Chapman, J. and Dearden, A. 2020. Britannia Mine: contamination, remediation and monitoring, pp. 255–263 *in: Ocean Watch Átl'ka7tsem/Txwnéwu7ts/Howe Sound Edition 2020 edited by A. Miller, J. Chapman, A. Dearden, and P. Ross*. Ocean Wise Research Institute, Ocean Wise Conservation Association, Vancouver, B.C., Canada, 388 pp. <http://oceanwatch.ca>.
- Danon-Shaffer, M.N. 2002. Investigation, remediation and cost allocation of contaminants from the Britannia Mine in British Columbia: A case study. *Environmental Forensics* **3**: 15–25.
- Golder Associates. 2010. Overall Closure Plan Framework Document. Britannia Mine Remediation Project. Submitted to: Ministry of Agriculture and Lands. 15 June 2010. Report Number 012-1830/32120.
- Golder Associates. 2017. Britannia Mine Environmental Monitoring, 2017 Data Report. Submitted to Ministry of Forests, Lands and Natural Resource Operations. 23 February 2018. Report Number: 1786668-001-R-Rev0-2000.
- Golder Associates. 2018. Britannia Mine Environmental Monitoring, 2018 Data Report. Submitted to Ministry of Forests, Lands and Natural Resource Operations. 27 February 2019. Report Number: 1786668-002-R-Rev0-2000.
- Government of BC, Ministry of Environment. 2016. Water Quality Criteria for Copper, Overview Report, July 1987, <http://www.env.gov.bc.ca/wat/wq/BCguidelines/copper/copper.html> (accessed September 7, 2016).
- Grout, J.A. and Levings, C.D. 2001. Effects of acid mine drainage from an abandoned copper mine, Britannia Mines, Howe Sound, British Columbia, Canada, on transplanted blue mussels (*Mytilus edulis*). *Marine Environmental Research* **51**: 265–288.
- Levings, C.D., Barry, K.L., Grout, J.A., Piercey, G.E., Marsden, A.D., Coombs, A.P. and Mossop, B. 2004. Effects of acid mine drainage on the estuarine food web, Britannia Beach, Howe Sound, British Columbia, Canada. *Hydrobiologia* **525**: 185–202.
- Levings, C.D., Varela, D.E., Mehlenbacher, N.M., Barry, K.L., Piercey, G.E., Guo, M. and Harrison, P.J. 2005. Effect of an acid mine drainage effluent on phytoplankton biomass and primary production at Britannia Beach, Howe Sound, British Columbia. *Marine Pollution Bulletin* **50**: 1585–1594.
- McCandless, R.G. 1995. The Britannia Mine: historic landmark or environmental liability? BC Professional Engineer, April 1995, pp. 4–7.
- Miller, A., Chapman, J., Dearden, A., and Ross, P. (Eds.) 2020. Ocean Watch Átl'ka7tsem/Txwnéwu7ts/Howe Sound Edition 2020. Ocean Wise Research Institute, Ocean Wise Conservation Association, Vancouver, B.C., Canada. 388 pp. <http://oceanwatch.ca>.
- O'Hara, G. 2007. Water management aspects of the Britannia Mine Remediation Project, British Columbia, Canada. *Mine Water and the Environment* **26**: 46–54.
- Warkentin, D. 2000. BC's Green Party News 49, Summer.

- Wernick, B.G., Nikl, L.H., Seguin, S.R. and Sinnett, G. 2010. Britannia Mine Remediation Project - Integrating ecological monitoring with remediation activities *in: Mine Closure 2010: Proceedings of the Fifth International Conference on Mine Closure edited by A. Rourie, M. Tibbett and J. Wiertz, Viña del Mar, Chile.*
- Wilson, B., Lang, B. and Pyatt, F.B. 2005. The dispersion of heavy metals in the vicinity of Britannia Mine, British Columbia, Canada. *Ecotoxicology and Environmental Safety* **60**: 269–276.

1.7 Assessment of marine debris pollution in the marine-coastal region of British Columbia, Canada

Kate Le Souef¹ and Juan José Alava^{1,2}

¹Ocean Pollution Research Program, Coastal and Ocean Research Institute, Vancouver Aquarium Marine Science Centre, Vancouver, British Columbia, Canada

²Institute for the Oceans and Fisheries, University of British Columbia, Vancouver, British Columbia, Canada

Since 1994, the Great Canadian Shoreline Cleanup (GCSC), a joint conservation initiative of the Vancouver Aquarium Marine Science Centre and World Wildlife Fund Canada, has been encouraging Canadians to coordinate cleanups at their local shoreline. The effort of this initiative offers an opportunity to survey, monitor and collect marine debris. Using data from the shoreline cleanups led by this program, we report the temporal trends and composition of marine debris in the Canadian Pacific (British Columbia) and the implications for marine conservation and management of solid waste.

Volunteers are inspired to participate to reduce the risk that shoreline litter poses to aquatic wildlife. Cleanups take place anywhere land connects to water, including beaches, lakes, rivers and streams. Participation in this national program grows year after year, with more than 2,000 cleanups taking place across the country throughout 2015 (Fig. 1.7.1). Each cleanup coordinator submits data about the litter collected. These data show the same patterns each year at most shorelines in Canada. Cigarette butts and single-use plastic items, such as food wrappers, straws, plastic bags and drink bottles usually dominate (*i.e.*, 80–90%) the litter found not only in the Canadian Pacific (Konecny *et al.*, 2018), but around all Canadian shorelines (Table 1.7.1).

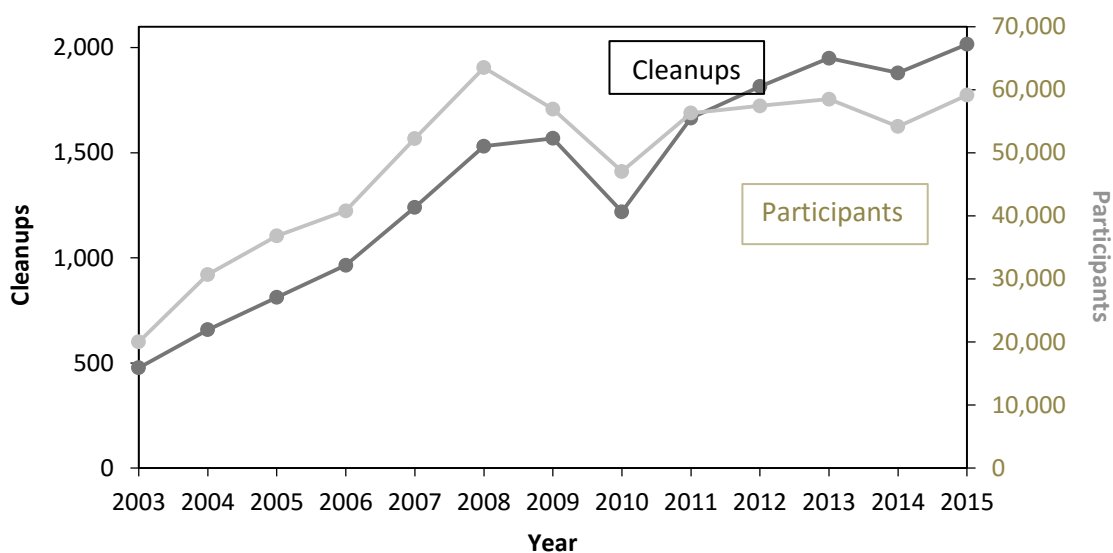


Fig. 1.7.1 Number of cleanups and participants registered across Canada in the Great Canadian Shoreline Cleanup since 2003.

Table 1.7.1 The 12 most common items (the ‘Dirty Dozen’) found on Canadian shorelines in 2015.

Ranking	Item	Number of items collected
1	Cigarette butts	409,417
2	Food wrappers	93,129
3	Plastic bottle caps	50,904
4	Plastic beverage bottles	37,769
5	Beverage cans	27,814
6	Other plastics and foam	27,110
7	Straws and stirrers	27,047
8	Other plastic bags	25,047
9	Metal bottle caps	22,093
10	Plastic grocery bags	20,492
11	Plastic lids	19,365
12	Paper cups and plates	17,819

In the southern Strait of Georgia, which includes the large urban area of Vancouver, and in the Victoria area, on Juan de Fuca Strait, cigarettes and cigarette filters, which are made of plastic, account for almost 50% of litter recovered (Konecny *et al.*, 2018). However, on the exposed coast of British Columbia, including the north coast of British Columbia (*i.e.*, Haida Gwaii and Prince Rupert), litter composition tends to be different, with large plastic bottles and plastic bags being the dominant items. As this coast is exposed to the Pacific Ocean, such items are from all over the world and wash up on this shoreline, including some single use plastics, but also large quantities of plastic from fishing, shipping and improperly managed land-based litter. Natural disasters, such as the Great East Japan Earthquake in 2011, have also contributed to shoreline litter in Canada in the form of ‘tsunami debris’ (Kataoka *et al.*, 2018). Anecdotal reports suggest that tsunami debris only represents a small fraction of the trash found on shorelines.

Cleanup data is opportunistic, because data will only be collected if volunteers decide to coordinate an event. The west coast of British Columbia is remote and difficult to access, and the volume of debris is often so large that helicopters, barges and trucks are required to move it. Therefore, only a small number of cleanups take place on the exposed Pacific coast. For example, in 2013, 19 cleanups took place on this coast, removing 2,295 kg of litter. In 2014, 22 cleanups took place, removing 13,860 kg of litter. In 2015, 31 cleanups took place, removing 16,916kg of litter. The GCSC recovered a total of 109,557 kg of trash ($27,389 \pm 66,634$ kg/year) in British Columbia between 2013 and 2016 (Konecny *et al.*, 2018). The most common items found on these cleanups were rope, fishing buoys and plastic water bottles. The litter data are difficult to compare year to year (Table 1.7.2 and Fig. 1.7.2) because significant funding was provided by the Japanese Government to the federal and provincial governments of Canada to support large-scale cleanups in 2014 and 2015.

Table 1.7.2 Number of cleanups and mass of litter collected from the exposed west coast of British Columbia.

Year	Number of cleanups	Litter collected (kg)
2006	31	2,749
2007	21	5,169
2008	18	2,292
2009	13	3,996
2010	24	4,166
2011	15	1,466
2012	28	5,273
2013	19	2,295
2014	22	13,860
2015	31	16,916

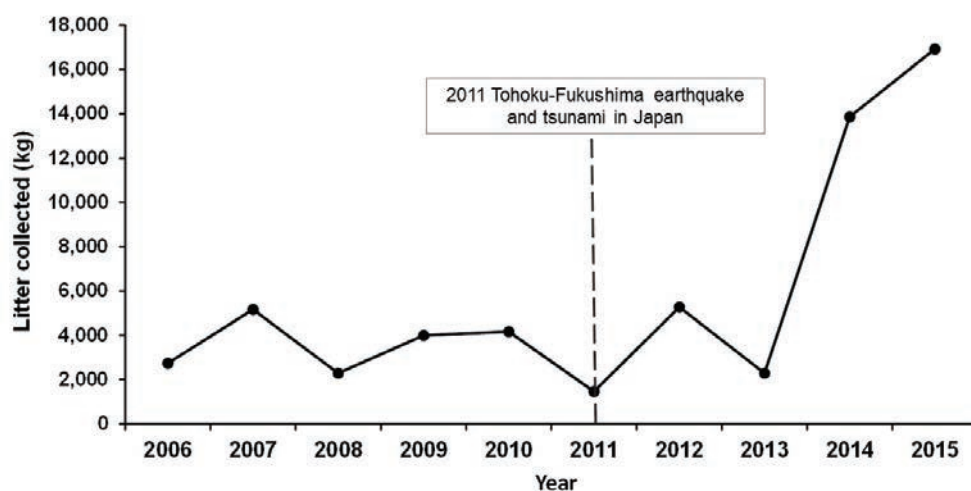


Fig. 1.7.2 Litter collected (kg) on the exposed west coast of British Columbia since 2006. Note that significant funding for large-scale cleanups was received in 2014.

The Great Canadian Shoreline Cleanup is recognized as one of the largest direct action conservation programs, as well as the most significant contributor to the International Coastal Cleanup, in Canada.

References

- Kataoka, T., Clarke Murray, C. and Isobe, A. 2018. Quantification of marine macro-debris abundance around Vancouver Island, Canada, based on archived aerial photographs processed by projective transformation. *Marine Pollution Bulletin* **132**: 44–51.
- Konecny, C., Fladmark, V. and De la Puente, S. 2018. Towards cleaner shores: Assessing the Great Canadian Shoreline Cleanup's most recent data on volunteer engagement and litter removal along the coast of British Columbia, Canada. *Marine Pollution Bulletin* **135**: 411–417.

1.8 Spatial assessment of microplastic pollution in the marine-coastal region of British Columbia, Canada

Juan José Alava¹, Alejandra Díaz², Ellika Crichton², Esther Gies², Marie Noel² and Peter S. Ross²

¹Institute for the Oceans and Fisheries, University of British Columbia, Vancouver, British Columbia, Canada

²Coastal and Ocean Research Institute, Vancouver Aquarium Marine Science Centre, Vancouver, British Columbia, Canada

Microplastic pollution in the Northeast Pacific represents a new emerging threat for marine organisms and ocean health (Doyle *et al.*, 2011; Desforges *et al.*, 2014; Alava, 2020). Along with other anthropogenic pollutants, microplastics have become one of the emerging contaminants of concern in the marine environment and sensitive ecosystems of British Columbia, Canada (Gies *et al.*, 2018; Kazmiruk *et al.*, 2018; Alava, 2019, 2020). An estimated 30 billion plastic particles are released annually from a wastewater treatment plant (WWTP; <https://www.sciencedirect.com/topics/agricultural-and-biological-sciences/wastewater-treatment>) near Vancouver to the ocean, highlighting the pollution risks by microplastic exposure to marine biota (Gies *et al.*, 2018). While a monitoring program to assess the impact of plastics or microplastics in the marine environment of British Columbia was implemented (*i.e.*, PollutionTracker, <https://pollutiontracker.org/contaminants/microplastics/>) by the Ocean Pollution Research Program at Ocean Wise Conservation Association (Ocean Wise, 2020), a pioneer study on the abundance, composition and distribution of marine microplastic pollution in the offshore and coastal marine environment of this region was recently documented by Desforges *et al.* (2014). In this report, we summarize the major finding from that study and other research efforts focused on microplastics in sediments (*e.g.*, Cluzard *et al.*, 2015; Kazmiruk *et al.*, 2018).

Sampling stations ($n = 34$) were surveyed along a transect encompassing the Northeast Pacific Ocean (NE Pacific), and several sampling sites off the west coast of Vancouver Island (WCVI), Queen Charlotte Sound (QCS), and the Strait of Georgia (SoG). Transects were completed aboard two oceanographic research cruises: the CCGS *John P. Tully* cruise in August 2012, and the La Perouse Monitoring Program cruise of September 2012 (Fisheries and Oceans Canada, Institute of Ocean Sciences). Samples were acid-digested and plastics were characterized using light microscopy by type (fibres or fragments) and size (<100, 100–500, 500–1000 and >1000 μm). The concentration of microplastics was 4 to 27 times greater at sites nearshore (SoG, WCVI, and QCS) than sites offshore in the NE Pacific Ocean. The highest concentrations of microplastics in subsurface seawater (4000–5000 particles/ m^3) were found within the SoG (Desforges *et al.*, 2014; Fig. 1.8.1).

The mean size of microplastic particles was $606 \pm 221 \mu\text{m}$. The smallest particle documented was $64.8 \mu\text{m}$, and the largest was $5810 \mu\text{m}$. The majority of identified particles were fibres/filaments and angular plastic fragments, with few cases of other types of plastics (thin films or round fragments). Fibres/filaments accounted for approximately three quarters of the identified particles in the collective dataset (Fig. 1.8.1). The contribution of fibres to total plastics decreased with increasing distance from shore (Fig. 1.8.1), including the percentage of fibres in QCS and the SoG, where they were greater than those in NE Pacific Ocean (Desforges *et al.*, 2014).

The extent of microplastic pollution impacts on marine biota was also assessed in two species of zooplankton (*i.e.*, the calanoid copepod *Neocalanus cristatus* and the euphausiid *Euphausia pacifica*),

resulting in ingestions of 1 particle/every 34 copepods and 1 particle/every 17 euphausiids (Desforges *et al.*, 2015). Microplastic pollution in coastal intertidal sediments (*i.e.*, 76,500 particles/m³) from a location (Bayne Sound) off Vancouver Island within the SoG has also been documented (Cluzard *et al.*, 2015). Recent data by Kazmiruk *et al.* (2018) showed that microplastics (*i.e.*, microfibers, microbeads and microfragments) are widely spread and found in all sampling locations within Baynes Sound and associated body waters around Lambert Channel. Microbeads occurred in the highest number ($\approx 25,000/\text{kg}$ dry sediment) relative to microfibers and microfragments, which occurred in similar amounts (100–300/kg dry sediment), indicating ubiquitous pollution with these emerging contaminants in this sensitive area (Kazmiruk *et al.*, 2018). Microplastics were also found in juvenile Chinook salmon (*i.e.*, maximum concentration > 2 microfibers/g of fish), sediments (*i.e.*, maximum concentration, ~ 50 microfibers/kg of sediment) and water column (*i.e.*, maximum concentration, ~ 1200 microfibers/m³) in Cowichan Bay, Vancouver Island (Collicutt *et al.*, 2019). These findings provide baseline information that can support risk management and control strategies by authorities and municipalities from cities around the region to reduce and mitigate sources of microplastic pollution in the ocean and impacts in marine biota.

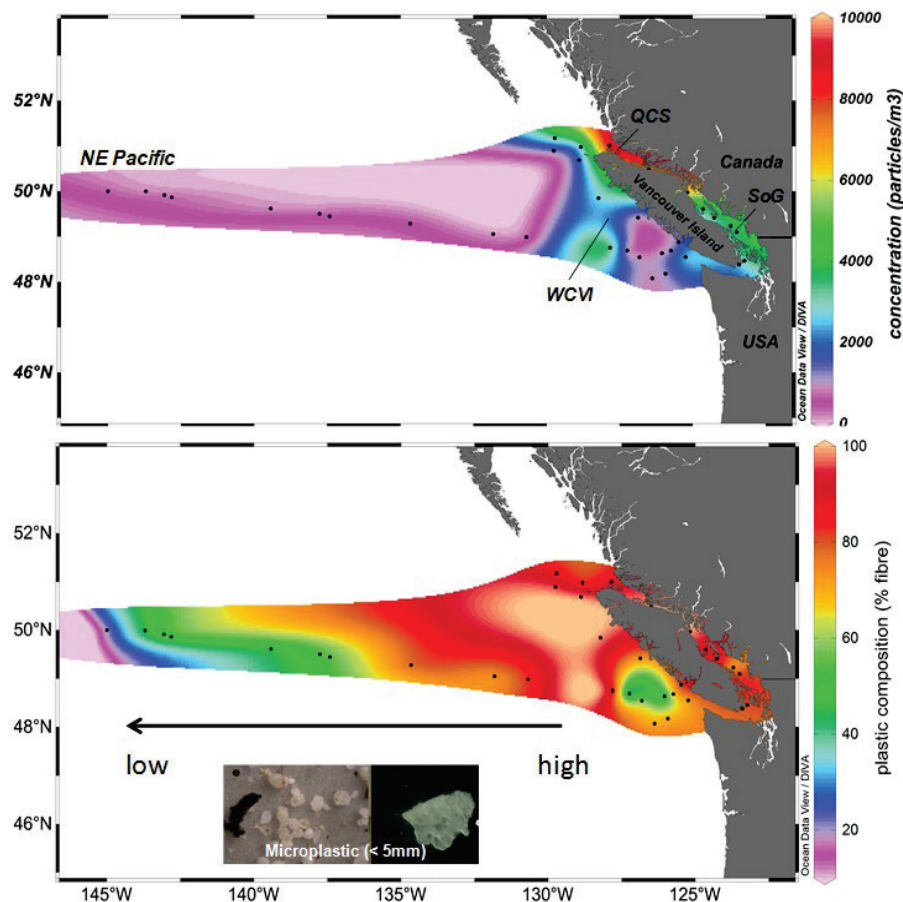


Fig. 1.8.1 Total microplastic concentration (particles/m³; detected particles $> 62 \mu\text{m}$) in subsurface waters, 4.5 m (top), and composition pattern expressed as % fibre (bottom) in offshore waters of British Columbia and the Strait of Georgia. The figure shows a high fibre content at nearshore stations and decrease with distance offshore. Sampling stations are depicted as black dots. NE Pacific = Northeast Pacific Ocean, WCVI = west coast Vancouver Island, QCS = Queen Charlotte Sound, and SoG = Strait of Georgia. Adapted from Desforges *et al.* (2014).

References

- Alava, J.J. 2019. Legacy and emerging pollutants in marine mammals' habitat from British Columbia, Canada: Management perspectives for sensitive marine ecosystems, pp. 87–114 *in*: Stewarding the Sound: The Challenge of Managing Sensitive Ecosystems *edited by* L.I. Bendell, P. Gallagher, L. Wood and S. McKeachie. CRC Press/Taylor and Francis Group. 148 pp.
<https://www.crcpress.com/Stewarding-the-Sound-The-Challenge-of-Managing-Sensitive-Coastal-Ecosystems/Bendell-Gallagher-McKeachie-Wood/p/book/9780367112035>.
- Alava, J.J. 2020. Modelling the bioaccumulation and biomagnification potential of microplastics in a cetacean foodweb of the Northeastern Pacific: A prospective tool to assess the risk exposure to plastic particles. *Frontiers in Marine Science* **7**: 566101, doi: 10.3389/fmars.2020.566101.
- Cluzard, M., Kazmiruk, T.N., Kazmiruk, V.D. and Bendell, L.I. 2015. Intertidal concentrations of microplastics and their influence on ammonium cycling as related to the shellfish industry. *Archives of Environmental Contamination and Toxicology* **69**: 310–319.
- Collicutt, B., Juanes, F. and Dudas, S.E. 2019. Microplastics in juvenile Chinook salmon and their nearshore environments on the east coast of Vancouver Island. *Environmental Pollution* **244**: 135-142.
- Desforges, J.P.W., Galbraith M., Dangerfield, N. and Ross, P.S. 2014. Widespread distribution of microplastics in subsurface seawater in the NE Pacific Ocean. *Marine Pollution Bulletin* **79**: 94–99.
- Desforges, J.P.W., Galbraith, M. and Ross, P.S. 2015. Ingestion of microplastics by zooplankton in the Northeast Pacific Ocean. *Archives of Environmental Contamination and Toxicology* **69**: 320–330.
- Gies, E.A., LeNoble, J.L., Noël, M., Etemadifar, A., Bishay, F., Hall, E.R. and Ross, P.S. 2018. Retention of microplastics in a major secondary wastewater treatment plant in Vancouver, Canada. *Marine Pollution Bulletin* **133**: 553–561, doi: 10.1016/j.marpolbul.2018.06.006.
- Kazmiruk, T.N., Kazmiruk, V.D. and Bendell, L.I. 2018. Abundance and distribution of microplastics within surface sediments of a key shellfish growing region of Canada. *PLoS ONE* **13**: e0196005, <https://doi.org/10.1371/journal.pone.0196005>.
- Ocean Wise. 2020. Microplastics. PollutionTracker. Coastal Ocean Research Institute. Vancouver, BC, Canada. <https://pollutiontracker.org/contaminants/microplastics/>.

2 China

2.1 Polychlorinated biphenyls (PCBs) in the marine environment of the China coast

Peng Zhang, Ying Bian, Chuanli Huo and Guangshui Na

National Marine Environmental Monitoring Center, Dalian, China

Polychlorinated biphenyls (PCBs) are one of the 12 legacy persistent organic pollutants (POPs) targeted by the Stockholm Convention on persistent organic pollutants (POPs) (UNEP, 2001). From 1950 to 2010, the total unintentionally produced PCB (UP-PCB) emissions in China were estimated to be 146.1 t, approximately 98.1% of which were from cement (132.5 t) and steel industries (6.3 t). The emissions from the Chinese coking industry were 115 kg, less than 0.1% of the total UP-PCB emissions (Cui *et al.*, 2013).

Regionally, the distribution of PCB pollution tends to be related to drainage area, industrial patterns, and pollution emissions during the two decades since their production and use were banned (Xing *et al.*, 2005). The concentration of PCBs in the atmosphere over China came from two sources, one of which was the combustion that emerged from contemporary high emissions, while the second source, a critical one, was re-emissions/volatilization (Hogarh *et al.*, 2012). A few previously published research papers on the legacy of PCBs in the coastal areas of Chinese marginal seas indicates that the concentrations of PCBs in sediment sampling from the vicinities of Dalian and Tianjin inshore, where chemical factories for producing paints were established, were relatively higher than those in other coastal areas (Zhang *et al.*, 2010; Zhao *et al.*, 2010; Hogarh *et al.*, 2012). Samples taken from Yellow River and Huaihe River and some branches of Yangtze River (Qiangtangjiang, Huangpujiang, Hanshui rivers and Dongting Lake) in central China had average PCB concentrations that have not reached the effects range low (ERL) level, suggesting a lack of or low risk posed by PCBs. Other areas, such as the Daliaohe, Haihe and Yellow River estuaries, as well as Jinzhou Bay near the Bohai Sea, had relatively low levels of contamination (Xing *et al.*, 2005).

The reports on PCB concentrations in seawater are relatively scarce. The existing data demonstrate that the PCB concentrations in surface seawater in the Bohai Sea and East China Sea are lower than EPA guideline limits. However, higher-level PCB concentrations have been detected in fish and shellfish samples collected from Dalian city (location 2), Yantai city (location 14) and Jiaozhou Bay (location 18) compared to other coastal areas in the Bohai Sea, Yellow Sea and East China Sea, as shown in Figure 2.1.1.

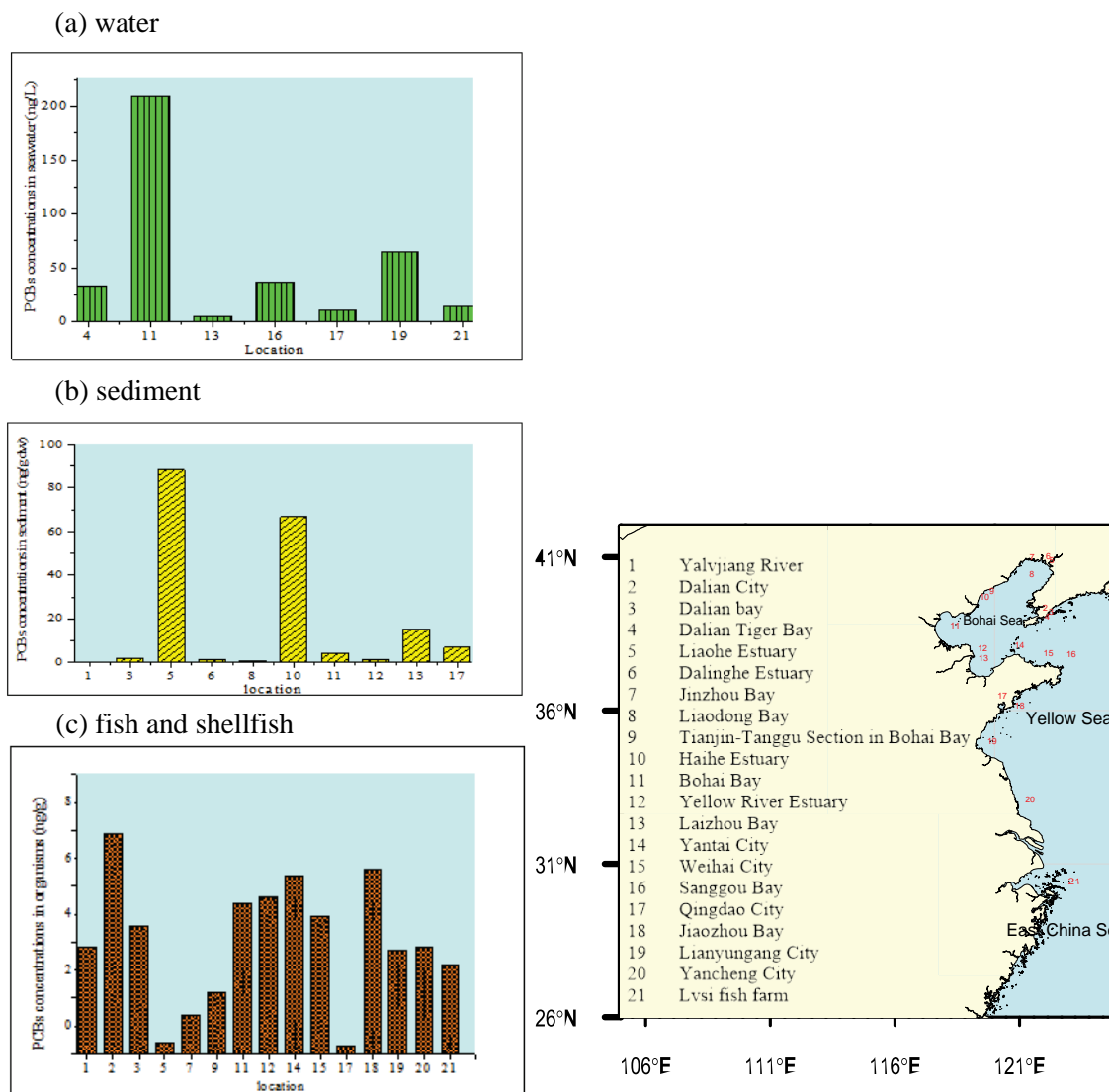


Fig. 2.1.1 Spatial distribution of polychlorinated biphenyls (PCBs) in the marine environment and organisms in Chinese marginal seas for (a) water, (b) sediments, and (c) fish and shellfish. Location numbers are identified in the map key.

However, in all the study areas, including cities, bays and estuaries shown in Figure 2.1.1c, the residual levels of PCBs in marine fish and shellfish were far below a federal tolerance of PCBs (2000 ng/g wet weight) provided by the United States Food and Drug Administration (USFDA, 2020) which was used as a criterion to assess the PCB contamination status in organisms, including fish and fisheries products such as fresh or saltwater finfish, crustaceans, and mollusks, as well as other forms of aquatic animal life (Xing *et al.*, 2005; Pan *et al.*, 2016). Xia *et al.* (2012) pointed out that the total PCB concentrations in yellow croaker and silver pomfret collected from Dalian, Qingdao, Shanghai and Zhoushan cities were at the low end of the global range, which may be related to the smaller usage and shorter consumption history of PCBs in China.

References

- Cui, S., Qi, H., Liu, L.Y., Song, W.W., Ma, W.L., Jia, H.L., Ding, Y.S. and Li, Y.F. 2013. Emission of unintentionally produced polychlorinated biphenyls (UP-PCBs) in China: Has this become the major source of PCBs in Chinese air? *Atmospheric Environment* **67**: 73–79.
- Hogarh, J.N., Seike, N., Kobara, Y., Habib, A., Nam, J.J., Lee, J.S., Li, Q., Liu, X., Li, J., Zhang, G. and Masunaga, S. 2012. Passive air monitoring of PCBs and PCNs across East Asia: A comprehensive congener evaluation for source characterization. *Chemosphere* **86**: 718–726.
- Pan, H., Geng J., Qin, Y., Tou, F., Zhou, J., Liu, M. and Yang, Y. 2016. PCBs and OCPs in fish along coastal fisheries in China: Distribution and health risk assessment. *Marine Pollution Bulletin* **111**: 483–487.
- UNEP (United Nations Environment Programme). 2001. UNEP in 2001. Part II. Safe chemical management.
- USFDA (United States Food and Drug Administration). 2020. CHAPTER 9, Environmental Chemical Contaminants and Pesticides *in*: Fish and Fisheries Products Hazards and Controls Guidance, 4th edition, SeafoodHazardsGuide_Fish_FisheryProducts_March2020.pdf.
- Xia, C., Lam, J.C.W., Wu, X., Xie, Z. and Lam, P.K.S. 2012. Polychlorinated biphenyls (PCBs) in marine fishes from China: Levels, distribution and risk assessment. *Chemosphere* **89**: 944–949.
- Xing, Y., Lu, Y., Dawson, R.W., Shi, Y., Zhang, H., Wang, T., Liu, W. and Ren, H. 2005. A spatial temporal assessment of pollution from PCBs in China. *Chemosphere* **60**: 731–739.
- Zhang, H., Zhao, X., Ni, Y., Lu, X., Chen, J., Su, F., Zhao, L., Zhang, N. and Zhang, X. 2010. PCDD/Fs and PCBs in sediments of the Liaohe River, China: Levels, distribution, and possible sources. *Chemosphere* **79**: 754–762.
- Zhao, L., Hou, H., Zhou, Y.Y., Xue, N.D., Li, H.Y. and Li, F.S. 2010. Distribution and ecological risk of polychlorinated biphenyls and organochlorine pesticides in surficial sediments from Haihe River and Haihe Estuary Area, China. *Chemosphere* **78**: 1285–1293.

2.2 Organochlorine pesticides (OCPs) in the marine environment of the China coast

Peng Zhang, Ying Bian, Chuanlin Huo and Guangshui Na

National Marine Environmental Monitoring Center, Dalian, China

The historical production and use of pesticides in China are shown in Figure 2.2.1. Because organochlorine pesticides (OCPs) accounted for 80% of the total pesticides produced in China before 1982, their regulation in 1982 resulted in a sharp decrease in the production of total pesticides for five years. After that, the production of total pesticides (mostly non-OCPs) increased again.

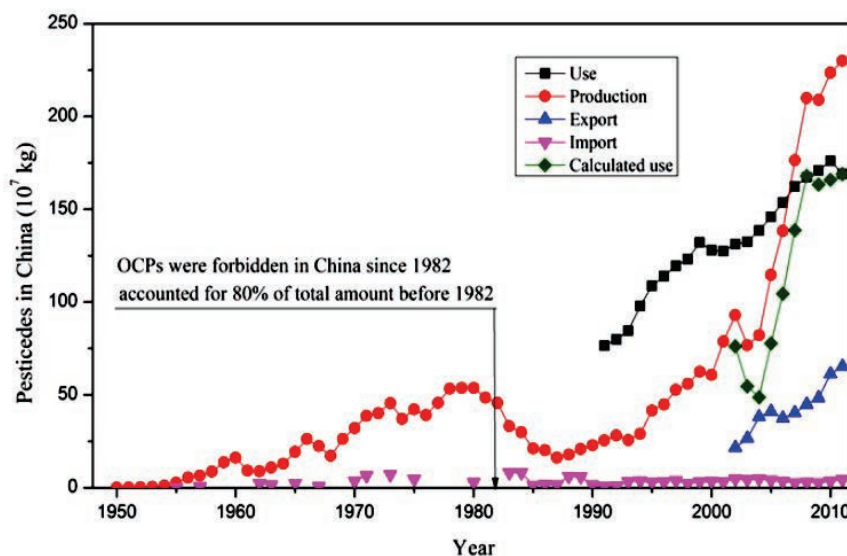


Fig. 2.2.1 Historical production, use, export and import of all pesticides in China. From Grung *et al.* (2015).

In 2003, α -hexachlorocyclohexane (α -HCH), a byproduct in the production the OCPs, had atmospheric concentrations over Chinese marginal seas that were quite uniform, with a H/L (highest/lowest) value of 9.6, which was also comparable to the atmospheric concentrations in the North Pacific Ocean and adjacent Arctic region. Furthermore, the α -HCH concentrations in 2006 declined by approximately 50 times over about two decades, which was likely related to the restricted usage of technical HCH and its relative quick degradation process in the environment. It was also observed that there was no significant difference in the spatial distribution of dichlorodiphenyltrichloroethane (DDT – another OCP) in the atmosphere, with a H/L value of 13. The results of a back trajectory (September 18 to October 17, 2006) indicate that the higher concentrations of the γ -HCH isomer and DDT compounds observed at some sampling sites in Chinese coastal waters were associated with air masses passing through Southeast China. The ratios of the α -HCH to γ -HCH isomer (α/γ -HCH ratios) range from 0.05 to 0.76, which are well below those in the technical HCH (4~7) range. The low α/γ -HCH ratio in the air implies that lindane is still in use. The ratios of *o,p'*-DDT/*p,p'*-DDT, ranging from 0.33 to 27, indicate the mixing source of technical DDT and dicofol-type DDT in the marginal seas (Lin *et al.*, 2012a).

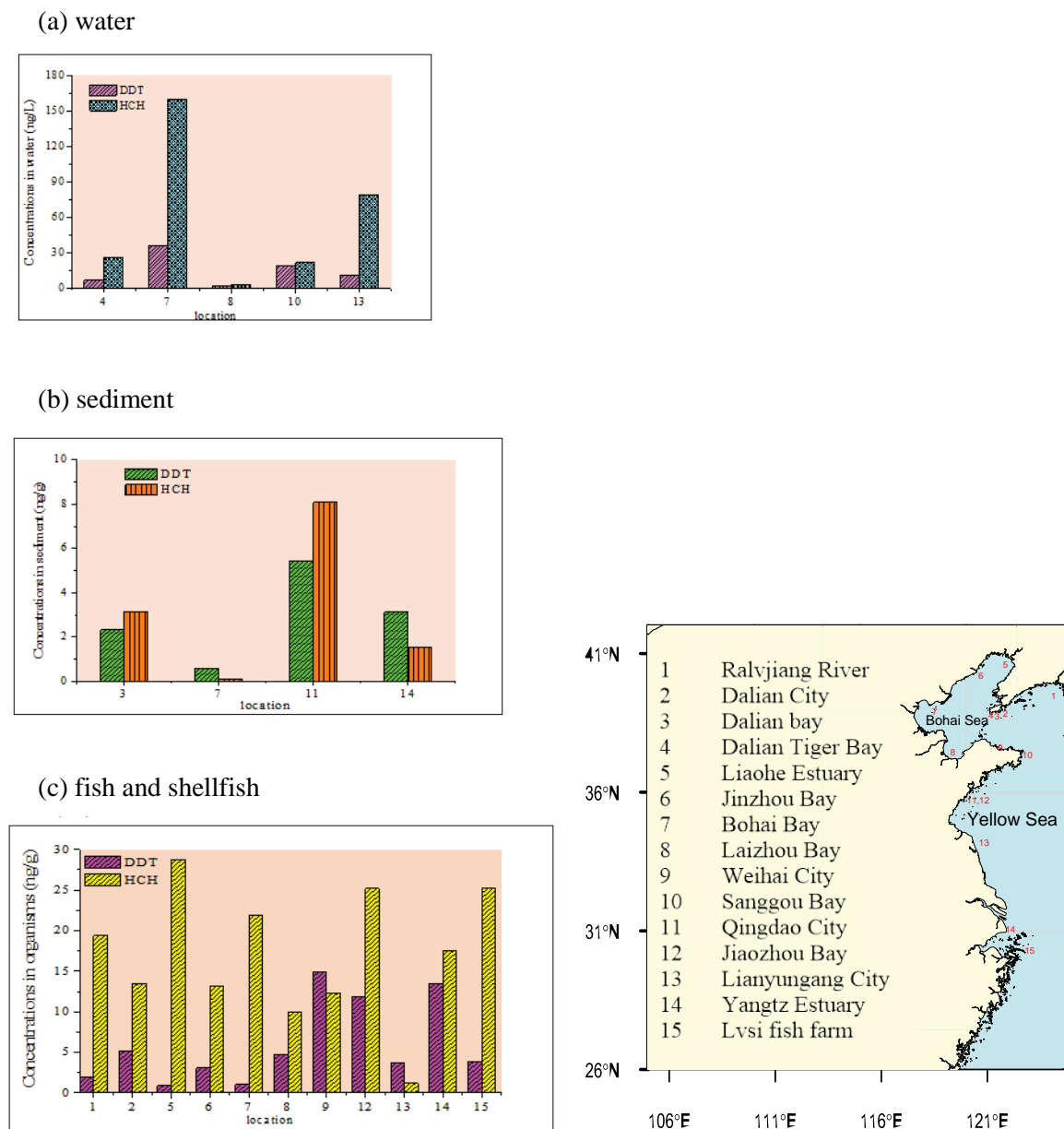


Fig. 2.2.2 Spatial distributions of organochlorine pesticides (OCPs), DDT and HCH, in the marine environment and organisms in Chinese marginal seas for (a) water, (b) sediments, and (c) fish and shellfish. Location numbers are identified in the map key.

The concentrations of DDTs and HCHs in seawater, sediment and organisms in Chinese marginal seas are illustrated in Figure 2.2.2 based on some previous researches (Xu *et al.*, 2007; Hu *et al.*, 2009; Zhao *et al.*, 2010; Lin *et al.*, 2012a,b; Zhou *et al.*, 2014). The OCP concentrations in seawater and sediment present a similar trend, *i.e.*, the HCH concentrations, are generally higher than DDTs except that DDT concentrations in sediment are higher than those of HCHs in Bohai Bay and Yangtze Estuary. In cities adjacent to the Bohai Sea, the OCP concentrations in water were relatively higher than in other areas of the Yellow Sea (Fig. 2.2.2a). The concentrations of OCPs in the sediment sampled from Qingdao were

relatively higher than other study areas (Fig. 2.2.2b). Due to huge differences in spatial distribution and the confounding influence of age, sex and diet sources of organisms, the distribution trends of OCPs in fish and shellfish (Fig. 2.2.2c) were too complex to interpret.

Generally, HCH concentrations were higher than for DDTs in the sites sampled for fish and shellfish (Fig. 2.2.2c). Shandong Province, bordering both the Bohai Sea and Yellow Sea, and Shanghai showed significantly higher concentrations of DDTs in organisms than in other areas of the Chinese marginal seas. Using the Norwegian classification system, risk levels of DDTs and lindane (HCHs in biota) were assessed by Grung *et al.* (2015). Their research revealed that the concentrations in biota from some Chinese coastal areas could be classified as “bad” or “very bad” for DDT, including Haihe Estuary and Yellow River Estuary. The risk in biota is much lower for HCH, for which the majority of the levels in biota can be classified as class I or II (*i.e.*, background or good, respectively).

References

- Grung, M., Lin, Y., Zhang, H., Steen, A.O., Huang, J., Zhang, G. and Larssen, T. 2015. Pesticide levels and environmental risk in aquatic environments in China - A review. *Environment International* **81**: 87–97.
- Hu, L., Zhang, G., Zheng, B., Qin, Y., Lin, T. and Guo, Z. 2009. Occurrence and distribution of organochlorine pesticides (OCPs) in surface sediments of the Bohai Sea, China. *Chemosphere* **77**: 663–672.
- Lin, T., Hu, L., Shi, X., Li, Y., Guo, Z. and Zhang, G. 2012a. Distribution and sources of organochlorine pesticides in sediments of the coastal East China Sea. *Marine Pollution Bulletin* **64**: 1549–1555.
- Lin, T., Li, J., Xu, Y., Liu, X., Luo, C., Chang, H., Chen, Y. and Zhang, G. 2012b. Organochlorine pesticides in seawater and the surrounding atmosphere of the marginal seas of China: Spatial distribution, sources and air-water exchange. *Science of the Total Environment* **435-436**: 244–252.
- Xu, X., Yang, H., Li, Q., Yang, B., Wang, X. and Lee, F.S. 2007. Residues of organochlorine pesticides in near shore waters of LaiZhou Bay and JiaoZhou Bay, Shandong Peninsula, China. *Chemosphere* **68**: 126–139.
- Zhao, L., Hou, H., Zhou, Y., Xue, N., Li, H. and Li, F. 2010. Distribution and ecological risk of polychlorinated biphenyls and organochlorine pesticides in surficial sediments from Haihe River and Haihe Estuary Area, China. *Chemosphere* **78**: 1285–1293.
- Zhou, S., Yang, H., Zhang, A., Li, Y.F. and Liu, W. 2014. Distribution of organochlorine pesticides in sediments from Yangtze River Estuary and the adjacent East China Sea: Implication of transport, sources and trends. *Chemosphere* **114**: 26–34.

2.3 Polybrominated diphenyl ether (PBDE) flame retardants in the marine environment of the China coast

Xindong Ma, Chuanlin Huo and Guangshui Na

National Marine Environmental Monitoring Center, Dalian, China

China is one of the largest producers and consumers of polybrominated diphenyl ethers (PBDEs) in the world. As a result, associated contamination of PBDE flame retardants in the marine environment has attracted widespread attention (Mai *et al.*, 2005). Since 2000, extensive investigations on the occurrence and distribution of PBDEs in the coastal environment have been conducted in South and East China (Mai *et al.*, 2005; Chen *et al.*, 2013). For example, high concentrations of PBDEs have been found in the Pearl River Delta and several e-waste recycling areas. In this report, we survey the present status of contamination with PBDEs in different marine matrices, *e.g.*, seawater, sediment and organisms.

PBDEs in seawater

The concentrations of PBDEs in matrices collected from the coastal waters of China show a typical regional pattern (Table 2.3.1). Results indicate that the concentrations of PBDEs in seawater could be affected by point sources. Because a lot of electronic manufacturing industries and e-waste dismantling areas exist on the Chinese coast, rainfall and surface runoff are likely to take atmospheric particulates and PBDE-contaminated soil into the coastal environment.

Table 2.3.1 Total PBDE concentrations in abiotic and biotic matrices collected from the coastal areas of China.

Matrix	Research area	Result	Congener number	Reference
Seawater	Lingding Bay	Σ PBDEs(mean): 5.0 ng/L	8	Luo <i>et al.</i> , 2008
	Pearl River Delta	Σ PBDEs(dissolved): 31.1~118.7 pg/L	17	Guan <i>et al.</i> , 2007
	Hong Kong	Σ PBDEs(dissolved): 31.1~118.7 pg/L	8	Wurl <i>et al.</i> , 2006
	Hong Kong	Σ PBDEs(particulate): 25.7~32.5 pg/L		
Sediment	Bohai Sea	Σ PBDEs: 2.5 ng/g (dw*)	14	Wang <i>et al.</i> , 2009
	Yangtze River Delta	Σ PBDEs: 13.6 ng/g (dw)	11	Chen <i>et al.</i> , 2013
	Laizhou Bay	Σ PBDEs: 240 ng/g (dw)	8	Jin <i>et al.</i> , 2008
	Haihe Estuary	Σ PBDEs: 60~2100 ng/g (dw)	27	Zhao <i>et al.</i> , 2011
	Liaohu Estuary	Σ PBDEs: 130~1980 ng/g (dw)	27	Zhao <i>et al.</i> , 2011
Organisms	Bohai Sea	Σ PBDEs: 0.68 ng/g (dw)	14	Wang <i>et al.</i> , 2009
	Laizhou Bay	Σ PBDEs: 457.2 ng/g (dw)	8	Jin <i>et al.</i> , 2009
	Liaodong Bay	Σ PBDEs: 1.3~8.8 ng/g (dw)	15	Ma <i>et al.</i> , 2013

*dw = dry weight

PBDEs in sediment

Compared with other countries, PBDEs concentrations in sediments of most coastal areas of China are relatively low (Table 2.3.1). Similar to the concentration in seawater, the spatial distribution of PBDEs also present a typical regional characteristic. It should be noted that while recent research has found that PBDE concentrations in surficial sediments in the Pearl River Delta display a decreasing trend, alternative flame retardants, including decabromodiphenyl ethane (DBDPE) show an increasing trend (Chen *et al.*, 2013).

PBDEs in organisms

PBDE residual levels in organisms from most coastal areas of China are also relatively low compared with those of other countries. However, the concentrations of PBDEs from the polluted areas are comparable to or even higher than those of developed regions (Table 2.3.1). According to a report by Shi *et al.* (2009), the average concentration of PBDEs in fish samples collected from the electronic waste recycling plant in Qingyuan (Guangdong Province) was 153.0 ng/g (lipid weight), and the mean value in waterfowl was up to 1165.2 ng/g (lw). Because of the high potential of bioaccumulation and biomagnification, the risk management of PBDEs in terms of the ecosystem and human health, especially in heavily polluted areas, should not be ignored.

References

- Chen, S.J., Feng, A.H., He, M.J., Chen, M.Y. and Mai, B.X. 2013. Current levels and composition profiles of PBDEs and alternative flame retardants in surface sediments from the Pearl River Delta, southern China: Comparison with historical data. *Science of the Total Environment* **444**: 205–211.
- Guan, Y., Wang, J., Ni, H., Luo, X., Mai, B. and Zeng, E. Y. 2007. Riverine inputs of polybrominated diphenyl ethers from the Pearl River Delta (China) to the coastal ocean. *Environmental Science and Technology* **41**: 6007–6013.
- Jin, J., Liu, W.Z., Wang, Y. and Tang, X.Y. 2008. Levels and distribution of polybrominated diphenyl ethers in plant, shellfish and sediment samples from Laizhou Bay in China. *Chemosphere* **71**: 1043–1050.
- Jin, J., Wang, Y., Yang, C., Hu, J., Liu, W., Cui, J. and Tang, X. 2009. Polybrominated diphenyl ethers in the serum and breast milk of the resident population from production area, China. *Environment International* **35**: 1048–1052.
- Luo, X.J., Yu, M., Mai, B.X. and Chen, S.J. 2008. Distribution and partition of polybrominated diphenyl ethers (PBDEs) in water of the Zhujiang River Estuary. *Chinese Science Bulletin* **53**: 493–500.
- Ma, X., Zhang, H., Yao, Z., Zhao, X., Wang, L., Wang, Z., Chen, J. and Chen, J. 2013. Bioaccumulation and trophic transfer of polybrominated diphenyl ethers (PBDEs) in a marine food web from Liaodong Bay, North China. *Marine Pollution Bulletin* **74**: 110–115.
- Mai, B.X., Chen, S.J., Luo, X.J., Chen, L.G., Yang, Q.S., Sheng, G.Y., Peng, P.A., Fu, J.M. and Zeng, E.Y. 2005. Distribution of polybrominated diphenyl ethers in sediments of the Pearl River Delta and adjacent South China Sea. *Environmental Science and Technology* **39**: 3521–3527.
- Shi, T., Chen, S.J., Luo X.J., Zhang, X.L., Tang, C.M., Luo, Y., Ma, Y.J., Wu, J.P., Peng, X.Z. and Mai, B.X. 2009. Occurrence of brominated flame retardants other than polybrominated diphenyl ethers in environmental and biota samples from southern China. *Chemosphere* **74**: 910–916.

- Wang, Z., Ma, X., Lin, Z., Na, G. and Yao, Z. 2009. Congener specific distributions of polybrominated diphenyl ethers (PBDEs) in sediment and mussel (*Mytilus edulis*) of the Bo Sea, China. *Chemosphere* **74**: 896–901.
- Wurl, O., Lam, P.K. and Obbard, J.P. 2006. Occurrence and distribution of polybrominated diphenyl ethers (PBDEs) in the dissolved and suspended phases of the sea–surface microlayer and seawater in Hong Kong, China. *Chemosphere* **65**: 1660–1666.
- Zhao, G.F., Zhou, H.D., Du, M., Yang, L., Li, K., Wu, Z. Y. and Gao, J.J. 2011. PBDEs in sediments from 14 principal tributaries of Haihe River and their potential risk. *Environmental Sciences* **32**: 2069–2073.

2.4 Perfluorinated compounds (PFCs) in the marine environment of the China coast

Yao Yao, Ruijing Li, Chuanlin Huo and Guangshui Na

National Marine Environmental Monitoring Center, Dalian, China

Recently, perfluorinated compounds (PFCs) have received worldwide attention because they are frequently detected in various environmental matrices, including coastal areas of China. In accordance with the screening criteria for persistent organic pollutants (POPs) under the Stockholm Convention, perfluorooctane sulfonate (PFOS) and its salts together with its precursor, perfluorooctane sulfonyl fluoride (PFOSF), were added to Annex B of the Convention in May of 2009, calling for restricted use worldwide. Among these compounds, perfluorooctanoic acid (PFOA) and PFOS are the most representative and commonly found in environmental matrices, since they have been used for many years in commercial production and are the terminal degradation products of several PFCs (Cao *et al.*, 2012). In this report, we survey the present status of contamination with PFCs in the marine environment of China's coast.

The concentrations of PFCs in the seawater of the coastal regions in China are summarized in Figure 2.4.1. PFOS and PFOA are the dominant PFCs found in these waters. Direct sewage emissions and rivers discharging into the sea might be the primary sources of PFCs in surface seawater. The contamination of PFCs in the Bohai Sea is more serious than other areas in China because of the presence of large chemical plants and rivers exhibiting a high content of urban wastewater input (Chen *et al.*, 2011). Compared with other countries, seawater concentrations of PFCs in most coastal areas of China are relatively low.

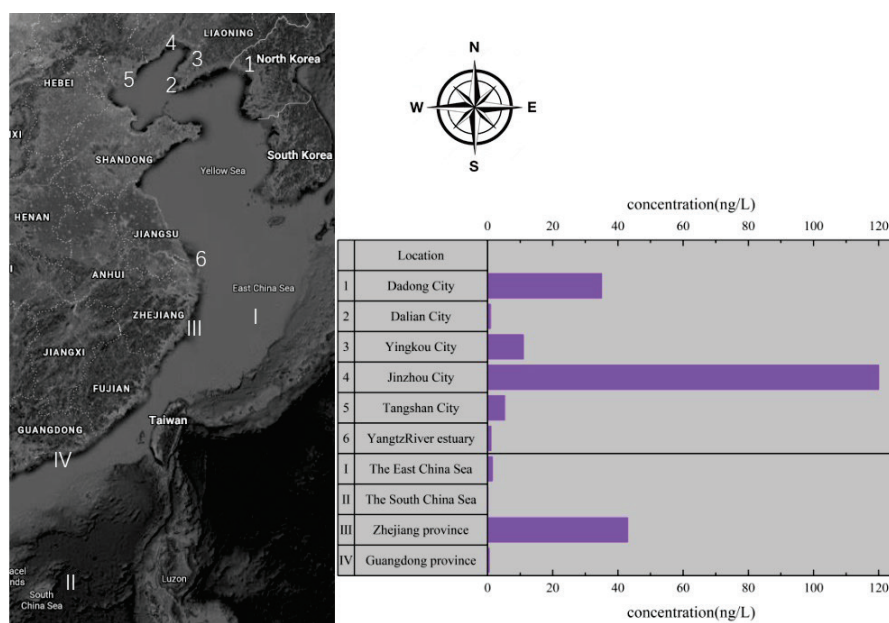


Fig. 2.4.1 Location and concentration of perfluorinated compounds (PFCs) in the seawater of the coastal regions of China (Chen *et al.*, 2011; Cai *et al.*, 2012; Lu *et al.*, 2015; Zheng *et al.*, 2017).

It has been reported that sediments and the deep ocean are two important sinks of PFASs in the environment (Konstantinos *et al.*, 2006). The concentration of perfluorinated compounds in the sediment of the coastal regions of China is summarized in Table 2.4.1. It was reported that the Σ_9 PFASs in sediments in the different sampling areas were ranked as Bohai Sea > East China Sea > Yellow Sea, based on both the mean and median, with an increasing trend from the lower to the upper layers (Gao *et al.*, 2014).

Table 2.4.1 Concentration of perfluorinated compounds (PFCs) in the sediment of Chinese marginal seas.

Location	Range (ng/g dw)	Predominant compound	Reference
Laizhou Bay	0.76	PFOA	Zhao <i>et al.</i> , 2013
South Bohai coastal watersheds	0.218–1.583	PFOS, PFBS	Zhu <i>et al.</i> , 2014
Bohai Sea	0.06–2.98	PFOA	Gao <i>et al.</i> , 2014
Yellow Sea	n.d.–2.76	PFOA	Gao <i>et al.</i> , 2014
Pearl River Delta	0.15–3.11	PFOS, PFBA	Zhao <i>et al.</i> , 2014
East China Sea	0.03–1.77	PFOA	Gao <i>et al.</i> , 2014
East China Sea	n.d.–34.8	PFOS, PFHpA, PFOA	Yan <i>et al.</i> , 2015

PFOA = perfluorooctanoic acid, PFOS = perfluorooctane sulfonate, PFBS = Perfluorobutane sulfonate, PFBA = pentafluorobenzoic acid, PFHpA = perfluoroheptanoic acid

PFCs have protein-binding, lipophilic and hydrophobic characteristics, which determine their bioaccumulative nature and tendency to accumulate in the liver of animals (Ju, 2010). The mean concentration of total PFCs in marine organisms appears to have the following trend: Bohai Sea < Yellow Sea < East China Sea (Pan *et al.*, 2014). Moreover, a negative correlation between Σ PFCs and trophic level was found for mollusks in the Bohai Sea region (Pan *et al.*, 2010).

References

- Cai, M.H., Zhao, Z., Yang, H.Z., Yin, Z.G., Hong, Q.Q., Sturm, R., Ebinghaus, R., Ahrens L., Cai, M.G., He, J.F. and Xie, Z.Y. 2012. Spatial distribution of per- and polyfluoroalkyl compounds in coastal waters from the East to South China Sea. *Environmental Pollution* **161**: 162–169.
- Cao, Y., Zhang, Y.H. and Lei, C.W. 2012. Environmental pollution and ecological toxicity of perfluorinated chemicals: A review. *Journal of Environment and Health* **29**: 561–567.
- Chen, C.L., Wang, T.Y., Khim, J.M., Luo, W., Jiao, W.T., Lu, Y.L., Naile, J.E., Hu, W.Y., Zhang, X., Geng, J., Bi, C.C., Li, J. and Giesy, J.P. 2011. Perfluorinated compounds in water and sediment from coastal regions of the northern Bohai Sea, China. *Chemistry and Ecology* **27**:165–176.
- Gao, Y., Fu, J., Zeng, L., Li, A., Li, H., Zhu, N., Liu, R., Liu, A., Wang, Y. and Jiang, G. 2014. Occurrence and fate of perfluoroalkyl substances in marine sediments from the Chinese Bohai Sea, Yellow Sea, and East China Sea. *Environmental Pollution* **194**: 60–68.
- Ju, X.D. 2010. Progress in research on marine environmental pollution of perfluorinated chemicals. *Marine Science* **34**: 93–99. (in Chinese)

- Konstantinos, P., Cousins, I.T., Buck, R.C. and Korzeniowski, S.H. 2006. Sources, fate and transport of perfluorocarboxylates. *Environmental Science and Technology* **40**: 32–44.
- Lu, Z.B., Song, L.N., Zhao, Z., Ma, Y.X., Wang, J., Yang, H.Z., Ma, H.M., Cai, M.H., Codling, G., Ebinghaus, R., Xie, Z.Y. and Giesye, J.P. 2015. Occurrence and trends in concentrations of perfluoroalkyl substances (PFASs) in surface waters of eastern China. *Chemosphere* **119**: 820–827.
- Pan, C.G., Zhao, J.L., Liu, Y.S, Zhang, Q.Q., Chen, Z.F., Lai, H.J., Peng, F.J., Liu, S.S. and Ying, G.G. 2014. Bioaccumulation and risk assessment of per- and polyfluoroalkyl substances in wild freshwater fish from rivers in the Pearl River Delta region, South China. *Ecotoxicology and Environmental Safety* **107**: 192–199.
- Pan, Y.Y., Shi, Y.L., Wang, Y.W., Cai, Y.Q. and Jiang, G.B. 2010. Investigation of perfluorinated compounds (pfc) in mollusks from coastal waters in the Bohai Sea of China. *Journal of Environmental Monitoring* **12**: 508–513.
- Yan, H., Zhang, C., Zhou, Q. and Yang, S. 2015. Occurrence of perfluorinated alkyl substances in sediment from estuarine and coastal areas of the East China Sea. *Environmental Science and Pollution Research* **22**: 1662–1669.
- Zhao, Y.G., Wan, H.T., Wong, M.H. and Wong, C.K.C. 2014. Partitioning behavior of perfluorinated compounds between sediment and biota in the Pearl River delta of south China. *Marine Pollution Bulletin* **83**: 148–154.
- Zhao, Z., Tang, J., Xie, Z., Chen, Y., Pan, X., Zhong, G., Sturm, R., Zhang, G. and Ebinghaus, R. 2013. Perfluoroalkyl acids (pfaas) in riverine and coastal sediments of Laizhou Bay, North China. *Science of the Total Environment* **447**: 415–423.
- Zheng, H.Y., Wang, F., Zhao, Z., Ma, Y.X., Yang, H.Z., Lu, Z.B., Cai, M.G. and Cai, M.H. 2017. Distribution profiles of per- and poly fluoroalkyl substances (PFASs) and their re-regulation by ocean currents in the East and South China Sea. *Marine Pollution Bulletin* **125**: 481–486.
- Zhu, Z., Wang, T., Wang, P., Lu, Y. and Giesy, J.P. 2014. Perfluoroalkyl and polyfluoroalkyl substances in sediments from south Bohai coastal watersheds, China. *Marine Pollution Bulletin* **85**: 619–627.

2.5 Polycyclic aromatic hydrocarbons (PAHs) in the marine environment of the China coast

Zhen Wang, Chuanlin Huo and Guangshui Na

National Marine Environmental Monitoring Center, Dalian, China

Polycyclic aromatic hydrocarbons (PAHs) are a class of hydrophobic semi-volatile organic compounds (SOCs) in the environment, and originate mainly from incomplete combustion of fossil fuels and biomass. With the increase of fossil fuels consumption in China, PAH concentrations in the coastal environment are also increasing.

PAHs in seawater

Table 2.5.1 lists the concentrations of PAHs in seawater in regions of China's marginal seas. The difference in concentrations among different regions is significant. In general, PAH concentrations in the Bohai Sea are comparable to those of the Yellow Sea and East China Sea while the levels of PAHs in the estuary are higher than those in the marginal seas. Overall, PAHs levels in the coastal waters of North China are comparable or lower to those of other coastal areas in the world.

Table 2.5.1 Concentrations of polycyclic aromatic hydrocarbons (PAHs) in Chinese coastal waters (ng/L).

Location	Range	Average	Number of PAHs congeners	Reference
South Yellow Sea	37.8 ~ 233.4	109.4	16	Han <i>et al.</i> , 2009
Northwest Bohai Sea	14.1 ~ 226	108	16	Zhang <i>et al.</i> , 2013
Qingdao coast	8.23 ~ 272.02	–	16	Li <i>et al.</i> , 2012
Yellow Sea–East China Sea coast	39.24 ~ 418.03	212.37	16	Song <i>et al.</i> , 2014
Yellow River estuary	473.7 ~ 1190.1	728.9	25	Lang <i>et al.</i> , 2008
Yangtze River estuary	478 ~ 6273	–	16	Ou <i>et al.</i> , 2007

PAHs in sediment

There is no significant difference among PAHs concentrations in sediments of various coastal areas, as presented in Table 2.5.2. However, a significant difference was observed between sampling sites. For example, PAH concentrations in sediments of the Bohai Sea were in the range of 24.7 to 2079.4 ng/g dry weight, with the highest concentration nearly two orders of magnitude higher than the lowest level. In other areas, the observed concentration difference was around one order of magnitude. The results indicate that there is a significant positive correlation between PAH pollution in sediments and industrial development in coastal areas, and imply that PAH pollution in sediments are influenced seriously by pollution sources in coastal areas especially.

Table 2.5.2 Concentrations of PAHs in coastal sediments of China (ng/g, dry weight).

Area	Location	Range	Average	References
Bohai Sea	Liaodong Bay	31.2 ~ 652.9	143.4	Lin <i>et al.</i> , 2005
	Qinhuangdao coast	202.2 ~ 2079.4	1081.9	Lin <i>et al.</i> , 2005
	Bohai Bay	24.7 ~ 34.6	28	Lin <i>et al.</i> , 2005
	Laizhou Bay	24.7 ~ 1392	55	Lin <i>et al.</i> , 2005
Yellow Sea	Dalian Bay	327 ~ 3558.9	1152.1	Liu <i>et al.</i> , 2001
	Jiaozhou Bay	82 ~ 4562	–	Yang <i>et al.</i> , 2003
	Rizhao coast	76.2 ~ 27512.2	2622.7	Lang <i>et al.</i> , 2009
	North Yellow Sea	222.1 ~ 776.3	–	Li <i>et al.</i> , 2002
	South Yellow Sea	90.4 ~ 732.65	299.57	Han <i>et al.</i> , 2009
East China Sea	Yangtze River estuary	698 ~ 7907	357.7	Chen <i>et al.</i> , 2011
	Hangzhou Bay	45.8 ~ 849.9	263.7	Chen <i>et al.</i> , 2011

Generally, PAHs in the sediment samples under investigation were mainly from the combustion of biomass, coal, and petroleum, a strong indication of anthropogenic impacts (Liu *et al.*, 2007; Liu *et al.*, 2012). The spatial variability of diagnostic ratios suggests that different PAH compounds underwent different degradation pathways from source to sediments, and hydrodynamic transport was not the dominant input mechanism. For example, PAHs in the Yellow Sea sediments were contributed largely from atmospheric input, especially via particle deposition (Ohkouchi *et al.*, 1999; Li *et al.*, 2002). Furthermore, the results of vertical distribution in core sediments show that, in general, PAH levels increased from the bottom to the surface, indicating the obvious influence of human activities on the vertical distribution of PAHs. Most studies suggest that the concentrations of PAHs in core sediments began to increase gradually from 1960s. However, the different core profiles of PAHs among different sites indicate that regional characteristics (*e.g.*, fuel consumption intensity, fuel types, economic development) also influence the PAH levels and distribution (Xu *et al.*, 2006).

References

- Chen, Z.M., Gao, X.J., Song, Z.G. *et al.* 2011. Distribution of OCPs in surface intertidal sediments of Hangzhou Bay. *China Environmental Science* **31**: 321–327. (in Chinese)
- Han, B., Jiang, F.H., Li, P.C., Zhang, X.Q. and Wang, X.R. 2009. Distribution and origin of polycyclic aromatic hydrocarbons in the sea water, pore water and sediment of the central area in the South Yellow Sea. *Advances in Marine Science* **27**: 233–242. (in Chinese)
- Lang, Y.H., Jia, Y.G., Liu, Z.F., Gao, Z.H. and Wang, X. 2008. Seasonal distribution characteristics and sources of polycyclic aromatic hydrocarbons (PAHs) in water samples from the Yellow River estuary. *Periodical of Ocean University of China* **38**: 640–646. (in Chinese)
- Lang, Y.H., Xue, L.D., Liu, A.X., *et al.*, 2009. Identification and source apportionment of polycyclic aromatic hydrocarbons (PAHs) in coastal surface sediments from Rizhao offshore area. *Periodical of Ocean University of China* **39**: 535–542. (in Chinese)
- Li, B., Wu, Y. and Zhang, J. 2002. Distribution and source of polycyclic aromatic hydrocarbons (PAHs) in surface sediment of the northern Yellow Sea. *China Environmental Science* **22**: 429–432. (in Chinese)

- Li, X.G., Deng, W., Zhou, X., Tang, X.L., Guo, X.Y. and Wang, Y. 2012. Distribution of PAHs in surface seawater of Qingdao preliminary apportionment. *China Environmental Science* **33**: 741–745. (in Chinese)
- Lin, X.M., Liu, W.X., Chen, J.L., Xu, S.S. and Tao, P. 2005. Distribution and ecological risk assessment of polycyclic aromatic hydrocarbons in surface sediments from Bohai Sea, China. *Acta Scientiae Circumstantiae* **25**: 70–75. (in Chinese)
- Liu, J.P., Xu, K.H., Li, A.C., Milliman, J., Velozzi, D.M., Xiao, S.B. and Yang, Z.S. 2007. Flux and fate of Yangtze River sediment delivered to the East China Sea. *Geomorphology* **85**: 208–224.
- Liu, L.Y., Wang, J.Z., Wei, G.L., Guan, Y.F. and Zeng, E.Y. 2012. Polycyclic aromatic hydrocarbons (PAHs) in continental shelf sediment of China: Implications for anthropogenic influences on coastal marine environment. *Environmental Pollution* **167**: 155–162.
- Liu, X.M., Xu, X.R., Zhang, X.T., Zhou, C.G. and Li, H. 2001. A preliminary study on PAHs in the surface sediment samples from Dalian Bay. *Acta Scientiae Circumstantiae* **21**: 507–509. (in Chinese)
- Ohkouchi, N., Kawamura, K. and Kawahata, H. 1999. Distributions of three- to seven-ring polynuclear aromatic hydrocarbons on the deep sea floor in the central Pacific. *Environmental Science and Technology* **33**: 3086–3090.
- Ou, D.N. 2007. Multi-media distribution and sources identification of polycyclic aromatic hydrocarbons (PAHs) in the Yangtze estuarine and coastal ecosystem [D]. East China Normal University. (in Chinese)
- Song, Y.M. 2014. Ecological assessment of polycyclic aromatic hydrocarbons in the seawater of ship routes [D]. Dalian Maritime University. (in Chinese)
- Xu, S.S., Liu, W.X. and Tao, S. 2006. Emission of polycyclic aromatic hydrocarbons in China. *Environmental Science and Technology* **40**: 702–708.
- Yang, Y.L., Mai, B.X., Pan, J., Yin, X.C. and Li, F.Y. 2003. Distribution and sources of polycyclic aromatic hydrocarbon in sediments of Jiaozhou Bay. *Marine Environmental Science* **22**: 38–43. (in Chinese)
- Zhang, Y.D., Tian, S.Y., Liu, X.B., Li, G.F., Zhu, W. and Jia, R. 2013. Distribution, source and ecological risk assessment of polycyclic aromatic hydrocarbons in surface water from the northwest Bohai Sea, China. *Oceanologia et Limnologia Sinica* **44**: 255–261. (in Chinese)

3 Japan

3.1 Temporal trends of PCB concentrations in mussels around the Japanese coast

Kazuhiko Mochida and Hiroyuki Tanaka

National Research Institute of Fisheries and Environment of Inland Sea, Fisheries Research and Education Agency, Hatsukaichi, Hiroshima, Japan

Polychlorinated biphenyls (PCBs) are ubiquitous persistent organic pollutants (POPs) in the marine environment. They have been used commercially in various materials and equipment such as electrical appliances, heat-transfer fluids, carbon-copy papers, and paints (Maruyama *et al.*, 1983). Although production, use, and import of PCBs has been prohibited since 1975 in Japan, ecological health impacts are still of concern because of their low degradability, toxicity, and high bioaccumulation in marine organisms (Ministry of the Environment, 2009). In this report, we survey the present status of concentrations of PCBs in blue mussel (*Mytilus galloprovincialis*) inhabiting the coastal areas of Japan.

PCBs in mussels inhabiting the coastal area

The Ministry of the Environment of Japan carried out monitoring surveys for PCBs in mussels from 2008 to 2014 in five different sampling sites in Japan, as shown in Figure 3.1.1 (<http://www.env.go.jp/chemi/kurohon/>). The mean \pm standard error (SE) and median of the concentration in mussels are 18 ± 9.5 ng/g wet weight (ww) and 15 ng/g ww, respectively. While a temporal decline in PCB concentrations in mussels is, in general, found in most sampling sites from 2008 to 2014, a prominent downward trend is not observed for the concentrations observed in mussels from some sites (sites A and B). At site (F), PCB concentrations in the hard-shelled mussel (*Mytilus caruscus*) were also investigated in 2008 and 2009 and the concentration in the mussel ($n = 6$, pooled) was 9.5 ng/g ww.

Because the regulation value in seafood in Japan is 0.5 to 3.0 mg/kg wet weight (https://www.mhlw.go.jp/web/t_doc?dataId=00ta5726&dataType=1&pageNo=1, in Japanese), the detected PCB concentrations in mussels are considered to be quite low.

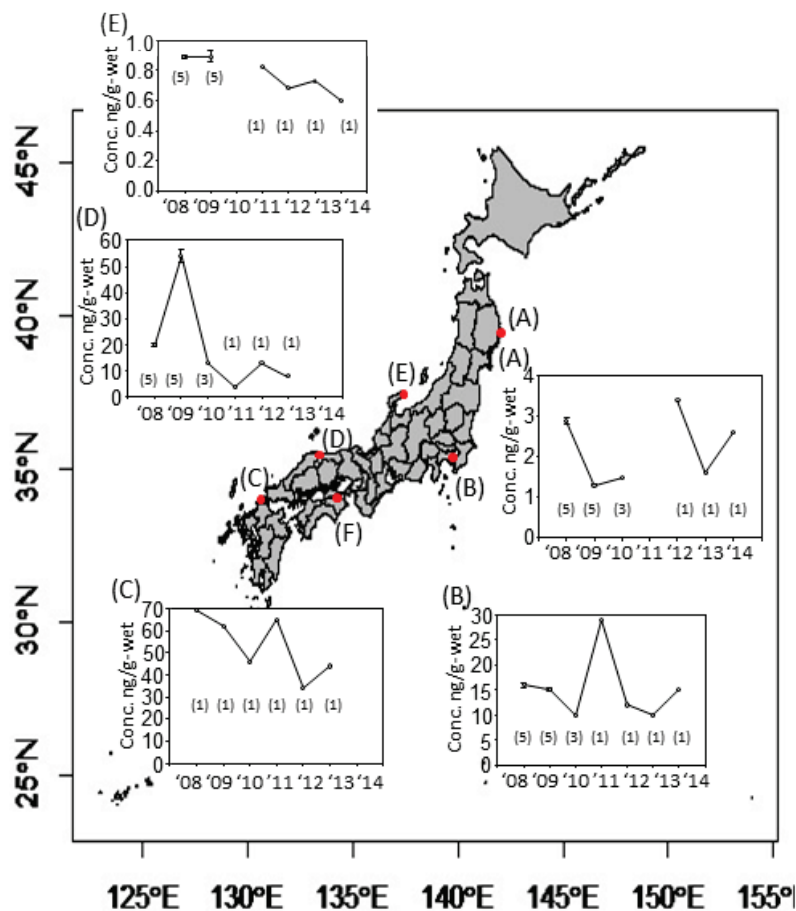


Fig. 3.1.1 Sampling sites and temporal changes in concentrations of polychlorinated biphenyls (PCBs) in blue mussel (*Mytilus galloprovincialis*) at sites around Japan. The sample at site (C) in '09 is for the purplish bifurcate mussel (*Septifer virgatus*). Data are expressed as mean. Error bars in (E) for '08 and '09 indicate standard error. Number in parentheses inside graphs shows the number of samples. Site (F) on map indicates hard-shelled mussel (*Mytilus coruscus*) sampled in '08 and '09.

References

- Maruyama, K., Sahrul, M., Tanabe, S. and Tatsukawa, R. 1983. Polychlorinated biphenyl pollution from shipbuilding in Nagasaki Bay, Japan. *Ecotoxicology and Environmental Safety* **7**: 514–520.
- Ministry of the Environment. 2009. Present status of marine pollution in the sea around Japan. http://www.env.go.jp/water/kaiyo/monitoring/status_report/.

3.2 Temporal and spatial trends of PCBs in coastal-marine sediments and seawater in Japan

Kazuhiko Mochida and Hiroyuki Tanaka

National Research Institute of Fisheries and Environment of Inland Sea, Fisheries Research and Education Agency, Hatsukaichi, Hiroshima, Japan

Polychlorinated biphenyls (PCBs) are ubiquitous persistent organic pollutants (POPs) in the marine environment. They have been used commercially in various materials and equipment, such as electrical appliances, heat-transfer fluids, carbon-copy papers, and paints (Maruyama *et al.*, 1983). In Japan, PCBs were designated as a Class I Specified Chemical Substance under the Chemical Substances Control Law in June 1974¹ since they are persistent, highly bioaccumulative, and chronically toxic in living organisms. In this report, we survey the present status of contamination with PCBs in coastal marine sediments and seawater of Japan.

PCBs in sediments of bays and offshore regions

The Ministry of the Environment of Japan carried out monitoring surveys for PCBs in both seawater and sediments from 2008 to 2013 in five different regions, including coastal and offshore areas (Fig. 3.2.1, <http://www.env.go.jp/water/kaiyo/monitoring.html>). While a low concentration level of

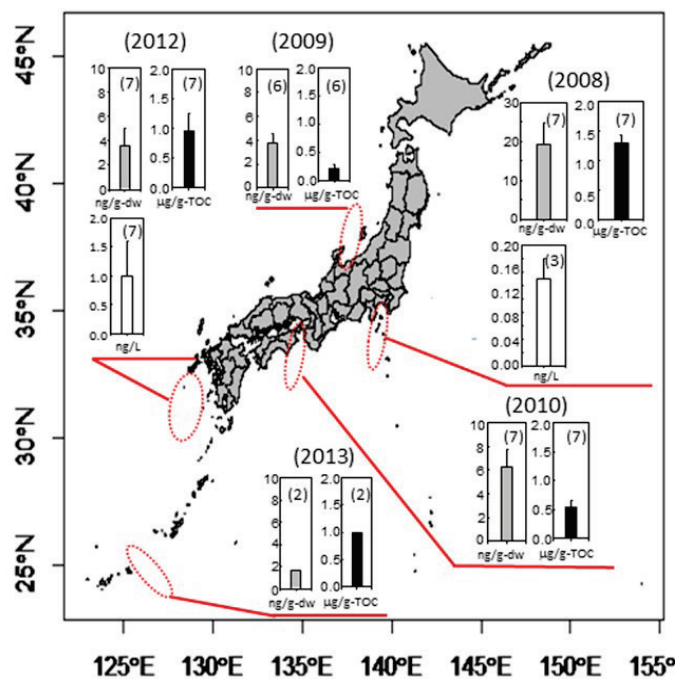


Fig. 3.2.1 Geographical distribution of PCBs in sediment (shaded column, ng/g-dw; closed column, $\mu\text{g/g-TOC}$) and seawater (open column, ng/L) of the marine environment around Japan. Data are expressed as mean \pm standard error in sediment and/or seawater samples except for the 2013 sample. Number in parentheses inside graphs represents number of samples.

¹ http://www.meti.go.jp/policy/chemical_management/kasinhou/about/about_index.html

PCBs in sediments remains detected in the offshore area, there is a tendency showing that concentrations of PCBs in the sediment of inner bays/coastal areas are higher. The highest concentrations in the sediment and seawater were 42.5 ng/g dry weight (dw) in the 2008 samples and 4.5 ng/L in the 2012 samples, respectively. In Japan, sediment quality and water quality criteria are 10 mg/kg dw and 0.0005 mg/L, respectively (<http://www.env.go.jp/hourei/05/000179.html> for sediment; <http://www.env.go.jp/kijun/index.html> for water). Compared to these values, the detected levels are quite low, even in metropolitan areas such as Tokyo Bay and Osaka Bay. Thus, the ecological risk imposed by PCBs is considered to be low.

PCBs in sediments of river mouths and coastal regions

The Ministry of the Environment also carried out monitoring surveys for PCBs in coastal–marine sediments from 2008 to 2014 in the 43 different river mouth and coastal regions in Japan (Fig. 3.2.2A, <http://www.env.go.jp/chemi/kurohon/>). In this survey, the mean concentrations of PCBs in sediments show a slight temporal decrease from 2008 to 2014 (*i.e.*, from 81 ± 18 ng/g dw to 52 ± 12 ng/g dw; Fig. 3.2.2B). The highest concentration detected in this survey was 950 ng/g dw from a metropolitan area in 2011. Other than these surveys, the PCB concentration levels in sediment samples from coastal areas near chemical manufacture facilities and a waste dumping site in the northern Shikoku Island was also reported (Kawano *et al.*, 2016). This survey was carried out from 2012 to 2013, and the concentration ranged from 0.085 to 220 ng/g dw (mean, 28 ng/g dw; median, 6.5 ng/g dw).

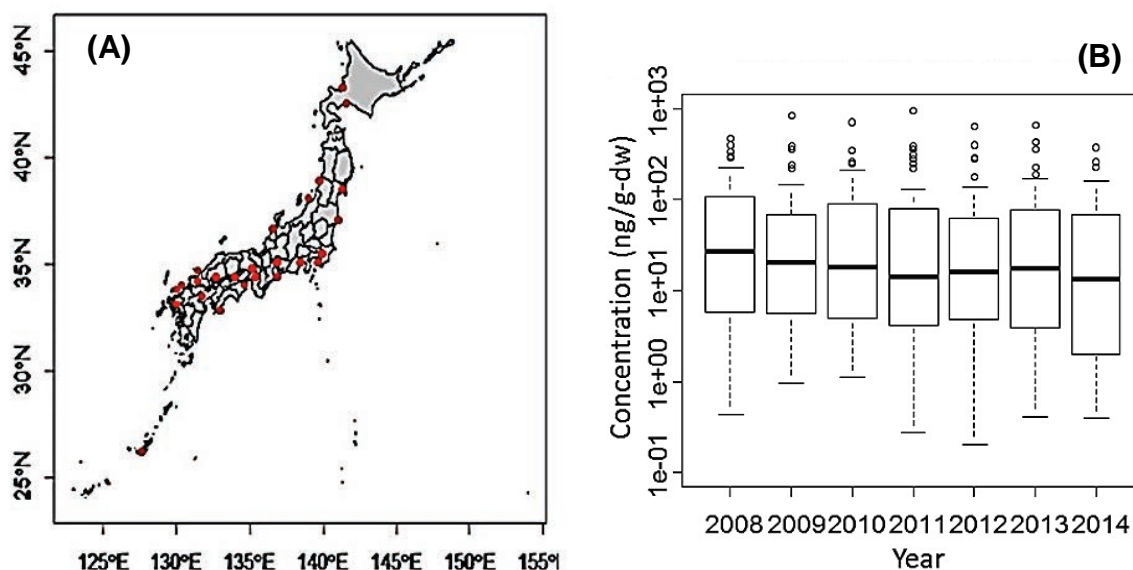


Fig. 3.2.2 (A) Sampling stations for PCBs in sediments of river mouth and coastal regions around Japan (red circles) and (B) boxplot for temporal changes in concentrations (ng/g dw) of PCBs for all the stations.

Given the sediment quality criteria, as mentioned above, these data also support the notion that the ecological risk level of PCBs is considered to be low in coastal and offshore areas of Japan. Due to the enactment of the Law Concerning Special Measures against PCB Waste (https://elaws.e-go.jp/search/elawsSearch/elaws_search/lsg0500/detail?lawId=413AC0000000065), the discharge

of PCBs into the environment may decrease. However, considering their low degradability and high bioaccumulation, continuous monitoring for PCB concentrations will be required in the future.

References

- Kawano, M., Nishiyama, T., Matsuda, M., Morita, M. and Takahashi, S. 2016. Dechlorane plus and dechlorane (Mirex) in surface sediment of the northern Shikoku Island, Japan. *Journal of Environmental Chemistry* **26**: 67–76. (in Japanese with English abstract)
- Maruyama, K., Sahrul, M., Tanabe, S. and Tatsukawa, R. 1983. Polychlorinated biphenyl pollution from shipbuilding in Nagasaki Bay, Japan. *Ecotoxicology and Environmental Safety* **7**: 514–520.

3.3 Current status of marine pollution by antifouling biocides in Japan

Kazuhiko Mochida

National Research Institute of Fisheries and Environment of Inland Sea, Fisheries Research and Education Agency, Hatsukaichi, Hiroshima, Japan

Antifouling biocides have been used mainly for ship hulls and fishing nets to prevent or reduce biofouling by colonizing marine organisms. In the 1970s, most of the ships in the world carried organotin (OT) based antifouling compounds, such as tributyltin (TBT) and triphenyltin (TPT), on their hulls. However, due to the severity of TBT-induced impacts on marine ecosystem health, Japan banned all applications of TBT usage in Japanese shipyards in 1991 prior to the action of the International Maritime Organization. Since the ban on the use of the OT compounds as antifouling paint biocides, many of the OT-free biocides have been registered on the Japanese market and used as biocide components of new antifouling paint products (Okamura and Mieno, 2006).

In this report, we survey the present status of contamination with the OT-based antifouling biocides that have already been out of use officially, and with a couple of the OT-free antifouling biocides in marine environment of Japan.

OT-based antifouling biocides

The Ministry of the Environment of Japan carried out yearly monitoring surveys for OTs in both seawater and sediment from 2008 to 2013 in five different regions (coastal to offshore) in Japan (Fig. 3.3.1, <http://www.env.go.jp/water/kaiyo/monitoring.html>).

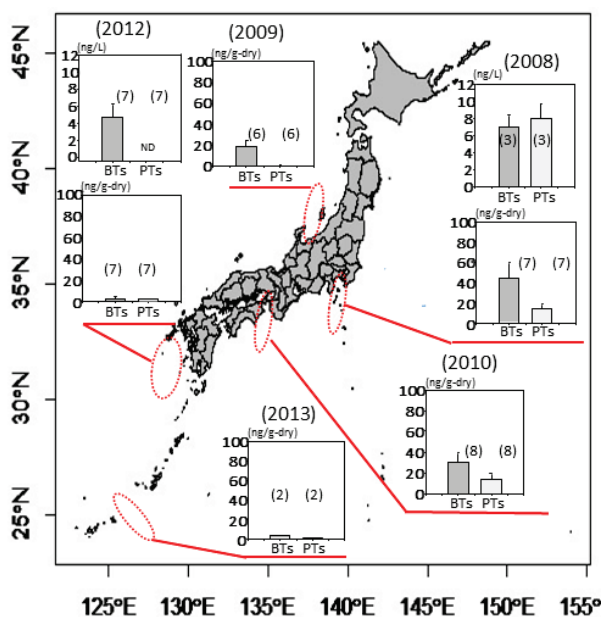


Fig. 3.3.1 Geographical distribution of organotin (OT) compounds, such as butyltins (BTs, shaded column) and phenyltins (PTs, open column), in the marine environment around Japan from 2008 to 2013. Data are expressed as mean \pm standard error of each compound in sediment and/or seawater sample except for the 2013 sample. Number in parentheses inside graphs represents number of samples.

OT-free antifouling biocides

Among the OT-free antifouling biocides, long-term monitoring data for two herbicides, including Irgarol 1051 (2-methylthio-4-*tertiary*-butylamino-6-cyclopropylamino-*s*-triazine) and Diuron (3-(3,4-dichlorophenyl)-1,1-dimethylurea) in the Seto Inland Sea are available (Balakrishnan *et al.*, 2012; Kaonga *et al.*, 2015). The concentrations of these biocides in both sediment and seawater had kept almost the same level from 2008 to 2013, with the exception of seawater samples collected in 2013 (Fig. 3.3.2). In the 2013 samples, the concentrations of both biocides tend to be high indicating that marinas and harbors contained markedly high concentration of these biocides.

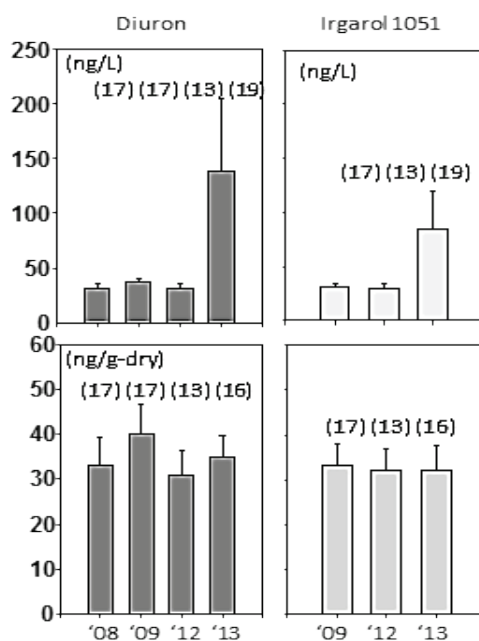


Fig. 3.3.2 Chronological changes in Diuron (left) and Irgarol 1051 (right) concentrations of seawater (upper graphs) and sediment (lower graphs) samples in the Seto Inland Sea. Data are expressed as mean \pm standard error. Number in parentheses represent number of samples.

The ecological risk of both biocides is considered to be low at the averaged concentration levels. However, the detected highest concentration, especially for Irgarol 1051, is $1.07 \mu\text{g/L}$ and the value corresponds to Lethal Concentration 50 (EC_{50}) values for some marine algal species (*e.g.*, $1.1 \mu\text{g/L}$ for 72-h EC_{50} of *Chaetoceros gracilis* (Mochida and Fujii, 2008)). Thus, continuous monitoring for this biocide is considered a priority. Incidentally, until now, biocides other than Irgarol 1051 and Diuron have never been detected at concentration levels that have the potential to affect marine organisms other than phytoplankton species involved in primary production in Japanese coastal waters. The potential impact of these antifouling biocides on primary production warrants further research.

References

- Balakrishnan, S., Takeda, K. and Sakugawa, H. 2012. Occurrence of Diuron and Irgarol in seawater, sediments and planktons of Seto Inland Sea, Japan. *Geochemical Journal* **46**: 169–177.
- Kaonga, C.C., Takeda, K. and Sakugawa, H. 2015. Antifouling agents and fenitrothion contamination in seawater, sediment, plankton, fish and selected marine animals from the Seto Inland Sea, Japan. *Geochemical Journal* **49**: 23–37.
- Mochida, K. and Fujii, K. 2008. Toxicity in plankton and fish, pp. 365–382 *in*: *Ecotoxicology of Antifouling Biocides* edited by T. Arai, H. Harino, M. Ohji and W. Langston, Springer-Verlag, Berlin.
- Okamura, H. and Mieno, H. 2006. Present status of antifouling systems in Japan: tributyltin substitutes in Japan, pp. 201–212 *in*: *The Handbook of Environmental Chemistry Vol. 5* edited by I. Konstantinou, Springer-Verlag, Berlin.

3.4 *Distribution and temporal trends of mercury in sediments from coastal seas around Japan*

Hideaki Maki

National Institute for Environmental Studies, Tsukuba, Ibaraki, Japan

The Minamata Legacy

As mercury is distributed ubiquitously in marine environments because of its multilateral sources, including atmospheric deposition originating from combustion of coal, gold mining, geological processes (*e.g.*, volcanic eruptions) and anthropogenic releases into water system, many monitoring efforts targeting various marine environmental media such as seawater, sediment and biota have been conducted to protect human health from mercury poisoning via dietary intakes of fisheries products. Significant attention has so far been paid to mercury pollution in Japan since the Minamata disease tragedy was recognized in the 1950s as a consequence of anthropogenic mercury pollution in Minamata Bay in Kumamoto, Kyushu. As a regulatory issue to protect human health, commercial deliveries of some marine fishes caught in the coastal seas of Japan have been occasionally banned because of accidental mercury contamination in them. To protect human health and the environment from the effects of anthropogenic emissions and release of mercury and to reduce mercury contamination in the marine environment, the Minamata Convention on Mercury (UNEP, 2016) was adopted as an international environment treaty and has already been signed by 128 countries. In Japan, industrial demand for mercury has been drastically reduced because of regulation and source controls (Fig. 3.4.1).

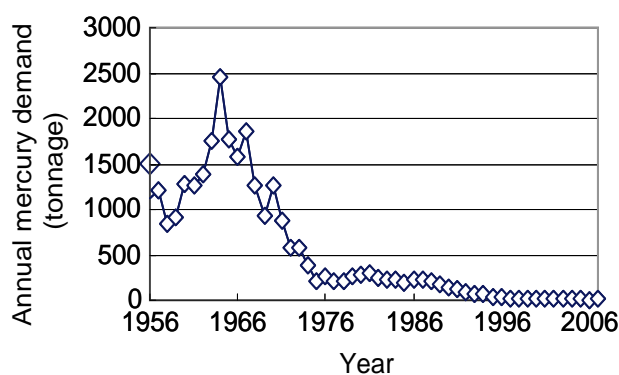


Fig. 3.4.1 Fifty-year trend of mercury demand from 1956 to 2006 in Japan (METI, 2009).

The Ministry of the Environment (MoE, formerly, Environmental Agency, the Government of Japan), has set provisional removal standards for mercury to prevent elutions from being discharged into the water column in marine environments (MoE, 2009). MoE has been conducting rigorous monitoring surveys of marine sediments by setting eight transects in the coastal seas around Japan since 1975 (Fig. 3.4.2; MoE, 2009). This monitoring work was earlier aimed to evaluate the influence of human

activities on land on the coastal sea environment but recently shifted to evaluation of ocean dumping of various sludges and waste soils. Water depths for sampling stations varied from 20 to 6,500 m.

Recent horizontal distribution of sedimentary THg

Figure 3.4.2 shows the horizontal distribution of total mercury (THg) concentrations ($\mu\text{g/g}$ dry weight, dw) in sediments in the coastal seas around Japan from 2004 to 2014. Data for each transect are summarized in Table 3.4.1. The concentrations in the Pacific Ocean (transects A, B and C) are higher than those in the East China Sea (transect E), excluding transect D, and the Sea of Japan (transects F, G and H; Fig. 3.4.2).

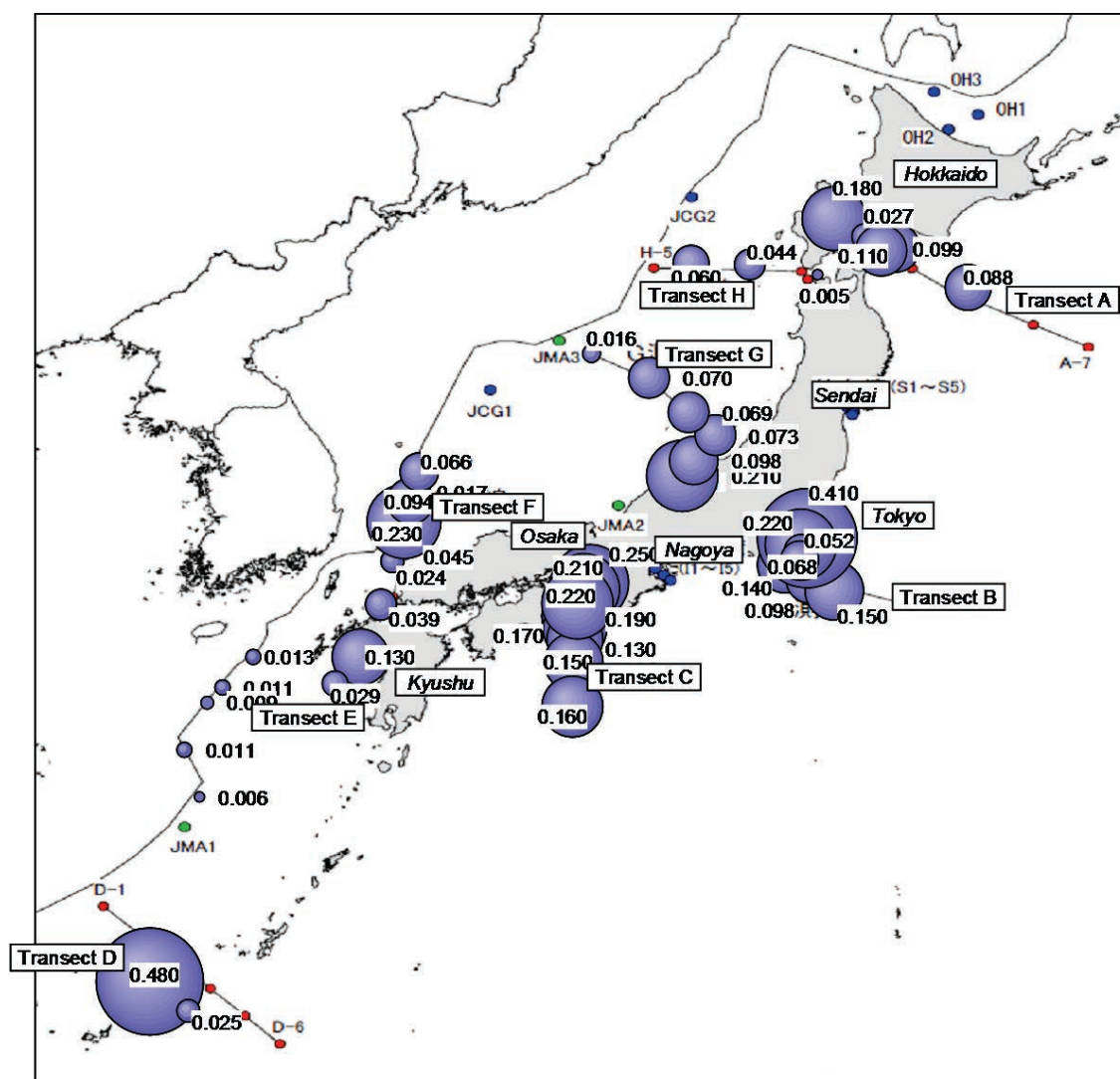


Fig. 3.4.2 Horizontal distribution of sedimentary mercury in the coastal seas around Japan from 2004 to 2014 (NIES, 2016). Value on each bubble shows THg concentration in $\mu\text{g/g}$ dry weight (dw).

Table 3.4.1 Total mercury (THg) concentration ($\mu\text{g/g}$ dry weight, dw) in sediments in eight transects in the coastal seas around Japan, from 2004 to 2014 (NIES, 2016).

Transect	THg concentration ($\mu\text{g/g}$ dw)			Data acquisition	Number of stations
	Lowest	Highest	Average	Year	
A	0.027	0.180	0.101	2005	5
B	0.098	0.150	0.129	2008	3
C	0.130	0.190	0.160	2010	5
D	0.025	0.480	0.253	2013	2
E	0.016	0.130	0.030	2012	8
F	0.017	0.230	0.074	2004	7
G	0.016	0.210	0.089	2009	6
H	0.005	0.060	0.036	2005	3

Average THg concentrations in transects B and C are 0.13 and 0.16 $\mu\text{g/g}$ dw, respectively, which are higher than in other transects and seem to reflect the significant influences of anthropogenic pollution from huge urbanized and heavily industrialized areas around Tokyo Bay (transect B) and Osaka Bay (transect C). All of the detected THg concentrations values are lower than the provisional removal standards for mercury to prevent elutions from being discharged into the water column in marine environments.

Chronological change of sedimentary THg

Figure 3.4.3 shows the chronological changes of sedimentary THg in transects B and C. Only transect C shows significant decay of sedimentary THg (rank correlation test, $p < 0.01$), but no significant THg reductions are observed in other transects deployed along Japan's coastal seas (*i.e.*, transects A, B, D, E, F, G, H and E; Fig. 3.4.2). This might be due to the difference in mercury sources into marine environment between each transect.

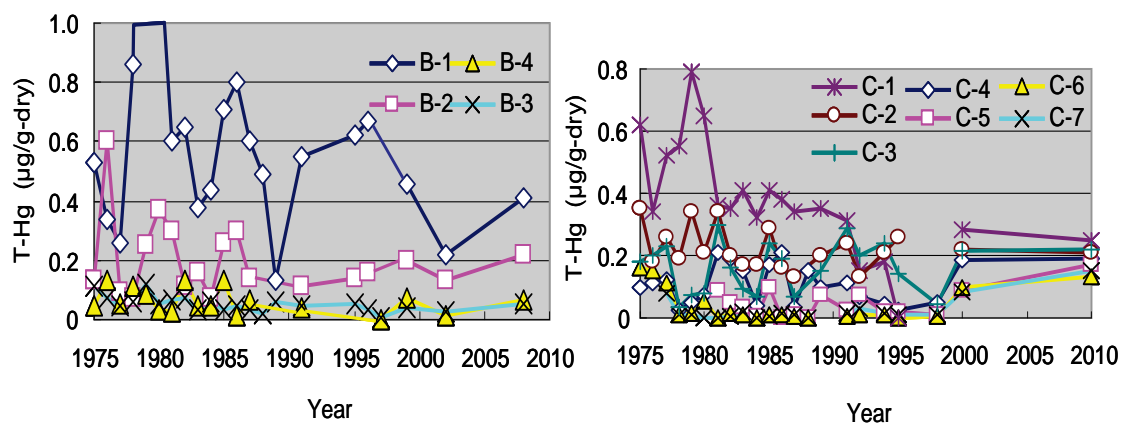


Fig. 3.4.3 Time series of annual changes of sedimentary mercury in transects B1 to B4 (left) and C1 to C7 (right) from 1975 to 2010, based on data from the National Institute for Environmental Studies (NIES, 2016).

Further successive monitoring effort is needed to know spacio-temporal distributions and background concentrations of mercury in coastal seas around Japan.

References

- METI (Agency for Natural Resources and Energy, Ministry of Economy, Trade and Industry). 2009. Non-Ferrous Metal Supply and Demand Statistics survey.
- MoE (Ministry of the Environment). 2009. Present status of marine pollution in the sea around Japan as based on data from Marine Environment Monitoring Survey results, Fiscal Years 1998–2007. The Government of Japan. https://www.env.go.jp/water/kaiyo/monitoring/status_report/en-1.pdf;
https://www.env.go.jp/water/kaiyo/monitoring/status_report/en-2.pdf.
- NIES (National Institute for Environmental Studies). 2016. Environmental GIS (in Japanese). <http://envgis.nies.go.jp/kaiyo/>.
- UNEP (United Nations Environment Programme). 2016. Minamata Convention on Mercury. <http://www.mercuryconvention.org/>.

4 Republic of Korea

4.1 Accumulation and temporal trends of polychlorinated biphenyls (PCBs), organochlorine pesticides (OCPs) and polybrominated diphenyl ethers (PBDEs) in finless porpoises (*Neophocaena asiaeorientalis*) in the Republic of Korea

Hyo-Bang Moon

Hanyang University, Ansan, Republic of Korea

Marine mammals have been considered important species for monitoring persistent organic pollutants (POPs) contamination and its long-term potential risk in marine ecosystems of Korea. Due to their smaller body size and regional distribution of their larger populations compared to other species, finless porpoises (*Neophocaena asiaeorientalis*) are frequently caught as bycatch by fishermen in the western and southern parts of the coastal waters of Korea (Fig. 4.1.1). As part of environmental monitoring and research efforts to track the contamination status in Korean coastal waters, the total concentrations of POPs, including polychlorinated biphenyls (PCBs), dichloro-diphenyl-trichloroethanes (DDTs), chlordane related compounds (CHLs), hexachlorocyclohexanes (HCHs), hexachlorobenzene (HCB), and polybrominated diphenyl ethers (PBDEs) were measured in the blubber of finless porpoises ($n = 52$) collected in 2003 and were in the ranges of 130–3300 ng/g lipid weight (lw), 480–45,300 ng/g lw, 20.3–840 ng/g lw, 50.3–3000 ng/g lw, 9.5–200 ng/g lw and 180–1600 ng/g lw, respectively (Moon *et al.*, 2010).



Fig. 4.1.1 Sampling map and geographical distribution of the finless porpoises (*Neophocaena asiaeorientalis*) collected from Korean coastal waters.

For the blubber of finless porpoises ($n = 77$) collected in 2010, the total concentrations of PCBs, DDTs, CHLs, HCHs, HCB, and PBDEs were 50.5–3200 ng/g lw, 240–10,100 ng/g lw, 11.3–300 ng/g lw, <LOQ–810 ng/g lw, 1.6–100 ng/g lw, and 48.0–1000 ng/g lw, respectively. Significant age- and sex differences were found for the concentrations and accumulation profiles of organochlorine pesticides (OCPs) and polybrominated diphenyl ethers (PBDEs) in blubber of finless porpoises collected in 2003 and 2010.

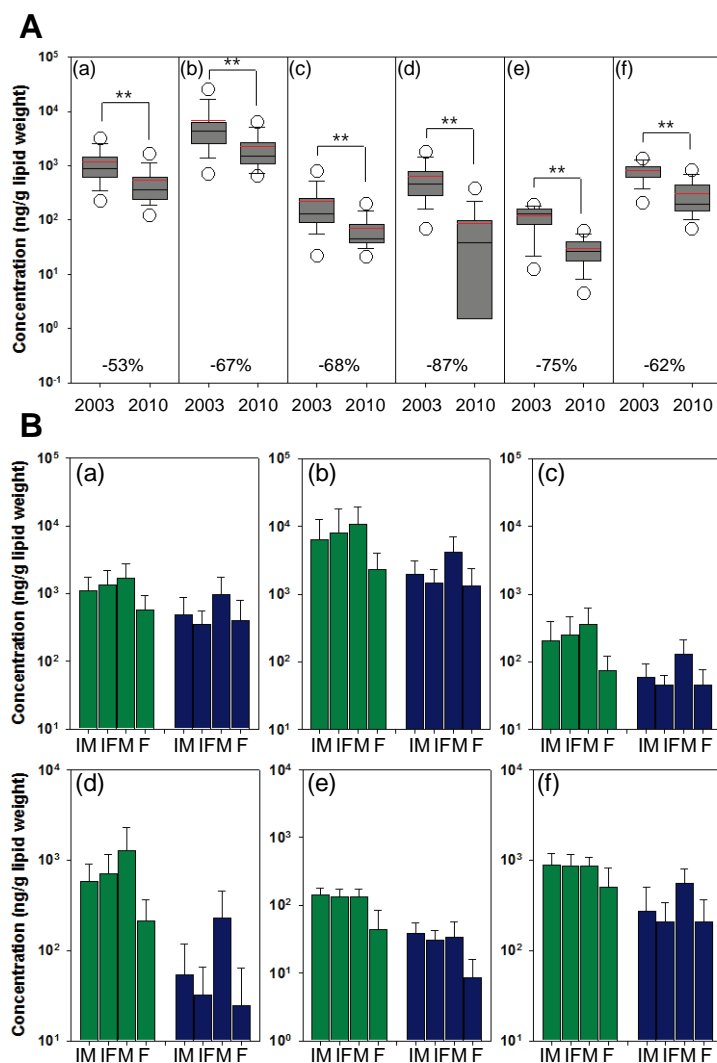


Fig. 4.1.2 Temporal trends of (a) PCBs, (b) DDTs, (c) CHLs, (d) HCHs, (e) HCB, and (f) PBDEs in (A) all of the samples and (B) groups according to sex and growth of finless porpoises collected between 2003 and 2010 from Korean coastal waters. Significant percentage declines are indicated for each chemical group ($*p < 0.05$; $**p < 0.005$). The box plot shows 10th, 25th, 50th, and 90th percentiles with error bars. Circles indicate the 5th percentile (lower) and the 95th percentile (upper). Arithmetic mean concentrations are given as red bars. Sample numbers for sex and growth groups were as follows: immature male ($n = 15$), immature female ($n = 20$), mature male ($n = 7$), and mature female ($n = 10$) collected in 2003 (total numbers = 52); immature male ($n = 20$), immature female ($n = 28$), mature male ($n = 19$), and mature female ($n = 10$) collected in 2010 (total numbers = 77).

The effects of life history variables (*i.e.*, age and sex) as confounding factors were investigated for the accumulation of POPs in finless porpoises collected between 2003 and 2010. The Mann-Whitney *U* test was performed for age and sex groups. There was no significant difference in size or age among groups ($p = 0.427\text{--}0.742$), indicating that age and growth stage are not influencing factors between time-gap samples of finless porpoises. The total concentrations of PCBs, OCPs and PBDEs in finless porpoises showed significant decreasing trends ($p < 0.001$) between the sampling years of 2003 and 2010 (Fig. 4.1.2). Based on the average concentrations of these contaminants, the reduction rates were 53%, 67%, 68%, 87%, 75%, and 62% for PCBs, DDTs, CHLs, HCHs, HCB, and PBDEs, respectively. Of 52 finless porpoise samples collected in 2003, 27% exceeded the NOAEL (no observed adverse effect level) of immunosuppression by DDTs of harbor seals (De Swart *et al.*, 1996) and 27% exceeded the total PCB-risk based toxic effect concentration (*i.e.*, endocrine disruption and immunotoxicity) of 1300 ng/g lw reported for harbor seals (Mos *et al.*, 2010), implying adverse health effects on this species.

In conclusion, finless porpoises can be used as sentinels of ecosystem health for the long-term monitoring of pollution by POPs in Korean coastal waters of the Yellow Sea.

References

- De Swart, R.L., Ross, P.S., Vos, J.G. and Osterhaus, A.D.M.E. 1996. Impaired immunity in harbor seals (*Phoca vitulina*) exposed to bioaccumulated environmental contaminants: review of a long-term feeding study. *Environmental Health and Perspectives* **104**: 823–828.
- Moon, H.-B., Choi, H.-G., An, Y.-R., Park, K.-J., Choi, S.-G., Moon, D.-Y. and Kannan, K. 2010. Contamination status and accumulation features of PCDDs, PCDFs and dioxin-like PCBs in finless porpoises (*Neophocaena phocaenoides*) from Korean coastal waters. *Journal of Hazardous Materials* **183**: 799–805.
- Mos, L., Cameron, M., Jeffries, S.J., Koop, B.F. and Roos, P.S. 2010. Risk-based analysis of polychlorinated biphenyl toxicity in harbor seals. *Integrated Environmental Assessment and Management* **6**: 631–640.

4.2 Accumulation and temporal trends of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/PCDFs) and dioxin-like polychlorinated biphenyls (DL-PCBs) in finless porpoises in the Republic of Korea

Hyo-Bang Moon

Hanyang University, Ansan, Republic of Korea

Polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) and polychlorinated biphenyls (PCBs) are introduced into the environment mainly by waste incineration and chemical manufacturing processes. In Korea, a nationwide inventory survey for PCDD/Fs reported that the primary sources of PCDD/Fs in the atmosphere are combustion processes from municipal solid waste incinerators (MSWIs) and the steel industry, which collectively accounted for 95% of the total PCDD/F emissions (Ministry of Environment, 2014). The total emissions of toxic equivalents (TEQs) of PCDD/Fs generated from waste incinerators in 2001 were reduced by 88% in the past 10 years (Ministry of Environment, 2014).

Marine mammals are top-level predators in the marine ecosystem and contain high levels of persistent organic pollutants (POPs) in their fat tissues. Due to their smaller body size and larger populations compared to other species, finless porpoises (*Neophocaena phocaenoides*) are frequently caught as bycatch by fishermen in the western and regional distribution of their southern parts in the coastal waters of Korea (Fig. 4.2.1). As part of environmental monitoring and research efforts to track the contamination status in Korean coastal waters, total concentrations of PCDDs, PCDFs, non-*ortho* PCBs and mono-*ortho* PCBs were measured in the blubber of finless porpoises ($n = 52$) collected in 2003 and ranged from 5.4–31 pg/g lipid weight (lw), 14–54 pg/g lw, 0.4–3.6 ng/g lw, and 27–519 ng/g lw,



Fig. 4.2.1 Sampling map and geographical distribution of the finless porpoises (*Neophocaena asiaeorientalis*) collected from Korean coastal waters.

respectively (Moon *et al.*, 2010). For the blubber of finless porpoises ($n = 76$) collected in 2010, the total concentrations of PCDDs, PCDFs, non-*ortho* PCBs, and mono-*ortho* PCBs were 1.01–27.5 pg/g lw, 0.92–123 pg/g lw, 0.001–0.93 ng/g lw, and 10.6–261 ng/g lw, respectively (Jeong *et al.*, 2016). Age- and sex-dependent accumulation patterns were found for PCDFs and DL-PCBs in blubber of finless porpoises collected in 2003 and 2010.

The TEQ concentrations of PCDDs, PCDFs, non-*ortho* PCBs, and mono-*ortho* PCBs in the blubber of finless porpoises collected in 2003 ranged from 0.1–4.1 pg/g lw, 1.7–7.6 pg/g lw, 1.9–8.6 pg/g lw, and 0.8–16 pg/g lw, respectively. In the blubber of finless porpoises collected in 2010, the TEQ

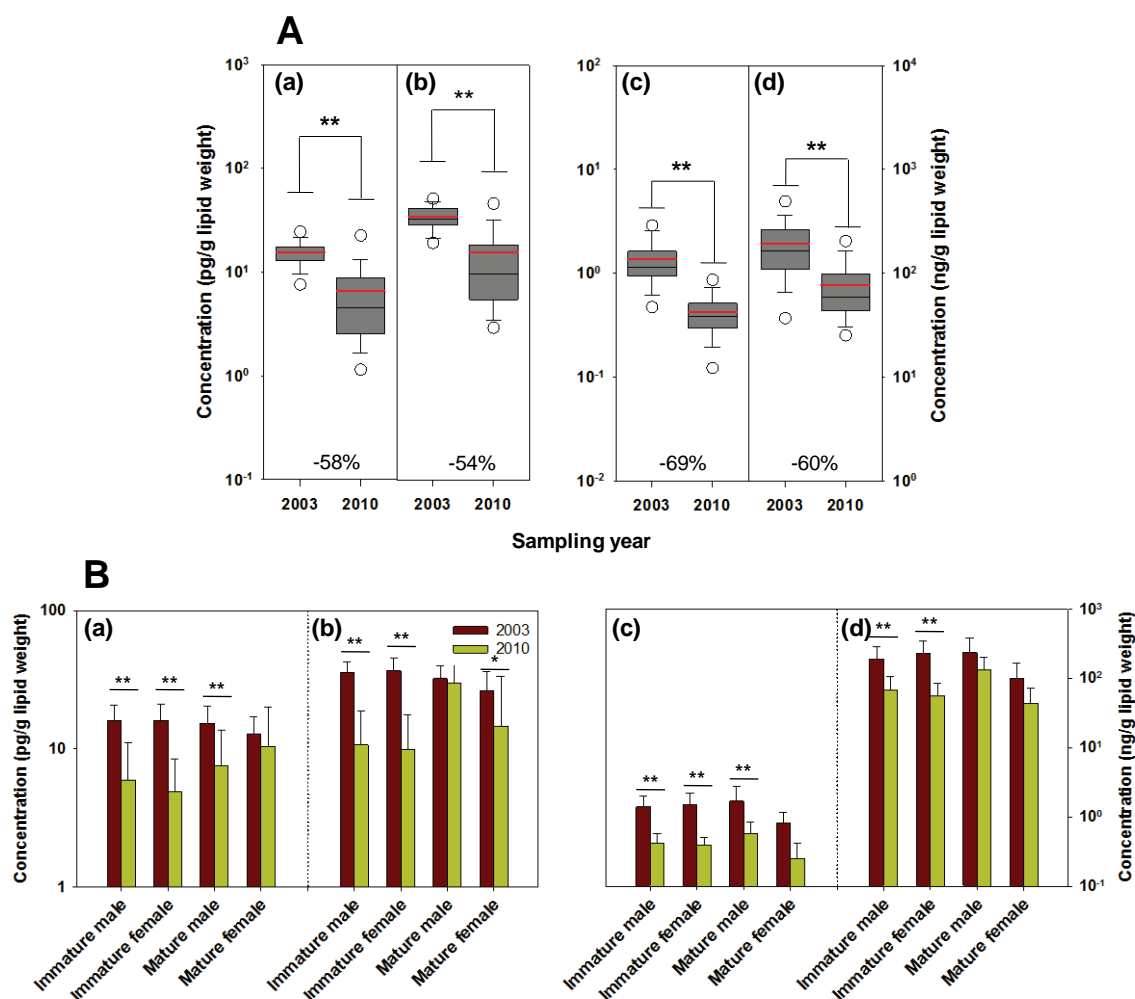


Fig. 4.2.2 Temporal trends of (a) PCDDs, (b) PCDFs, (c) non-*ortho* PCBs, and (d) mono-*ortho* PCBs in (A) all of the samples and (B) groups according to sex and growth of finless porpoises collected between 2003 and 2010 from Korean coastal waters. Significant percentage declines are indicated for each chemical group (* $p < 0.05$; ** $p < 0.005$). The box plot shows 10th, 25th, 50th, and 90th percentiles with error bars. Circles indicate the 5th percentile (lower) and the 95th percentile (upper). Arithmetic mean concentrations are given as red bars. Sample numbers for sex and growth groups were as follows: immature male ($n = 15$), immature female ($n = 20$), mature male ($n = 7$), and mature female ($n = 10$) collected in 2003 (total numbers = 52); immature male ($n = 19$), immature female ($n = 28$), mature male ($n = 19$), and mature female ($n = 10$) collected in 2010 (total numbers = 76).

concentrations of PCDDs, PCDFs, non-*ortho* PCBs, and mono-*ortho* PCBs ranged from 0.002–14.1 pg/g lw, <LOQ to 36.5 pg/g lw, <LOQ to 8.3 pg/g lw, and 0.3–7.8 pg/g lw, respectively. The total TEQ levels in blubber of finless porpoises from both sampling years were lower than the NOAEL (no observed adverse effect level; 62 pg TEQ/g) and LOAEL (lowest observed adverse effect level; 209 pg TEQ/g) known to have immunomodulatory effects due to dioxin-like contaminants in harbor seals (*Phoca vitulina*; Ross *et al.*, 1995). To account for the effects of confounding factors in the bioaccumulation of these contaminants in finless porpoises collected between 2003 and 2010, the Mann-Whitney *U* test was performed for age and sex groups. There was no significant difference in size or age among groups ($p = 0.427\text{--}0.742$), indicating that age and growth stage are not influencing factors between time-gap samples of finless porpoises. The total concentrations of PCDD/Fs and DL-PCBs in finless porpoises showed significant decreasing trends ($p < 0.001$) between 2003 and 2010 (Fig. 4.2.2). Based on the average concentrations of these contaminants, the reduction rates were 58%, 54%, 69%, and 60% for PCDDs, PCDFs, non-*ortho*, and mono-*ortho* PCBs, respectively. In addition, the congener-specific temporal trends of PCDD/Fs and DL-PCBs also significantly decreased between time-gap samples.

Finless porpoises can be used as sentinels of ecosystem health for the long-term monitoring of pollution by dioxins and furans in Korean coastal waters of the Yellow Sea. Our results suggest that the implementation of source control and regulations on dioxin-like contaminants has been effective for marine mammals in Korea.

References

- Jeong, Y., Kim, S.-J., Shin, K.-H., Hwang, S.Y., An, Y.-R. and Moon, H.-B. 2016. Accumulation and temporal changes of PCDD/Fs and dioxin-like PCBs in finless porpoises (*Neophocaena asiaeorientalis*) from Korean coastal waters: Tracking the effectiveness of regulation. *Marine Pollution Bulletin* **105**: 30–36.
- Ministry of Environment. 2014. Report on the effect of the regulation to dioxin emission. Ministry of Environment of Korea, Seoul, Korea. (in Korean)
- Moon, H.-B., Choi, H.-G., An, Y.-R., Park, K.-J., Choi, S.-G, Moon, D.-Y. and Kannan, K., 2010. Contamination status and accumulation features of PCDDs, PCDFs and dioxin-like PCBs in finless porpoises (*Neophocaena phocaenoides*) from Korean coastal waters. *Journal of Hazardous Materials* **183**: 799–805.
- Ross, P.S., De Swart, R.L., Reijnders, P.J.H., Van Loveren, H., Vos, J.G. and Osterhaus, A.D.M.E. 1995. Contaminant-related suppression of delayed-type hypersensitivity and antibody responses in harbor seals fed herring from the Baltic sea. *Environmental Health Perspectives* **103**: 162–167.

4.3 Contamination status and temporal trends of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/PCDFs) in sediments and bivalves from coastal areas in the Republic of Korea

Hyo-Bang Moon¹ and Minkyu Choi²

¹Hanyang University, Ansan, Republic of Korea

²National Institute of Fisheries Science (NIFS), Busan, Republic of Korea

Polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) are mainly introduced into the environment by waste incineration and chemical manufacturing processes. In Korea, a nationwide inventory survey for PCDD/Fs reported that the primary sources of PCDD/Fs in the atmosphere are combustion processes from municipal solid waste incinerators (MSWIs) and the steel industry, which collectively accounted for 95% of the total PCDD/F emissions (Ministry of Environment, 2014). The total emissions of toxic equivalents (TEQs) of PCDD/Fs generated from waste incinerators in 2001 were reduced by 88% in the past 10 years (Ministry of Environment, 2014).

The nation-wide monitoring of PCDD/Fs in sediments and bivalves from the coastal waters of Korea was launched by the National Institute of Fisheries Science (NIFS) in January 2000 to assess the contamination status, spatial distribution, and temporal trends of these persistent organic pollutants (POPs) in coastal–marine environments of Korea. Surface sediments and two kinds of bivalve species, including mussels (*Mytilus coruscus* and *M. edulis*) and oysters (*Crassostrea gigas*), were sampled at 25 locations from the Korean coast during the period 2000–2012 (Fig. 4.3.1).

The toxic equivalent (TEQ) concentrations of PCDD/Fs in 2000–2012 ranged from <LOQ to 18.8 (mean: 1.61) pg/g dry weight (dw) for all of the sediment samples and from <LOQ to 1.66 (mean: 0.19) pg/g wet weight for all of the bivalve samples. The highest concentrations of PCDD/Fs in sediments were observed in the southeastern part of Korea (Ulsan, Onsan, Busan, Masan, Jinhae bays), which are the locations close to the largest industrial complexes and harbors (Fig. 4.3.1). This result indicates that intensive industrial and shipping activities are major contamination sources of PCDD/Fs in Korean coastal waters (Moon *et al.*, 2008; Choi *et al.*, 2011).

The spatial distribution of PCDD/Fs in bivalves showed a relatively homogenous pattern compared to that of sediment contamination (Choi *et al.*, 2010). A significant decreasing trend of PCDD/Fs was found in sediments from Korean coastal waters from 2010 to 2012, as shown in Figure 4.3.2, suggesting the association with a strong regulation on PCDD/Fs in flue gas from waste incinerators in Korea (Ministry of Environment, 2014). However, no significant temporal trends were found for PCDD/Fs in bivalves from Korean coastal waters (Fig. 4.3.2). To evaluate the ecotoxicological effects of PCDD/Fs in coastal sediment from Korea, sediment quality guidelines (SQGs) such as probable effect level (PEL) and threshold effect level (TEL) from Florida State University were used. Our evaluation showed that almost half of the sediment samples (46%) had exceeding values of PCDD/Fs for the TEL (0.85 pg TEQ/g dw), implying ecotoxicological potential risks to marine organisms in Korean coastal waters.

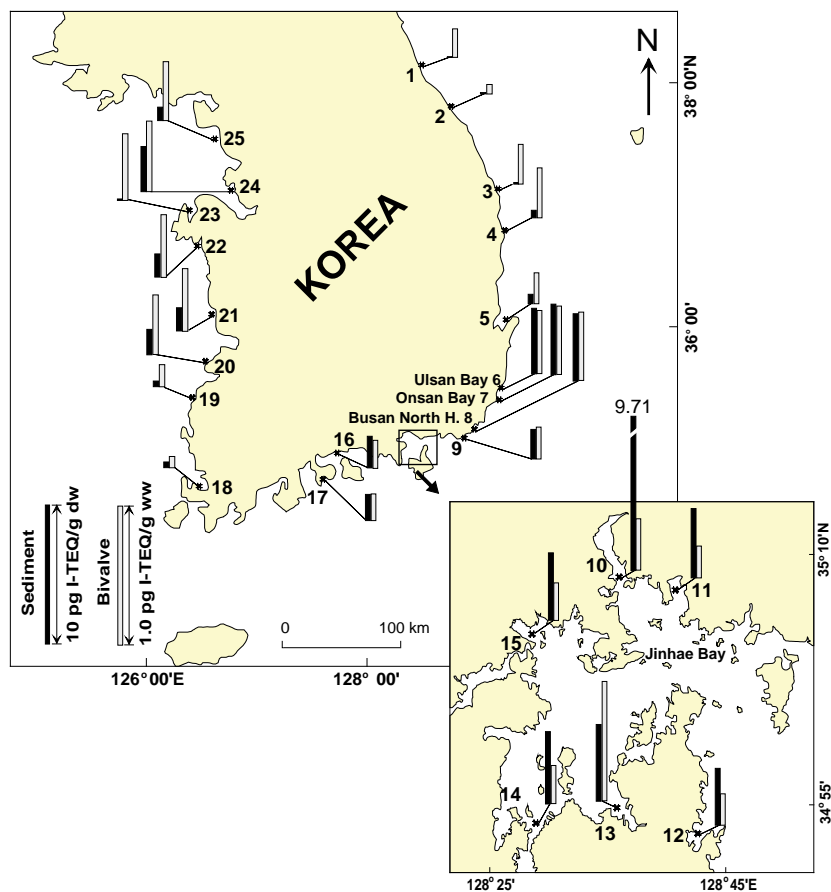


Fig. 4.3.1 Spatial distribution of mean concentrations of PCDD/Fs in sediment and bivalves (mussels and oysters) from 25 locations in Korean coastal waters during the period of 2000–2012.

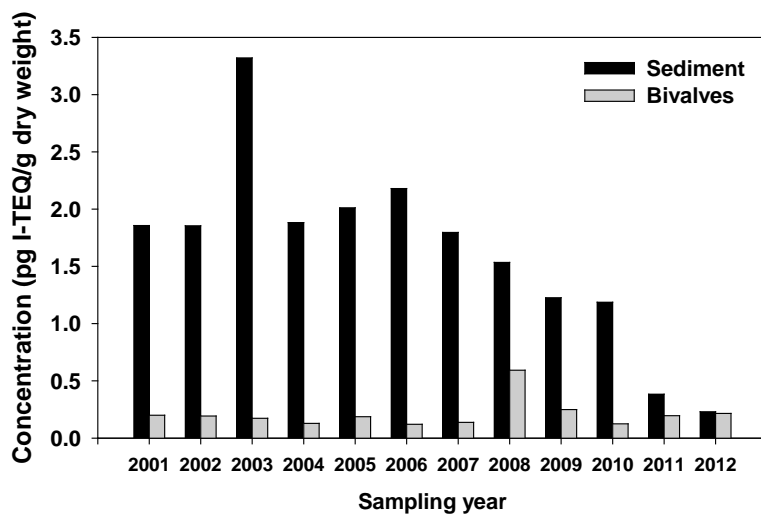


Fig. 4.3.2 Temporal trends of mean concentrations of PCDD/Fs in sediment and bivalves (mussels and oysters) from 25 locations in Korean coastal waters during the period 2000–2012.

In this context, marine sediments and bivalves are key environmental compartments used as ocean pollution indicators to track dioxin and furan pollution in the coastal waters of Korea.

References

- Choi, H.-G., Moon, H.-B., Choi, M., Yu, J. and Kim, S.-S. 2010. Mussel watch program for organic contaminants along the Korean coast, 2001-2007. *Environmental Monitoring and Assessment* **169**: 473–485.
- Choi, H.-G., Moon, H.-B., Choi, M. and Yu, J. 2011. Monitoring of organic contaminants in sediments from the Korean coast: Spatial distribution and temporal trends (2001–2007). *Marine Pollution Bulletin* **62**: 1352–1361.
- Ministry of Environment. 2014. Report on the effect of the regulation to dioxin emission, Ministry of Environment of Korea, Seoul, Korea. (in Korean)
- Moon, H.-B., Choi, H.-G., Lee, P.-Y. and Ok, G. 2008. Congener-specific characterization and sources of polychlorinated dibenzo-*p*-dioxins, dibenzofurans and dioxin-like polychlorinated biphenyls in marine sediments from industrialized bays of Korea. *Environmental Toxicology and Chemistry* **27**: 323–333.

4.4 Contamination status and temporal trends of polycyclic aromatic hydrocarbons (PAHs) in sediments and bivalves from coastal areas in the Republic of Korea

Hyo-Bang Moon¹ and Minkyu Choi²

¹Hanyang University, Ansan, Republic of Korea

²National Institute of Fisheries Science (NIFS), Busan, Republic of Korea

Polycyclic aromatic hydrocarbons (PAHs) are ubiquitous environmental pollutants of public concern and are derived mainly from anthropogenic activities in coastal areas of Korea. PAHs are generated by incomplete fuel combustion, domestic and industrial wastewaters, and spillage of crude oil and its refined products. PAHs derived from various sources can be transported to marine environments via riverine inputs and atmospheric deposition.

The nation-wide monitoring of 16 PAHs, which are recommended as the toxic priority pollutants, in sediments and bivalves from coastal waters of Korea was launched by the National Institute of Fisheries Science (NIFS) in January 2000 to assess the contamination status, spatial distribution, and temporal trend of these contaminants in the coastal environment of Korea. Surface sediments and two kinds of bivalve species, including mussels (*Mytilus coruscus* and *M. edulis*) and oysters (*Crassostrea gigas*), were sampled at 25 locations from the Korean coast over the period 2000–2012 (Fig. 4.4.1).

The total concentrations of PAHs in 2000–2012 ranged from 1.71 to 3060 (mean: 216) ng/g dry weight (dw) for all of the sediment samples and from 4.36 to 2653 (mean: 182) pg/g wet weight for all of the bivalve samples. The highest concentrations of PAHs in sediments and bivalves were observed in the southeastern part of Korea (Ulsan, Onsan, Busan, Masan, Jinhae bays), which are the locations close to the largest industrial complexes and harbors (Fig. 4.4.2).

This result indicates that intensive industrial and shipping activities are the major contamination sources of PAHs in Korean coastal waters (Choi *et al.*, 2010, 2011). No significant temporal trend was found for PAHs in sediments and bivalves from Korean coastal waters, implying the presence of wide ranges and on-going sources of PAHs in the coastal environment of Korea. The total concentrations of PAHs in sediments from Korean coastal waters did not exceed the effect range low (ERL) and effect range median (ERM) suggested by the National Oceanic and Atmospheric Administration (NOAA) as screening values for sediment quality (Long and Morgan, 1990).

This report validates the use of marine sediments and bivalves as key environmental compartments that can serve as ocean pollution indicators to track pollution by PAHs in the coastal waters of Korea.

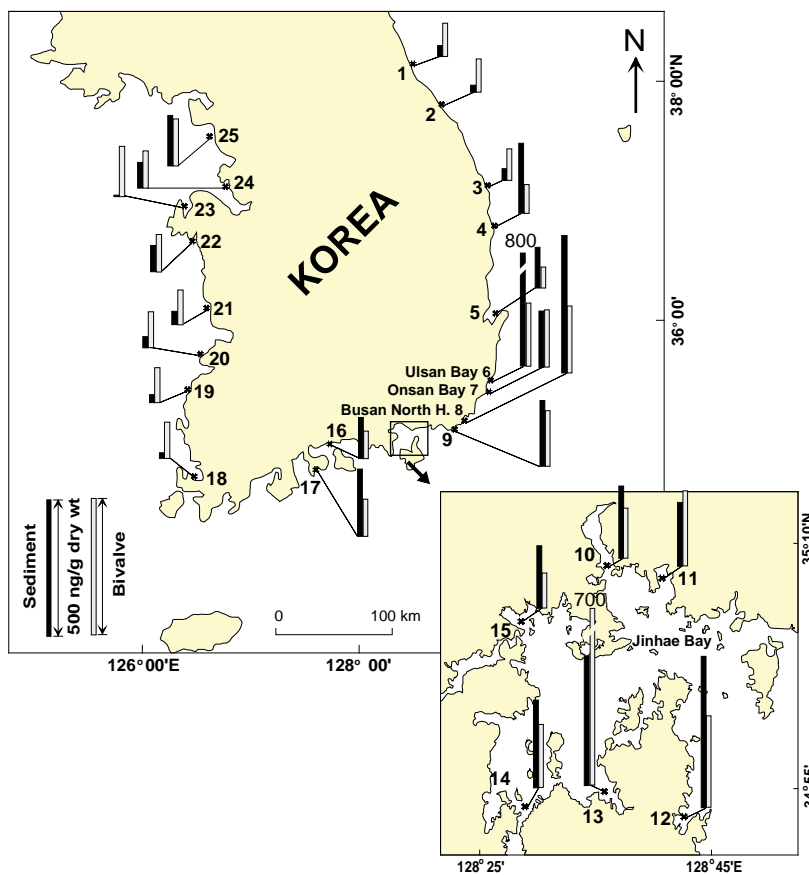


Fig. 4.4.1 Spatial distribution of mean concentrations of PAHs in sediment and bivalves (mussels and oysters) from 25 locations in Korean coastal waters during the period of 2000–2012.

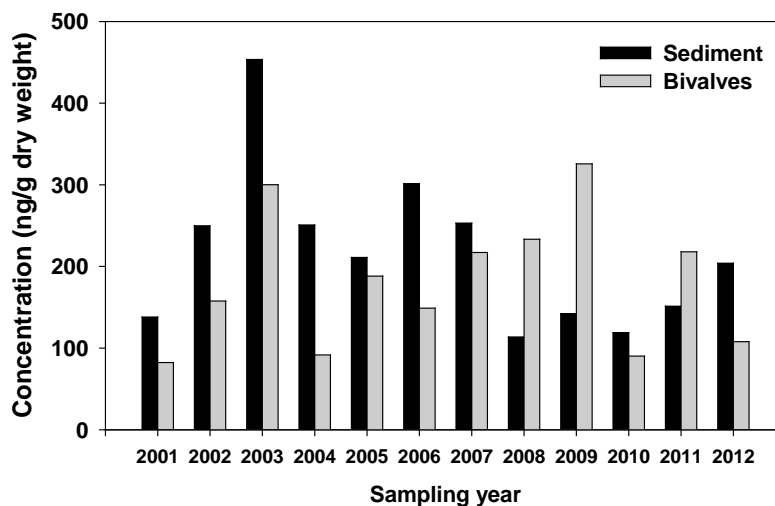


Fig. 4.4.2 Temporal trends of mean concentrations of PAHs in sediment and bivalves (mussels and oysters) from 25 locations in Korean coastal waters during the period of 2000–2012.

References

- Choi, H.-G., Moon, H.-B., Choi, M., Yu, J. and Kim, S.-S. 2010. Mussel watch program for organic contaminants along the Korean coast, 2001-2007. *Environmental Monitoring and Assessment* **169**: 473–485.
- Choi, H.-G., Moon, H.-B., Choi, M. and Yu, J. 2011. Monitoring of organic contaminants in sediments from the Korean coast: Spatial distribution and temporal trends (2001-2007). *Marine Pollution Bulletin* **62**: 1352–1361.
- Long, E.R. and Morgan, L.G., 1990. The potential for biological effects of sediment-sorbed contaminants tested in the National Status and Trends Program. NOAA Technical Memorandum NOS OMA 52, Seattle, Washington, USA.

4.5 Temporal trends of antifouling biocides (Butyltins) in rock shells (*Thais clavigera*) and finless porpoises (*Neophocaena asiaorientalis*) in the Republic of Korea

Minkyu Choi¹ and Hyo-Bang Moon²

¹National Institute of Fisheries Science (NIFS), Busan, Republic of Korea

²Hanyang University, Ansan, Republic of Korea

Biocidal additives have been used in antifouling paints for ship hulls and marine structures to prevent the immersed solid surfaces from fouling by colonizing marine organisms. Antifouling biocides can be directly introduced from ship hulls to marine environments and can cause adverse biological effects on non-target marine organisms (U.S. Environmental Protection Agency, 2004). Many studies have reported that organotins, including butyltins (BTs), can cause growth, developmental, and reproductive disorders in non-target marine species (Leung *et al.*, 2006) and are toxic to the immune systems of marine mammals (Frouin *et al.*, 2008). Accumulation of BTs in marine mammals is suspected to have an immunosuppressive effect, which can lead to subsequent infectious diseases or opportunistic infections by pathogens (Nakata *et al.*, 2002; Frouin *et al.*, 2008; Nakayama *et al.*, 2009). In Korea, restrictions addressing the use of TBT-based antifouling paints were partially introduced in 2000 for small boats, but their use was totally banned in 2003, and their residues on ships were banned in 2008 (MOF, 2008), following the action of the International Maritime Organization.

Imposex (*i.e.*, endocrine disruption of gonad development in marine invertebrates) and BT levels in rock shells (*Thais clavigera*; $n = 2,165$) were monitored on the Korean coast between 2004 and 2009 (Fig. 4.5.1) to investigate the temporal trend of BT contaminations. Mean imposex frequency (percentage occurrence of imposex female animals) showed values $> 95\%$ between 2004 and 2006 and decreasing from 2007 (Fig. 4.5.2a). Relative penis length index (RPLI) decreased from 2004 to 2009 (Fig. 4.5.2b), and significant differences between sampling years were found ($p < 0.05$). The concentrations of tributyltin (TBT) in *T. clavigera* also decreased over time, and levels of TBT and BT in 2004 were significantly greater than those in 2009 ($p < 0.05$). These indicate that rock shells were able to recover from imposex caused by BT contamination.

Finless porpoises (*Neophocaena asiaorientalis*) are frequently caught as bycatch by fishers in the western and southern parts of the coastal waters of Korea due to their smaller body size and regional distribution of their larger populations compared to other species (Fig. 4.5.3). We measured the concentrations of BTs in liver samples of *N. asiaorientalis* collected in 2003 and 2010 from Korean coastal waters to assess the effectiveness of legislative action against BTs. Butyltins were detected in all livers of finless porpoises ($n = 63$). The concentrations of monobutyltin (MBT), dibutyltin (DBT), TBT, and total BTs in finless porpoises were different between 2003 and 2010 (Fig. 4.5.4). The total concentrations of BTs in finless porpoises collected in 2003 (701 ± 371 ng/g wet weight, ww) were significantly higher than those measured in 2010 (155 ± 58.4 ng/g ww), indicating a clear decrease in BTs in marine mammals associated with TBT regulation in Korea. Earlier studies confirmed a clear decreasing trend in the concentrations of TBT in seawater and bivalves from Korean coastal waters (Choi *et al.*, 2009, 2010). The total ban on BT usage in Korea was found to be effective in reducing its level in marine environments, from seawater to marine mammals.

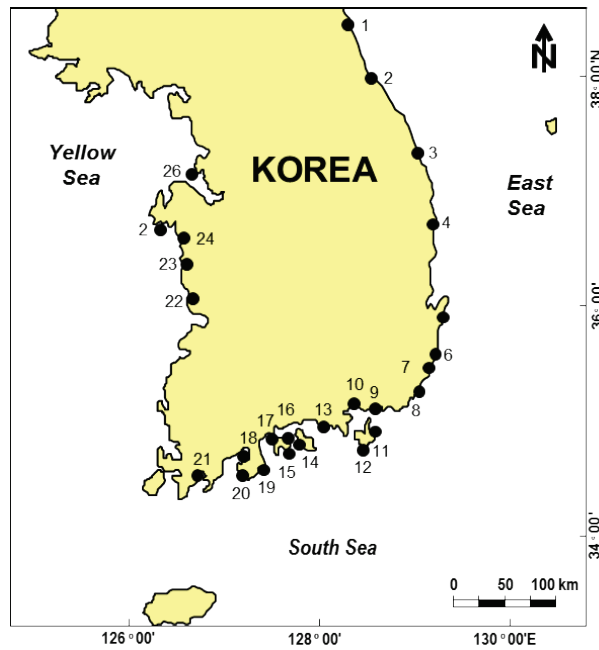


Fig. 4.5.1 Sampling sites of rock shells (*Thais clavigera*) along the Korean coast.

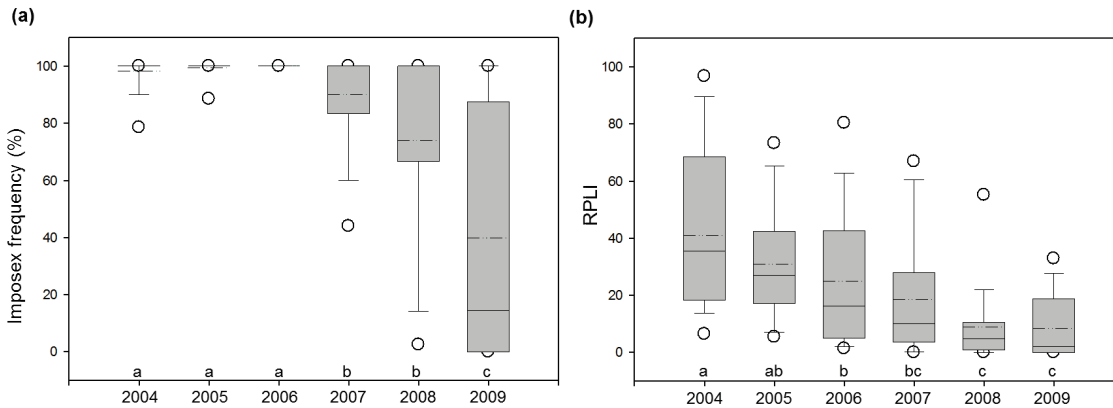


Fig. 4.5.2 Temporal trends in (a) imposex frequency and (b) relative penis length index (RPLI) in *T. clavigera* on the Korean coast. The box plot shows 10th, 25th, 50th, and 90th percentiles with error bars. Circles indicate the 5th percentile (lower) and the 95th percentile (upper).



Fig. 4.5.3 Sampling map and geographical distribution of finless porpoises (*Neophocaena asiaeorientalis*) collected from Korean coastal waters.

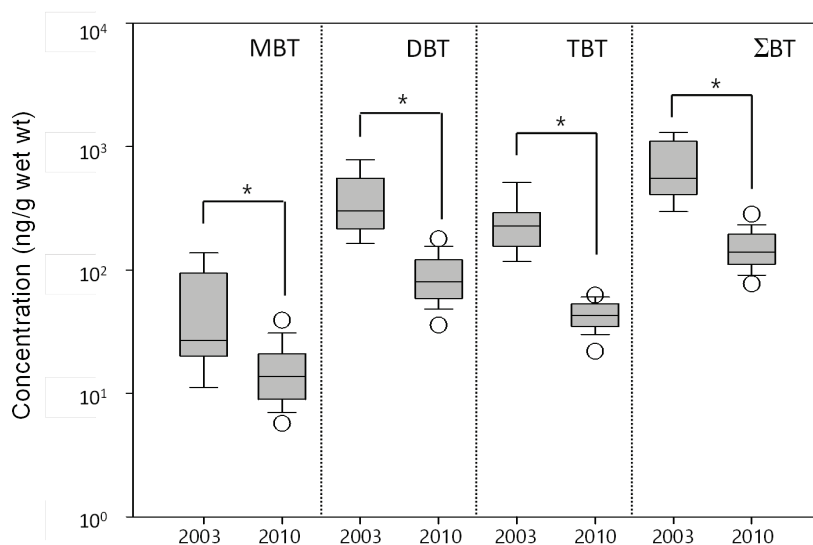


Fig. 4.5.4 Temporal trends of (a) monobutyltin (MBT), (b) dibutyltin (DBT), (c) tributyltin (TBT), and (d) total BTs in all of the samples of finless porpoises collected between 2003 and 2010 from Korean coastal waters. Significant percentage declines are indicated for each chemical group (* $p < 0.05$). The box plot shows 10th, 25th, 50th, and 90th percentiles with error bars. Circles indicate the 5th percentile (lower) and the 95th percentile (upper).

References

- Choi, M., Choi, H.-G., Moon, H.-B. and Kim, G.-Y. 2009a. Spatial and temporal distribution of tributyltin (TBT) in seawater, sediments and bivalves from coastal areas of Korea during 2001–2005. *Environmental Monitoring and Assessment* **151**: 301–310.
- Choi, M., Moon, H.-B., Yu, J., Eom, J.-Y. and Choi, H.-G. 2010. Temporal trend of butyltins in seawater, sediments, and mussels from Busan Harbor of Korea between 2002 and 2007: tracking the effectiveness of tributyltin regulation. *Archives of Environmental Contamination and Toxicology* **58**: 394–402.
- Frouin, H., Lebeuf, M., Saint-Louis, R., Hammill, M., Pelletier, E. and Fournier, M. 2008. Toxic effects of tributyltin and its metabolites on harbor seal (*Phoca vitulina*) immune cells in vitro. *Aquatic Toxicology* **90**: 243–251.
- Leung, K.M.Y., Kwong, R.P.Y., Ng, W.C., Horiguchi, T., Qui, J.W., Yang, R., Song, M., Jiang, G., Zheng, G.J. and Lam, P.K.S. 2006. Ecological risk assessments of endocrine disrupting organotin compounds using marine neogastropods in Hong Kong. *Chemosphere* **65**: 922–938.
- MOF (Ministry of Oceans and Fisheries). 2008. Marine Environmental Management Act 40.
- Nakata, H., Sakakibara, A., Kanoh, M., Kudo, S., Watanabe, H., Nagai, N., Miyazaki, N., Asano, Y. and Tanabe, S. 2002. Evaluation of mitogen-induced responses in marine mammal and human lymphocytes by in-vitro exposure of butyltins and nonortho coplanar PCBs. *Environmental Pollution* **120**: 245–253.
- Nakayama, K., Matsudaira, C., Tajima, Y., Yamada, T.K., Yoshioka, M., Isobe, T., Takahashi, S. and Tanabe, S. 2009. Temporal and spatial trends of organotin contamination in the livers of finless porpoises (*Neophocaena phocaenoides*) and their association with parasitic infection status. *Science of the Total Environment* **407**: 6173–6178.
- U.S. Environmental Protection Agency. 2004. Ambient aquatic life water quality criteria for tributyltin (TBT)-Final. USEPA, EPA 822-R-03-031, Washington, DC.

4.6 Contamination status and temporal trends of antifouling biocides (TBT, Diuron and Irgarol) in coastal areas in the Republic of Korea

Minkyu Choi¹, Byoung Seok Yoon² and Won Joon Shim³

¹ National Institute of Fisheries Science (NIFS), Busan, Republic of Korea

² Korea Marine Environment Management Corporation (KOEM), Busan, Republic of Korea

³ Korea Institute of Ocean Science and Technology (KIOST), Geoje, Republic of Korea

Biocidal additives have been used in antifouling paints for ship hulls and marine structures to prevent the immersed solid surfaces from fouling of colonizing marine organisms. Antifouling biocides can be directly introduced from ship hulls to marine environments and can cause adverse biological effects on non-target marine organisms (U.S. Environmental Protection Agency, 2004).

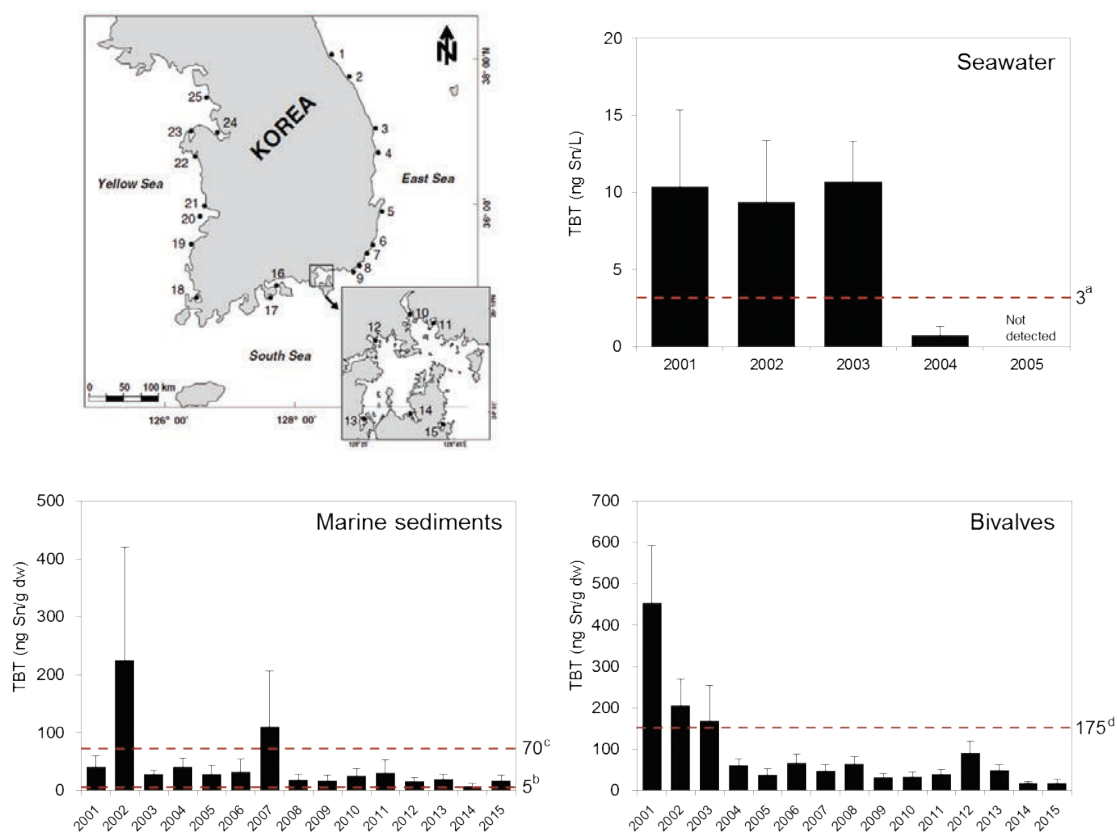


Fig. 4.6.1 Temporal trends of the mean tributyltin (TBT) concentrations in seawater, marine sediments, and bivalves at 25 sites along the coast of Korea in 2001–2015, with a comparison of screening values reported elsewhere. ^a Chronic criterion of USEPA (2004); ^bAMWQG-LT (Australian Marine Water Quality Guideline-low trigger value); ^cAMWQG-HT (high trigger value; Burton *et al.*, 2004); ^dUpper and lower ecotoxicological assessment criteria of OSPAR (2004).

Tributyltin (TBT), an antifouling agent found in seawater, bivalves and sediments, has been monitored annually at 25 sites along the coast of Korea by the Korean Ministry of Oceans and Fisheries (MOF) since 2001 (Fig. 4.6.1). As a consequence of a total ban on TBT use in Korea in 2003, the mean concentrations of TBT in seawater and bivalves have significantly decreased. However, TBT levels in sediments have not been significantly reduced due to its persistence in benthic environments. In Korea, restrictions addressing the use of TBT-based antifouling paints were partially introduced in 2000 for small boats, but their use was totally banned in 2003, and their residues on ships were banned in 2008 (MOF, 2008) following the action of the International Maritime Organization. The total ban on BT usage in Korea was found to be effective in reducing its level in seawater and biota, but longer periods are required for reduction of sedimentary TBT levels.

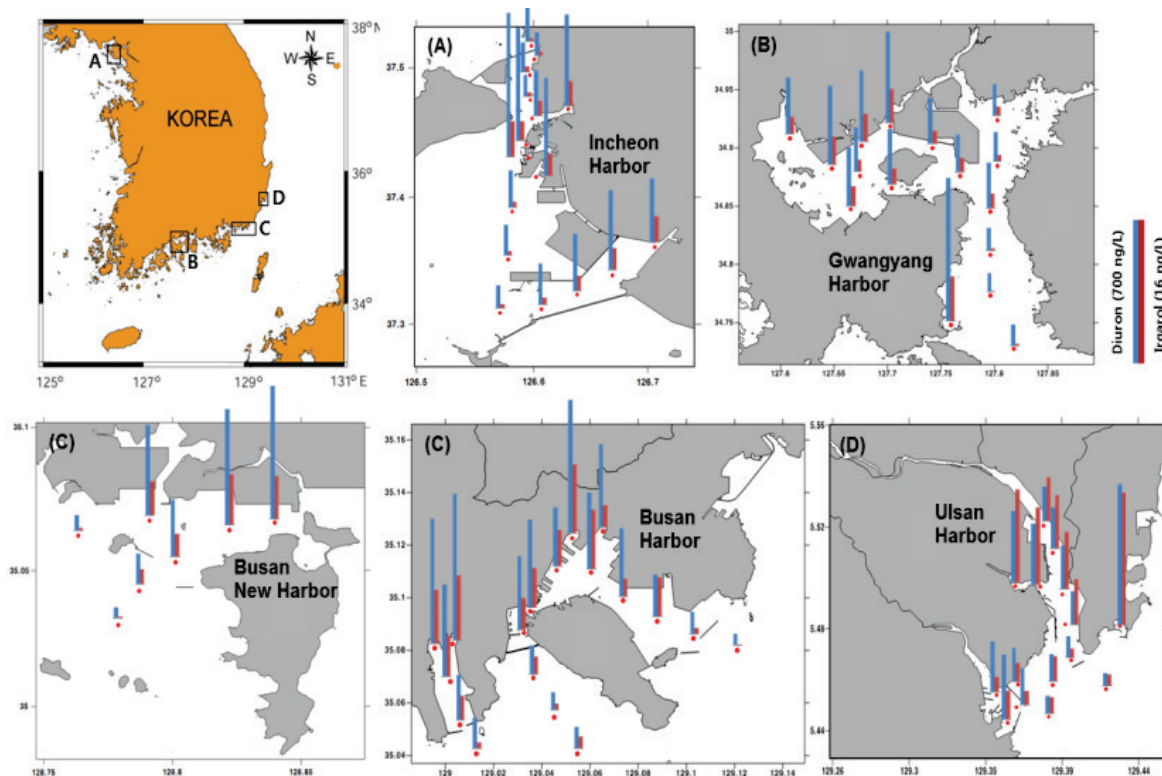


Fig. 4.6.2 Spatial distribution of Diuron and Irgarol in seawater from major industrialized harbor areas in Korea. (A) Incheon Harbor, (B) Gwangyang Harbor, (C) Busan Harbor and Busan New Harbor, and (D) Ulsan Harbor.

Following the ban of TBT-based antifouling agents, alternative biocides were introduced into the market. There is currently only limited data available regarding the occurrence of alternative antifouling agents (*i.e.*, Organotin (OT)-free antifouling biocides) in marine environments of Korea. In the 2009–2010 survey (Kim *et al.*, 2014), it was found that Diuron and Irgarol in seawater were widely distributed along the coastal areas of Korea, while the levels of pyrethroids and Dichlofluanid were below the detection limits in all sites, and Sea-Nine 211 was detected occasionally at some sites. In 2015, seawater contamination levels by Diuron and Irgarol were monitored at 71 stations from five major industrialized harbors in Korea (Fig. 4.6.2; NIFS, 2015). These antifouling chemicals were commonly detected in seawater, with concentrations ranging from 33 to 640 ng/L for Diuron and from 0.1 to 15 ng/L for

Irgarol. The current Diuron concentrations in Korean coastal water occasionally exceed the EU-predicted no effect concentration (PNEC, 200 ng/L) and UK Environmental quality standard (EQS, 100 ng/L), while Irgarol levels are not high enough to cause adverse health effects on marine organisms (EU PNEC 43.9 ng/L and UK EQS 24 ng/L). These monitoring results indicate that Diuron is an antifouling agent of concern in Korean coastal waters since the ban of TBT use.

References

- Burton, E.D., Phillips, I.R. and Hawker, D.W. 2004. Sorption and desorption behavior of tributyltin with natural sediments. *Environmental Science and Technology* **38**: 6694–6700.
- Kim, N.S., Shim, W.J., Yim, U.H., Hong, S.H., Ha, S.Y., Han, G.M. and Shin, K.H. 2014. Assessment of TBT and organic booster biocide contamination in seawater from coastal areas of South Korea. *Marine Pollution Bulletin* **78**: 201–208.
- MOF (Ministry of Oceans and Fisheries). 2008. Marine Environmental Management Act 40.
- NIFS (National Institute of Fisheries Science). 2015. Final Report - Occurrence and distribution of Diuron and Irgarol 1051 in seawater from five major harbors of Korea. NIFS, Busan, 25 pp.
- OSPAR, 2004. OSPAR/ICES workshop on the evaluation and update of background reference concentration (BRCs) and ecotoxicological assessment criteria (EACs) and how these assessment tools should be used in assessing contaminants in water, sediment and biota (Final report), 9–13 February 2004, The Hague. OSPAR Commission and ICESCIEM
- U.S. Environmental Protection Agency. 2004. Ambient aquatic life water quality criteria for tributyltin (TBT)-Final. USEPA, EPA 822-R-03-031, Washington, DC.

4.7 Contamination status and temporal trends of mercury (Hg) in mussels (*Mytilus edulis*) and oysters (*Crassostrea gigas*) from coastal areas in the Republic of Korea

Dong-Woon Hwang¹ and Yong-Woo Lee²

¹National Institute of Fisheries Science (NIFS), Busan, Republic of Korea

²Korea Marine Environment Management Corporation (KOEM), Busan, Republic of Korea

Seafoods, including marine fish and shellfish, are important sources of animal protein, nutrition, and omega-3 polyunsaturated fatty acids for people (Qiu *et al.*, 2011). Unfortunately, most people are exposed to mercury due to seafood consumption. Mercury is a highly toxic metal and occurs naturally, and is introduced into the coastal environment through various pathways such as river runoff and atmospheric deposition. In addition, the amount of mercury (Hg) released to coastal waters by human activities is increasing every year. Marine bivalves such as mussels and oysters are often used as bioindicators for monitoring metal contamination in coastal environments (Shulkin *et al.*, 2003; Alfonso *et al.*, 2013).

The Ministry of Oceans and Fisheries of the Republic of Korea (MOF) investigated the concentrations of trace metals in soft tissues of mussels (*Mytilus edulis*, $n = 271$) and oysters (*Crassostrea gigas*, $n = 108$) collected at 25 stations along the coast of Korea from 2000 to 2015. The Hg concentrations in mussels showed large spatial differences, while the Hg concentrations in oysters did not display any spatial differences during the monitoring periods. The mean Hg concentration in mussels and oysters ranged from 0.006 to 0.155 mg/kg wet weight (ww) (mean 0.020 mg/kg ww) and from 0.009 to 0.015 mg/kg ww (mean 0.011 mg/kg ww), respectively (Fig. 4.7.1). Relative to mussels, oysters have low Hg concentrations. Compared to the safety level (0.5 mg/kg ww) of Hg for shellfish applied in Korea (MFDS, 2015), Hg concentrations in mussels and oysters were below the safety level in all samples except for mussels of the Onsan coast (southeast Korea) in 2010. The highest Hg concentrations were observed in mussels and oysters near urban and industrial complexes (Onsan coast) and close to large-scale oyster farms (Garolim Bay, west coast of Korea), respectively. However, low Hg concentrations in mussels and oysters were also found near urban and industrial complexes (Jinhae Bay (south coast) for mussels and Gunsan coast (west coast) for oysters). These findings indicate that Hg concentrations in two shellfish species of Korea are widely distributed along the Korean coast and may not be dependent solely on anthropogenic activities in the coastal zone.

Mean Hg concentrations in mussels did not show any distinctive temporal trend during the monitoring periods (Fig. 4.7.2A), while mean Hg concentrations in oysters were below 0.02 mg/kg ww and were uniform over time since 2004 (Fig. 4.7.2B). Presently, it is difficult to find any reasonable cause for temporal variations in Hg concentrations in the tissues of mussels and oysters. Generally, there are distinct differences in bioaccumulation potential for trace metals between mussels and oysters. In addition, various biological and environmental factors, including changes in biogeochemical conditions and growth dilution affecting metal bioaccumulation, influence the metal concentration in tissues of shellfish (Kimbrough *et al.*, 2008; Apeti *et al.*, 2012). Therefore, more extensive and long-term monitoring for Hg in mussels and oysters is necessary to understand the temporal variation of Hg

concentrations in the tissues of shellfish to provide relevant information for a mercury management in coastal zones of Korea.

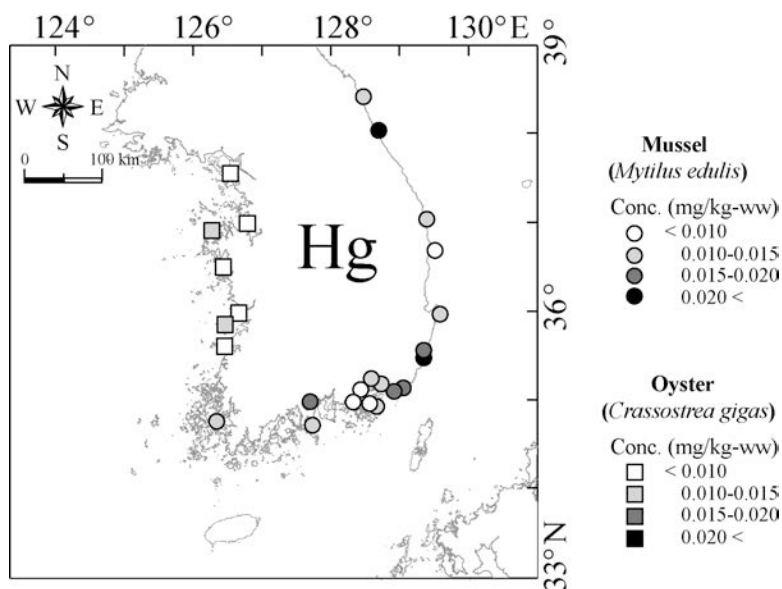


Fig. 4.7.1 Horizontal distributions of mean Hg concentration (mg/kg wet weight, ww) in the tissues of mussels and oysters collected at 25 stations along the Korean coast from 2000 to 2015.

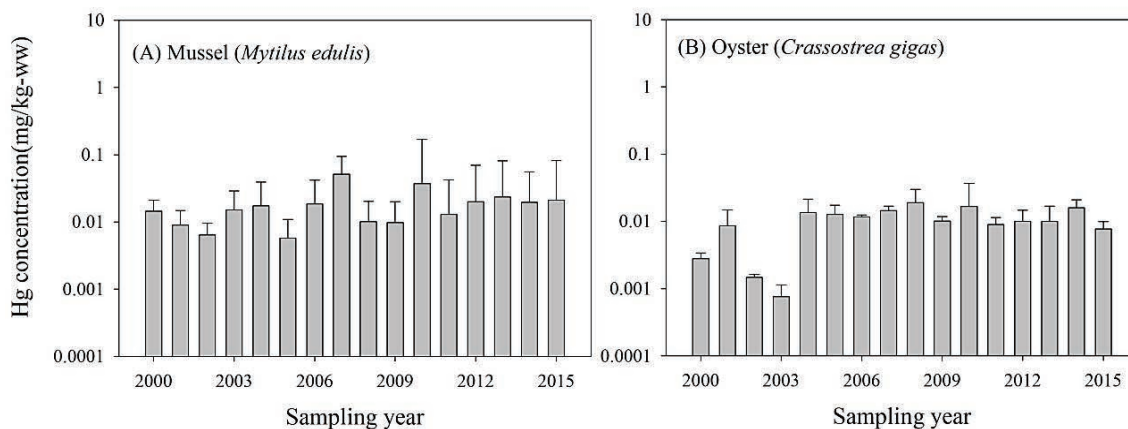


Fig. 4.7.2 Annual variations of mean Hg concentration (mg/kg ww) in the tissues of (A) mussels and (B) oysters collected at 25 stations along the Korean coast from 2000 to 2015.

References

- Alfonso, J.A., Handt, H., Mora, A., Vásquez, Y., Azocar, J. and Marcato, E. 2013. Temporal distribution of heavy metal concentrations in oysters *Crassostrea rhizophorae* from the central Venezuelan coast. *Marine Pollution Bulletin* **73**: 394–398.
- Apeti, D.A., Lauenstein, G.G. and Evans, D.W. 2012. Recent status of total mercury and methyl mercury in the coastal waters of the northern Gulf of Mexico using oysters and sediments from NOAA's mussel watch program. *Marine Pollution Bulletin* **64** : 2399–2408.
- Kimbrough, K.L., Johnson, W.E., Lauenstein, G.G., Christensen, J.D. and Apeti, D.A. 2008. An assessment of two decades of contaminant monitoring in the nation's coastal zone. NOAA Technical Memorandum NOS NCCOS 74, Silver Spring, MD, 105 pp.
- MFDS (Ministry of Food and Drug Safety). 2015. Korean Food Code – Chapter 6, A standards for marine products, 1505 pp.
- Shulkin, V.M., Presley, B.J. and Kavun, V.I. 2003. Metal concentrations in mussel *Crenomytilus grayanus* and oyster *Crassostrea gigas* in relation to contamination of ambient sediments. *Environment International* **29**: 493–502.
- Qiu, Y.W., Lin, D., Liu, J.Q. and Zeng, E.Y. 2011. Bioaccumulation of trace metals in farmed fish from South China and potential risk assessment. *Ecotoxicology and Environmental Safety* **74**: 284–293.

4.8 Contamination status and temporal trends of mercury (Hg) in sediments from coastal areas in the Republic of Korea

Dong-Woon Hwang¹ and Yong-Woo Lee²

¹National Institute of Fisheries Science (NIFS), Busan, Republic of Korea

²Korea Marine Environment Management Corporation (KOEM), Busan, Republic of Korea

Mercury (Hg) is a highly toxic and bioaccumulating trace metal. It occurs naturally at low levels in rock, soil, and natural waters including seawater. However, about half of all mercury in the natural environment originates from anthropogenic sources, including the burning of fossil fuels, waste incineration, mining and other industrial activities.

The Ministry of Oceans and Fisheries of the Republic of Korea (MOF) investigated the concentrations of trace metals in Korean coastal sediments collected from 71 stations from 2004 to 2015 ($n = 828$). Hg concentrations in coastal sediments showed large spatial differences. Mean Hg concentrations in each sampling site during the monitoring periods ranged from 0.009 to 0.466 mg/kg dry weight (dw) (mean 0.038 mg/kg dw) and the highest mean concentration was found on the Onsan coast located in the southeastern region of Korea (Fig. 4.8.1A). Compared to the threshold effect level (*i.e.*, TEL, 0.11 mg/kg dw) for Hg used in Korea as sediment quality guidelines (MOF, 2013), mean Hg concentrations were below the TEL value at all sampling sites, except for one site (Onsan coast). Most of the mean Hg concentrations were much higher in the sediments from the southeastern and northeastern coasts of Korea.

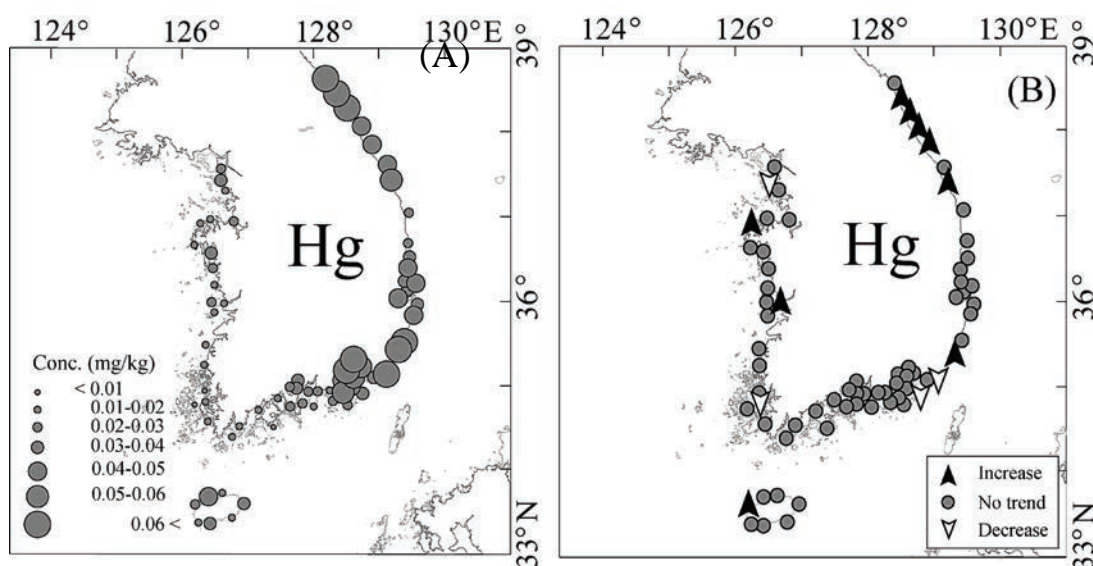


Fig. 4.8.1 Horizontal distributions (A) and temporal trends (B) of mean Hg concentration (mg/kg dry weight, dw) in coastal sediment collected at 71 stations along the Korean coast from 2004 to 2015.

Mean Hg concentrations in Korean coastal sediments generally showed a gradual increase in every year during the monitoring periods (Fig. 4.8.2). In detail, the Hg concentration in sediments did not show a temporal change at most of the sampling sites. However, Hg concentrations in some coastal sediments increased and decreased significantly at nine and four sites, respectively, as shown in Figure 4.8.1B. Although there are no special anthropogenic sources like large metropolitan areas, industrial complexes, or large-scale fish and shellfish farms along the northeastern coast of Korea, increasing trends in Hg concentrations were mainly found in the sediments from the northeastern coast. Such a trend could have been caused by atmospheric deposition of trace metals released from land-based sources in Korea and China (Hwang *et al.*, 2016). Generally, it is well known that monsoons can carry dust (*e.g.*, Asian dust) from the arid and semi-arid region of northern and northwestern China to other countries, including Korea and Japan (Ishikawa *et al.*, 1998; Kang *et al.*, 2009), and Asian dust collected in Korea was also reported to contain anthropogenic trace metals originating from some heavily industrialized Chinese cities (Lee *et al.*, 2013).

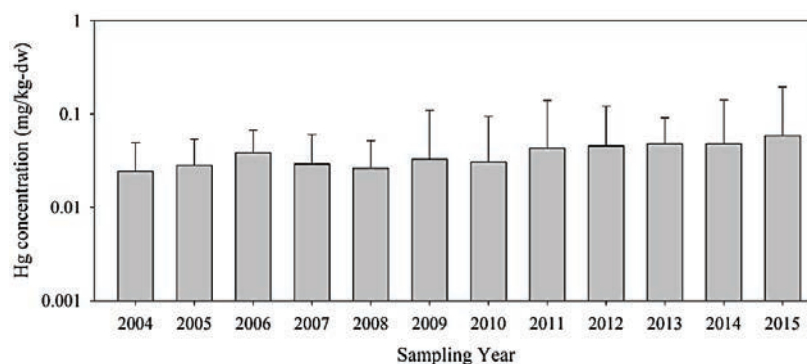


Fig. 4.8.2 Annual variation of mean Hg concentration (mg/kg dw) in coastal sediment collected at 71 stations along the Korean coast from 2004 to 2015.

On the other hand, the monitoring results for trace metals is one of the major factors to decide a marine policy and to devise a management plan for improving the coastal environment. These mercury findings have been used to manage the sediment quality of the Korean coast to implement a marine environmental policy (*e.g.*, regulation of contaminant loadings in special management areas and dredging of contaminated sediment in harbors). Thus, continuous monitoring for trace metals, including mercury in Korean coastal sediment, is necessary to understand the trend in temporal changes of mercury concentrations and to manage coastal sediment quality.

References

- Hwang, D.W., Kim, S.G., Choi, M., Lee, I.S., Kim, S.S. and Choi, H.G. 2016. Monitoring of trace metals in coastal sediments around Korean Peninsula. *Marine Pollution Bulletin* **102**: 230–239.
- Ishikawa, Y., Yoshimura, K., Mori, A., and Hara, H. 1998. High sulfate and nitrate concentrations in precipitation at Nagasaki impacted by long-distant and local sources. *Atmospheric Environment* **32**: 2939–2945.
- Kang, C.H., Kim, W.H., Ko, H.J., and Hong, S.B. 2009. Asian dust effects on total suspended particulate (TSP) compositions at Gosan in Jeju Island, Korea. *Atmospheric Research* **94**: 345–355.
- Lee, P.K., Youm, S.J., and Jo, H.Y. 2013. Heavy metal concentrations and contamination levels from Asian dust and identification of sources: A case-study. *Chemosphere* **91**: 1018–1025.
- MOF (Ministry of Oceans and Fisheries). 2013. Marine Environment Management Act – Articles 8: Marine Environmental Standards. MOF Notice No. 2013-186.

4.9 Contamination status and temporal trends of lead (Pb) in mussels (*Mytilus edulis*) and oysters (*Crassostrea gigas*) from coastal areas in the Republic of Korea

Dong-Woon Hwang¹ and Yong-Woo Lee²

¹ National Institute of Fisheries Science (NIFS), Busan, Republic of Korea

² Korea Marine Environment Management Corporation (KOEM), Busan, Republic of Korea

Lead (Pb) is one of the trace metals that occur naturally in the earth's crust. Pb concentrations in the natural environment have significantly increased worldwide over the past century because of anthropogenic activities such as manufacturing processes, paint and pigment applications, and fossil fuel burning (Kimbrough *et al.*, 2008). Marine bivalves have been used extensively as bioindicators to assess the pollution of the coastal environment because of their sedentary behavior, broad distribution, abundance, and high ability to accumulate metals (Weng and Wang, 2014).

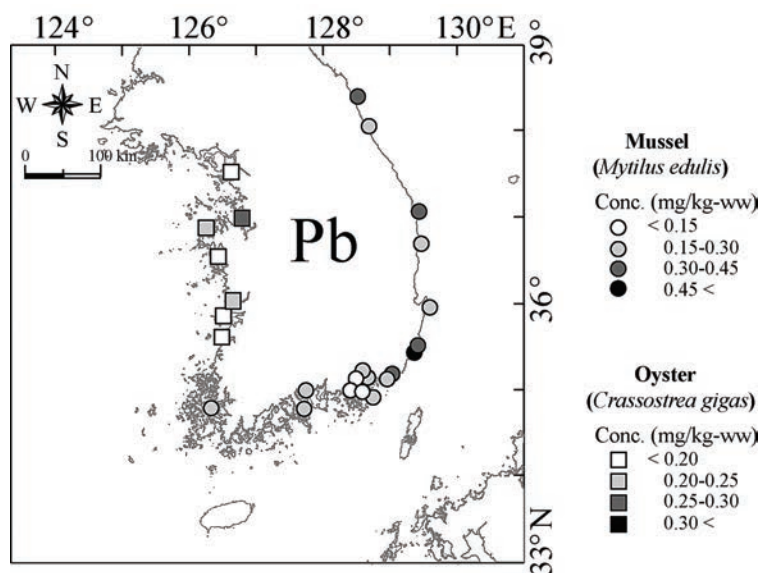


Fig. 4.9.1 Horizontal distributions of mean Pb concentration (mg/kg wet weight, ww) in the tissues of mussels and oysters collected at 25 stations along the Korean coast from 2000 to 2015.

The concentration of trace metals in the soft tissues of mussels (*Mytilus edulis*, $n = 270$) and oysters (*Crassostrea gigas*, $n = 107$) collected at 25 stations along the coast of Korea were investigated by the Ministry of Oceans and Fisheries of the Republic of Korea (MOF) from 2000 to 2015. The Pb concentrations in two shellfish species showed large spatial differences during the monitoring period. The mean Pb concentration in mussels and oysters of each sampling site ranged from 0.10 to 0.83 mg/kg wet weight (ww) (mean 0.27 mg/kg ww) and from 0.14 to 0.28 mg/kg ww (mean 0.19 mg/kg ww), respectively (Fig. 4.9.1). Relative to mussels, oysters had low Pb concentrations. Compared to the

safety level (2.0 mg/kg ww) of Pb for shellfish applied in Korea (MFDS, 2015), the Pb concentrations in mussels and oysters were below the safety level in all samples except for mussels of the Onsan coast (southeast Korea) in 2014. The highest mean Pb concentrations were found in mussels and oysters near urban and industrial areas (Onsan coast for mussels and Asan coast (northwest Korea) for oysters). However, mussels and oysters at some sampling sites near large urban centers and industrial complexes showed low Pb concentrations. This finding implies that Pb concentrations in two shellfish species of Korea are not significantly dependent on anthropogenic activities in the coastal zone.

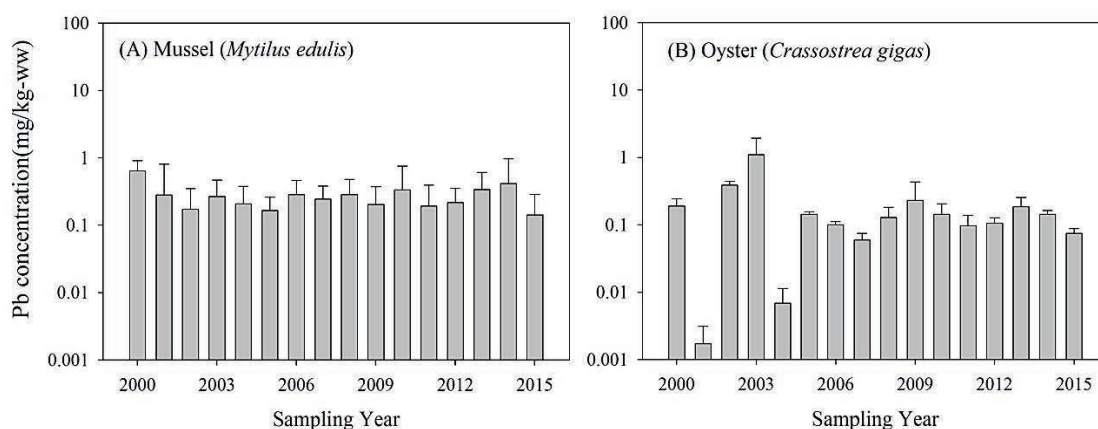


Fig. 4.9.2 Annual variations of mean Pb concentration (ww) in the tissues of (A) mussels and (B) oysters collected at 25 stations along the Korean coast from 2000 to 2015.

Mean Pb concentrations in mussels were below 0.5 mg/kg ww except in 2000 and exhibited little temporal variation during the monitoring periods (Fig. 4.9.2A), while mean Pb concentrations in oysters showed large annual variation (Fig. 4.9.2B). Presently, it is difficult to find the main cause for these variations because there are distinct differences in bioaccumulation potential between mussels and oysters for trace metals (Kimbrough *et al.*, 2008) and the metal accumulation in the tissues of bivalves depends on various biological (*i.e.*, size, age, clearance rate, reproductive cycle, *etc.*) and environmental factors (*i.e.*, temperature, salinity, pH, and dissolved organic matter) (Langston and Spence, 1995). Thus, more extensive interdisciplinary studies are necessary to understand the temporal variations of Pb concentrations in the tissues of mussels and oysters on the Korean coast.

References

- Kimbrough, K.L., Johnson, W.E., Lauenstein, G.G., Christensen, J.D. and Apeti, D.A. 2008. An assessment of two decades of contaminant monitoring in the nation's coastal zone. NOAA Technical Memorandum NOS NCCOS 74, 105 pp., Silver Spring, MD.
- Langston, W.J. and Spence, S.K. 1995. Biological factors involved in metal concentrations observed in aquatic organisms, pp. 407–478 *in*: Metal Speciation and Bioavailability in Aquatic Systems *edited by* A. Tessier and D.R. Turner, John Wiley and Sons, Chichester.
- MFDS (Ministry of Food and Drug Safety). 2015. Korean Food Code – Chapter 6 A standards for marine products, 1505 pp.
- Weng, N. and Wang, W.X. 2014. Variations of trace metals in two estuarine environments with contrasting pollution histories. *Science of the Total Environment* **485-486**: 604–614.

4.10 Contamination status and temporal trends of lead (Pb) in sediments from coastal areas in the Republic of Korea

Dong-Woon Hwang¹ and Yong-Woo Lee²

¹National Institute of Fisheries Science (NIFS), Busan, Republic of Korea

²Korea Marine Environment Management Corporation (KOEM), Busan, Republic of Korea

Human activities associated with urbanization and industrialization influence the levels and distributions of toxic metals in the marine environment. Lead (Pb) in coastal environments mainly originates from anthropogenic sources, including the combustion of fossil fuels and ore processes, rather than natural processes such as the weathering and erosion of soils and rocks.

The Ministry of Oceans and Fisheries of the Republic of Korea (MOF) carried out monitoring and field surveys for trace metals in Korean coastal sediments collected from 71 stations from 2004 to 2015 ($n = 833$). The Pb concentrations in coastal sediments showed large spatial differences during the monitoring period. Mean Pb concentrations in each sampling site ranged from 17.1 to 92.2 mg/kg dry weight (dw) (mean 32.3 mg/kg dw) and the highest mean Pb concentration was found on the Onsan coast located in the southeastern region of Korea (Fig. 4.10.1A). Compared to the threshold effect level (*i.e.*, TEL, 44.0 mg/kg dw) for Pb applied in Korea as sediment quality guidelines (MOF, 2013), the mean Pb concentrations were below the TEL value at all sampling sites, except for two sites (Onsan coast and Masan Bay (southeastern coast)). In addition, most of the mean Pb concentrations were significantly higher in sediments from the southeastern coast of Korea where large metropolitan areas, industrial complexes, and fish and shellfish farms are heavily concentrated. Thus, these results indicate that human activities in the Korean coastal zone significantly influence the distribution of Pb concentrations in the coastal sediment (Hwang *et al.*, 2016).

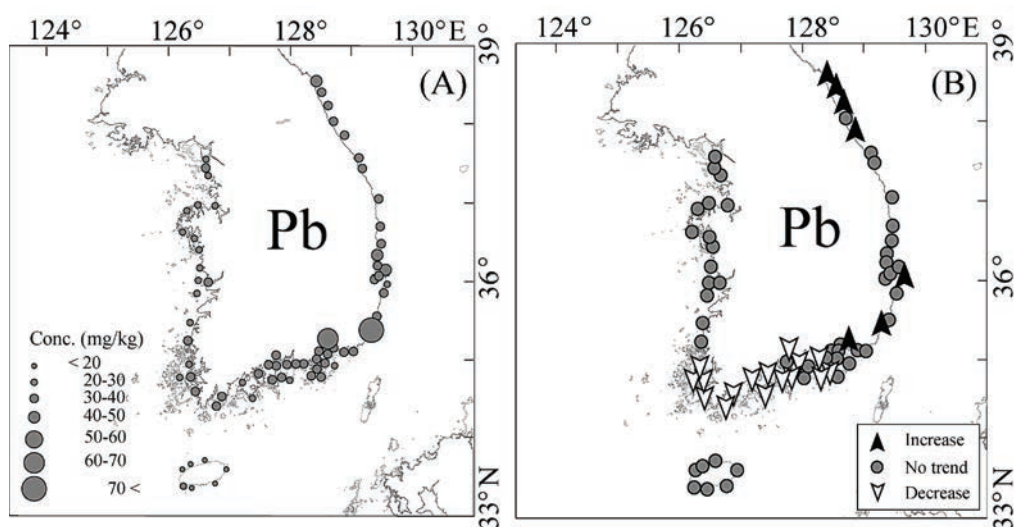


Fig. 4.10.1 Horizontal distributions (A) and temporal trends (B) of mean Pb concentration (mg/kg dry weight, dw) in coastal sediment collected at 71 stations along the Korean coast from 2004 to 2015.

Mean Pb concentrations in Korean coastal sediments did not show any marked annual variation during the monitoring periods (Fig. 4.10.2). In detail, there were no discernable temporal trends of Pb concentration in sediments at most of the sampling sites. However, Pb concentrations in sediments increased and decreased significantly at seven and sixteen sites, respectively, as shown in Figure 1B. Especially, the decreasing trends in Pb concentrations were mainly found in sediments from the southern coast although there are many anthropogenic sources, such as metropolitan areas, industrial complexes, and fish and shellfish farms along the southern coast. Such a trend could have resulted from sustainable coastal management fostered by the Korean Government because most parts of the southern coast have been designated special management areas, marine environment conservations areas, and marine protected areas (Hwang *et al.*, 2016).

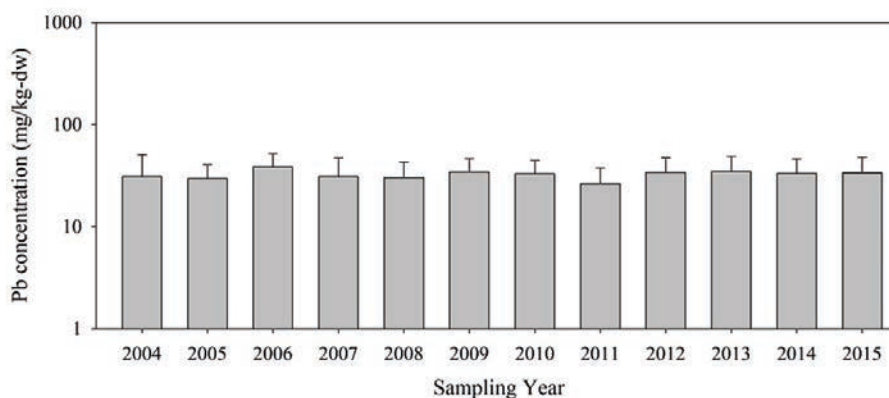


Fig. 4.10.2 Annual variation of mean Pb concentration (mg/kg dw) in coastal sediment collected at 71 stations along the Korean coast from 2004 to 2015.

On the other hand, these findings have been used to manage Pb concentrations in Korean coastal sediment, and to decide marine policy (*e.g.*, regulation of contaminant loadings in special management areas, dredging of contaminated sediment in harbors, and designation of marine protected areas) for improving the coastal environment. Also, the monitoring results for trace metals will be one of the major factors used to determine if a region needs a special management plan to implement a marine environmental policy. Therefore, it is necessary to do continuous monitoring for trace metals, including lead, in Korean coastal sediments.

References

- Hwang, D.W., Kim, S.G., Choi, M., Lee, I.S., Kim, S.S. and Choi, H.G. 2016. Monitoring of trace metals in coastal sediments around Korean Peninsula. *Marine Pollution Bulletin* **102**: 230–239.
- MOF (Ministry of Oceans and Fisheries). 2013. Marine Environment Management Act – Articles 8: Marine Environmental Standards. MOF Notice No. 2013-186.

4.11 Contamination status and temporal trends of marine debris at beaches in the Republic of Korea

Sunwook Hong¹ and Won Joon Shim²

¹ Korea Marine Litter Institute, Our Sea of East Asia Network (OSEAN), Tongyeong, Republic of Korea

² Korea Institute of Ocean Science and Technology (KIOST), Geoje, Republic of Korea

Marine debris is a pollution issue of global concern. A total of 20 beaches have been monitored along the coasts of the Republic of Korea since 2008 with governmental support (Fig. 4.11.1a; Hong *et al.*, 2014). A 100-m-long transect located with GPS coordinates at each beach has been surveyed by trained non-governmental organizations every two months (*i.e.*, at the end of odd numbered months ± 5 days) following the exactly same protocol. All debris larger than 2.5 cm in diameter between the low tide mark and the beginning of vegetation or artificial barriers was collected. The debris was classified into 12 categories such as plastics other than Styrofoam, metal, wood, cloth, among others and measured in terms of number, weight and volume. Individual debris collected was classified into 94 items to identify abundant debris and contributing sources. Before the survey (*i.e.*, January 2008), all existing marine debris on the selected beaches was removed in order to remove any possible effects accumulated during the past years.

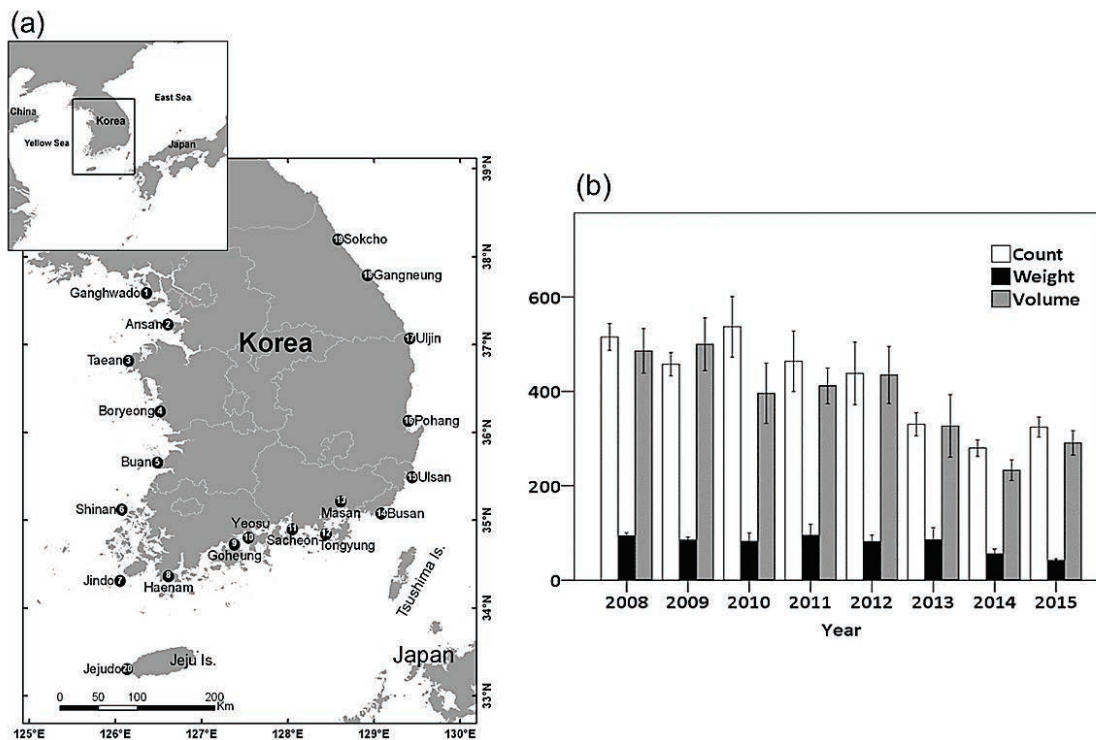


Fig. 4.11.1 (a) Location of marine debris monitoring sites in Korea since 2008 (Hong *et al.*, 2014) and (b) temporal trend of marine debris quantities over 8 years (mean \pm SE) (Unit: counts/100 m; black: kg/100 m; grey: 10^{-3} m³/100 m).

Hong *et al.* (2014) reported the level of marine debris pollution and identified its main sources based on the analysis of the first 2-year surveys (*i.e.*, 220 surveys in total from March 2008 to November 2009). The mean quantities of marine debris were estimated at 481.9 (± 267.7 SD) count/100 m for number, 86.5 (± 78.6 SD) kg/100 m for weight, and 0.48 (± 0.38 SD) m³/100 m per volume. Plastics and Styrofoam occupied the majority of debris composition in terms of number (66.7%) and volume (62.3%). Wood (timber) was the major material in weight. Main sources of debris were estimated to be generated from fishing activities (51.3%). Styrofoam buoy debris was the most frequent item, potentially originating from aquaculture. The sources have been also supported by other studies (Jang *et al.*, 2014a, b).

According to the recent preliminary analysis, the number, weight, and volume of marine debris has been decreasing significantly over 8 years, from 2008 to 2015, as shown in Figure 4.11.1b (MOF and KOEM, 2015). Decreasing inputs from land-based and/or sea-based sources, active cleaning by local authorities or residents, less intense rainfall and wind, and effective policies could be factors affecting the overall decrease in the amount of beach debris. There has been observed seasonalities in which the marine debris increases from summer to early fall (late July to late September) and decreases in winter. Typhoons, wind direction change, or beachgoer activities in the summer holiday season may affect the result. Further analysis should be done to identify which factors have contributed to the decrease and seasonality more significantly.

Marine debris has negatively affected wildlife because of entanglements or ingestion (Hong *et al.*, 2013). Forty-five cases collected showed that at least 21 species, including 18 species of birds, two species of mammals, and one species of crustacean, have ingested or been entangled by marine debris, especially recreational fishing gears (*e.g.*, fishing hooks, monofilament lines, sinkers) and commercial fishing gears (*e.g.*, nets, ropes, traps). Five threatened or protected species were included among them: black-faced spoonbill *Platalea minor*, whooper swan *Cygnus cygnus*, greater painted snipe *Rostratula benghalensis*, finless porpoise *Neophocaena phocaenoides*, and water deer *Hydropotes inermis*.

The studies on quantities of marine debris and their impacts on wildlife show that fishing gears (both commercial and recreational) should be prioritized when managing marine debris in Korea.

References

- Hong, S., Lee, J., Jang, Y.C., Kim, Y.J., Kim, H.J., Han, D., Hong, S.H., Kang, D. and Shim, W.J. 2013. Impacts of marine debris on wild animals in the coastal area of Korea. *Marine Pollution Bulletin* **66**: 117–124.
- Hong, S., Lee, J., Kang, D., Choi, H.-W. and Ko, S.-H. 2014. Quantities, composition, and sources of beach debris in Korea from the results of nationwide monitoring. *Marine Pollution Bulletin* **84**: 27–34.
- Jang, Y.C., Lee, J., Hong, S., Mok, J.Y., Kim, K.S., Lee, Y.J., Choi, H.-W., Kang, H. and Lee, S. 2014a. Estimation of the annual flow and stock of marine debris in South Korea for management purposes. *Marine Pollution Bulletin* **86**: 505–511.
- Jang, Y.C., Lee, J., Hong, S., Lee, J.S., Shim, W.J. and Song, Y.K. 2014b. Sources of plastic marine debris on beaches of Korea: More from the ocean than the land. *Ocean Science Journal* **49**: 151–162.
- MOF (Ministry of Oceans and Fisheries), KOEM (Korea Marine Environment Management Corporation), 2015. 2015 National Marine Debris Monitoring in Korea, KOEM, 186 pp. (in Korean)

5 Russia

5.1 *The human impact on the mercury accumulation in modern sediments of Amur Bay (the Sea of Japan)*

Kirill Aksentov

V.I. Il'ichev Pacific Oceanological Institute, Far Eastern Branch, Russian Academy of Science, Vladivostok, Russia

Studying the processes influencing the distribution and migration of mercury in the environment is of paramount importance because of its high toxicity. Since the onset of the industrial period, anthropogenic emissions of mercury have increased and its global cycling has been significantly altered (Schuster *et al.*, 2002; Fitzgerald *et al.*, 2007). The Amur Bay has been being exposed to the intense anthropogenic influence since the middle of the 20th century. The sources of pollutants are discharges from industrial enterprises located in the Razdol'naya River basin and on the Murav'ev-Amurskii Peninsula (Vladivostok city). This study investigates the reconstruction of mercury accumulation in bottom sediments of Amur Bay.

Mercury in sediment cores

The sediment samples were collected by the small gravity core GOIN-1.5 in the northern part of Amur Bay and the Zolotoi Rog Bay (also known as Golden Horn Bay, Vladivostok). Total mercury concentration in samples (dry weight, dw) was determined at 50°C by atomic absorption spectroscopy on an RA-915+ analyzer equipped with an RP-91C (Lumex Ltd., St. Petersburg, Russia) pyrolysis system. The accuracy of the method was tested using a Certified Reference Material Marine Sediments (MESS-3 and PACS-2) (Aksentov, 2013).

Mercury content in surficial sediments of the northern part Amur Bay ranged from 50 to 760 ppb dw with an average value of 120 ppb dw (Polyakov *et al.*, 2008). Higher concentrations were found in Zolotoi Rog Bay (2500 ppb) (Aksentov, 2013). The modern sedimentation rate of 4.1 mm/year was determined in core I08-3 based on unsupported ^{210}Pb (Kalugin *et al.*, 2015). Vertical mercury distribution shows the stages of its accumulation in sediments associated with the impact of increased human activities (Fig. 5.1.1).

Cleanup of the Zolotoi Rog Bay and dumping of contaminated sediments in the Amur Bay is demonstrated by mercury concentrations in cores M06-34 and I07-8, respectively (Fig. 5.1.1). The baseline levels of mercury, estimated from core horizons and deposited before the sediments were impacted by human activities, are in the order of 30 ppb dw. There are also bottom sediments with high mercury content located near natural sources. Sand in the wave-cut terrace along the abrasion bench in

Cape Nizkii Popov Island (~20 km south of Zolotoi Rog) contains mercury concentrations ranging 1000–2500 ppb dw (Aksentov, 2015). At this point, these concentrations are influenced by the deep Murav'ev fault. Nevertheless, it is not widespread. In this context, bottom sediments of Amur Bay are polluted at different degrees. Development in the Southern Primorye has increased the accumulation of mercury in the bottom sediments.

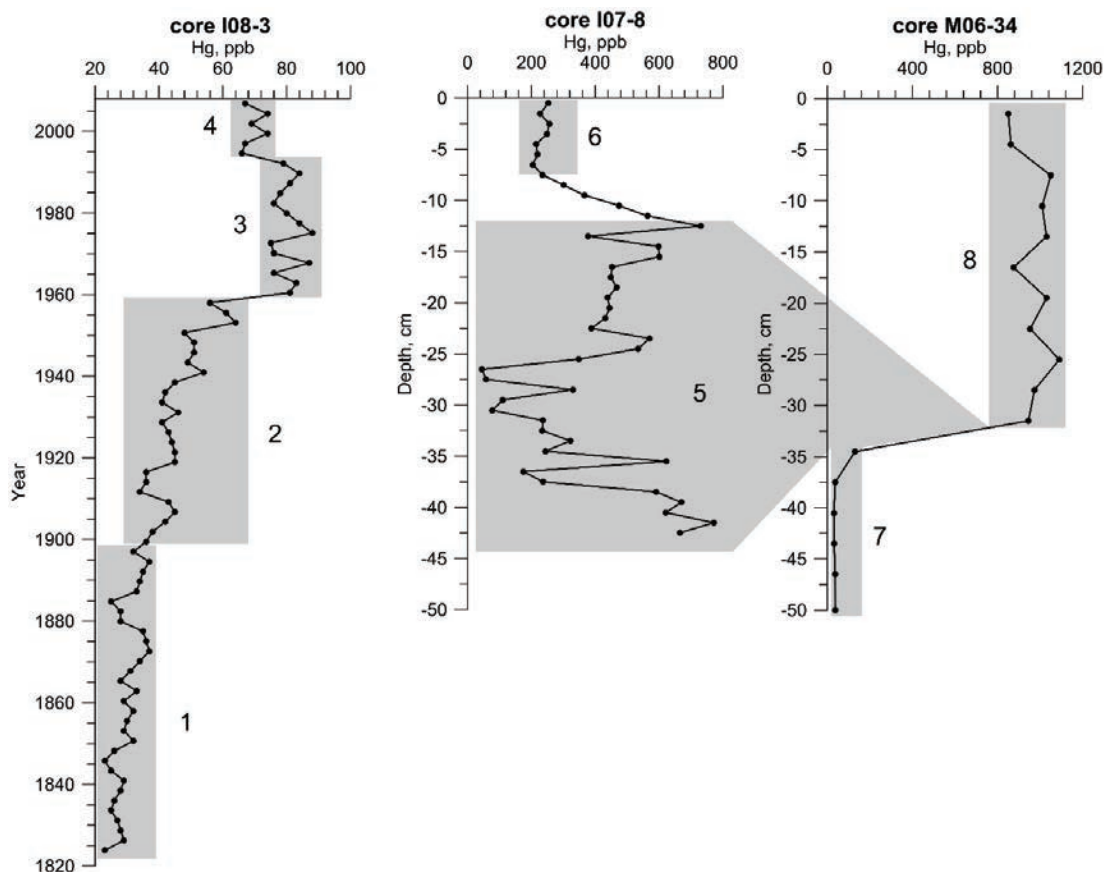


Fig. 5.1.1 Vertical profiles of total mercury (Hg) concentrations (dry weight, dw) in the sediment cores from the Amur Bay (I08-3, I07-8) and the Zolotoi Rog Bay (M06-34). 1, 7 – background concentration; 2 – moderate impact, 3, 5 – intensive contamination; 4, 6, 8 – recent level. I08-3 ($43^{\circ} 10,743' N 131^{\circ} 48,192' E$); I07-8 ($43^{\circ} 05,373' N 131^{\circ} 50,335' E$); M06-34 ($43^{\circ} 06,105' N 131^{\circ} 52,850' E$).

References

- Aksentov, K.I. 2013. Mercury in bottom sediments of the Peter the Great Bay. Ph.D. Thesis, V.I. Il'ichev Pacific Oceanological Institute, FEB RAS, Vladivostok, 140 pp. (in Russian)
- Aksentov, K.I. 2015. Mercury in the sea water of the Amur Bay (the Sea of Japan): Recent content and geochemical processes. *Russian Meteorology and Hydrology* **40**: 606–611.
- Fitzgerald, W.F., Lamborg, C.H. and Hammerschmidt, C.R. 2007. Marine biogeochemical cycling of mercury. *Chemical Reviews* **107**: 641–62.

- Kalugin, I., Astakhov, A., Darin, A. and Aksentov, K. 2015. Anomalies of bromine in the estuarine sediments as a signal of floods associated with typhoons. *Chinese Journal of Oceanology and Limnology* **33**:1489–1495.
- Polyakov, D.M., Aksentov, K.I. and Ivanov, M.V. 2008. Mercury in the bottom sediments of the marginal filter of the Razdol'naya River, Amur Bay. *Geochemistry International* **46**: 614–621.
- Schuster, P.F., Krabbenhoft, D.P., Naftz, D.L., Cecil, L.D., Olson, M.L., Dewild, J.F., Susong, D.D., Green, J.R. and Abbott M.L. 2002. Atmospheric mercury deposition during the last 270 years: a glacial ice core record of natural and anthropogenic sources. *Environmental Science and Technology* **36**: 2303–2310.

5.2 *Inter-annual dynamics of Pb concentrations in bottom sediments of Peter the Great Bay (the Sea of Japan) around Vladivostok, Russia*

Alexander V. Sevastianov, Tatyana S. Lishavskaya and Tatyana A. Belan

Far Eastern Regional Hydrometeorological Research Institute, Vladivostok, Russia

For thousands of years the presence of humans caused no harm to the coastal environment. Fishing and other forms of exploitation resulted in local environmental changes only in the coastal zone. By the end of the 20th century, however, the condition of pristine habitats and communities has changed due to the introduction of a large number of anthropogenic chemicals into the marine environment. The habitats and communities are now characterized by exhibiting low quality and impaired quantitative parameters.

Vladivostok, situated along the Peter the Great Bay coastline (Fig. 5.2.1), is one of the main seaports in eastern Russia affected by high anthropogenic pressure from human populations (about 600,000 individuals), industry, and transportation. The main sources of pollution near Vladivostok are municipal and industrial waste waters, urban runoff, marine transportation, and dredged materials. As a result of chemical loads and run-off input (*i.e.*, including nutrients and dissolved organic matter) into the marine-coastal zone, the vicinity near Vladivostok is undergoing chronic anthropogenic impacts.

For this report, three areas with different pollution levels were detected in Peter the Great Bay by Principal Components Analysis. Concentrations of petroleum hydrocarbons, trace metals (*i.e.*, lead, Pb) and chlorinated hydrocarbons in bottom sediments were used to assess the degree of pollution.

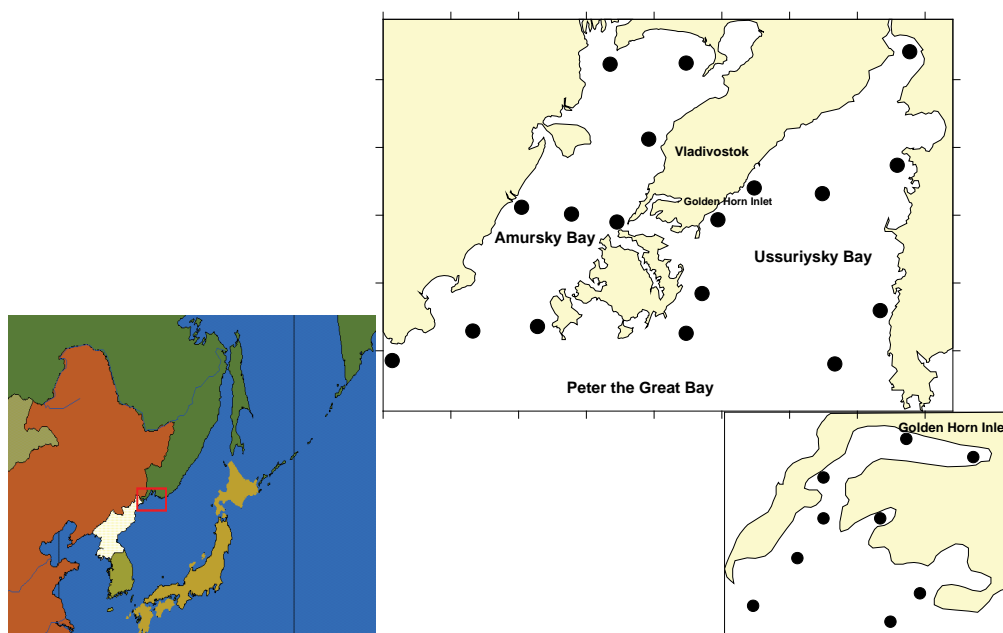


Fig. 5.2.1 Map illustrating the spatial sampling design in Peter the Great Bay, Vladivostok, Russia.

Golden Horn Inlet (Vladivostok inner harbor; Fig. 5.2.1) was considered to be extremely polluted by lead, while Amursky Bay exhibited high to moderate levels of sediment contamination by lead. These locations are semi-closed coastal areas showing low hydrodynamic processes and containing silt sediments with a high concentration of total organic carbon (up to 127.4 mg/kg dry weight, dw), petroleum hydrocarbons (up to 2.83 mg/kg dw), and trace metals. Moderate pollution levels were observed in bottom sediments from Ussuriysky Bay. For example, the inter-annual dynamics of Pb concentrations (mg/kg dw) in bottom sediments in the different sampling locations of Peter the Great Bay are shown in Figures 5.2.2–5.2.4. In general, no discernable trends of Pb sediment concentrations over time were observed for each specific area.

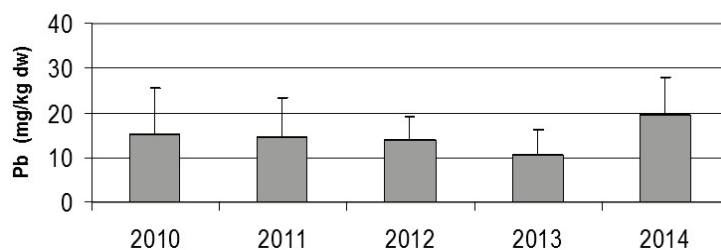


Fig. 5.2.2 Inter-annual dynamics of Pb concentrations (mg/kg dry weight, dw) in bottom sediments of Amursky Bay.

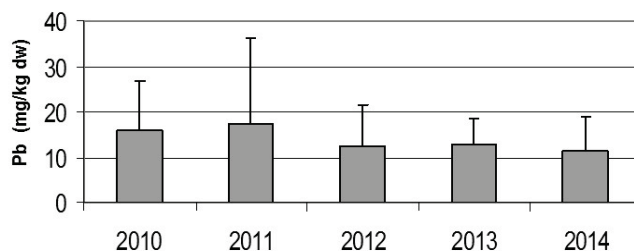


Fig. 5.2.3 Inter-annual dynamics of Pb concentrations (mg/kg dw) in bottom sediments of Ussuriysky Bay.

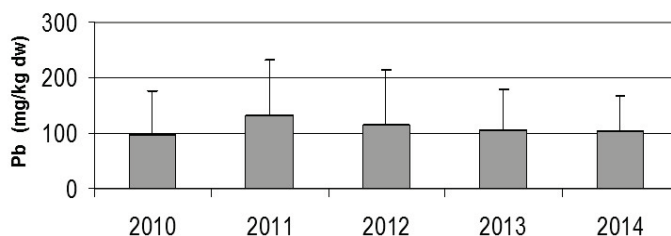


Fig. 5.2.4 Inter-annual dynamics of Pb concentrations (mg/kg dw) in bottom sediments of Golden Horn Inlet.

The Pb content in Amursky Bay and Ussuriysky Bay does not exceed the threshold level (45–85 mg/kg), whereas in Golden Horn Inlet the maximum Pb content in bottom sediments (131.68 mg/kg) was higher than this value.

5.3 Organochlorine pesticides in tissues of marine mammals from the western part of the Bering Sea, Russia

Olga Lukyanova¹ and Vasiliy Tsygankov²

¹ Pacific Research Fisheries Centre (TINRO-Centre), Vladivostok, Russia

² Far Eastern Federal University, Vladivostok, Russia

Concentrations of persistent organochlorine pesticides (OCP) were measured in the liver and muscles of grey whales (*Eschrichtius robustus*) and Pacific walrus (*Odobenus rosmarus divergens*). The tissue samples were collected from male and female specimens of different ages hunted by local indigenous people in 2010 and 2011 within the coastal waters of Lorino village (Mechigmsky Bay, Bering Sea). The International Whaling Commission (IWC) recognises the exclusive rights of indigenous people of Chukotka and Alaska to hunt grey whales as a means of supporting their traditional lifestyle. Here, we report the results of analysis of α -, β -, and γ -isomers of hexachlorocyclohexane (HCH), dichlorodiphenyl-trichloroethane (DDT), and its metabolites (1,1-dichloro-2,2-bis(p-chlorophenyl)ethylene or DDE, and 1,1-dichloro-2,2-bis(p-chlorophenyl)ethane or DDD) measured in seven specimens of grey whales and eight specimens of Pacific walruses.

A total of seven grey whales, including four males and three females, and a total of eight Pacific walruses, including five males and three females, were hunted by local indigenous people during the summers of 2010 and 2011 in the Mechigmsky Bay (Fig. 5.3.1). The concentrations of OCP in liver and muscle tissues were analyzed by gas chromatography–mass spectrometry (GC–MS) using a Shimadzu gas chromatograph mass spectrometer GCMS-QP2010Ultra at the Pacific Research Fisheries Centre (TINRO-Centre, Vladivostok).

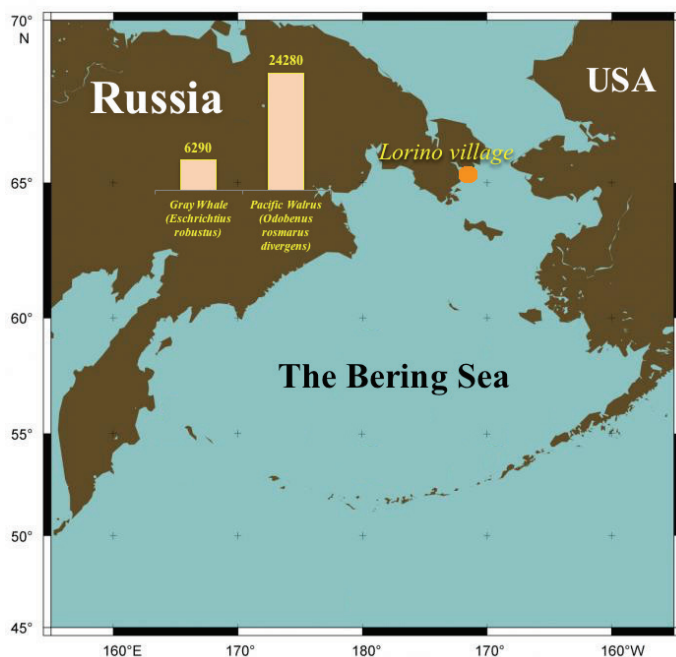


Fig. 5.3.1 Sampling location of grey whales and Pacific walruses in the Bering Sea. Bars indicate median concentration of total organochlorine pesticides (OCP) in whale and walrus liver (ng/g lipid weight).

OCP concentrations in grey whale tissues

Organochlorine pesticides were found in all samples analyzed for grey whales. In muscle tissue, the total concentration of OCP (Σ HCH + Σ DDT) ranged from 297 to 3581 ng/g lipid weight (lw), while in liver tissue it ranged from 769 to 13,808 ng/g lw. There was no statistically significant difference in OCP concentrations in muscles of males compared to females; however, the concentration of OCP in males was slightly higher for all detected compounds. Also, the concentration of α -HCH was significantly ($p = 0.05$) higher in the liver of male whales compared to females. β -HCH concentrations were significantly higher in liver tissue compared to muscle tissue ($p = 0.002$) for both genders.

When comparing the medians of the concentrations of Σ HCH, Σ DDT, and Σ OCP, only the Σ OCP concentration was significantly higher in liver compared to muscle tissue ($p = 0.048$). The total concentrations of Σ HCH and Σ DDT were also higher in liver tissue compared to muscles, although the differences were not statistically significant.

OCP concentrations in Pacific walrus tissues

The total OCP concentrations (Σ HCH + Σ DDT) in the liver of walrus ranged from 4900 to 90,300 ng/g lw. These concentrations were substantially higher compared to muscle tissue, where the total concentration of OCP ranged from 200 to 5700 ng/g lw. The total concentrations of all constituents measured were higher in females compared to males except for α -HCH, although the difference was not statistically significant. For the liver tissue, concentrations of all pesticides were higher in females, except for β -HCH, which was higher in males, but these differences were also statistically insignificant.

Concentrations of two constituents, *i.e.*, α -HCH and DDT, were significantly higher in liver compared to muscle tissue ($p = 0.016$ and $p = 0.021$ for males and females, respectively). For the other compounds, the differences were statistically insignificant. The γ -HCH and DDE concentrations were higher in liver tissue, whereas the β -HCH concentration was higher in muscles.

A comparison of the median concentrations of Σ HCH, Σ DDT, and Σ OCP concentrations in muscle tissue *versus* liver shows the prevalence of the pollutants in liver with all the differences being statistically significant ($p = 0.001$ – 0.036).

References

- Tsygankov, V., Boyarova, M.D. and Lukyanova, O.N. 2015. Bioaccumulation of persistent organochlorine pesticides (OCPs) by gray whale and Pacific walrus from the western part of the Bering Sea. *Marine Pollution Bulletin* **99**: 235–239, DOI: 10.1016/j.marpolbul.2015.07.020.
- Tsygankov, V.Y., Boyarova, M.D., Lukyanova, O.N. and Khristoforova, N.K. 2017. Bioindicators of organochlorine pesticides in the Sea of Okhotsk and the Western Bering Sea. *Archives of Environmental Contamination and Toxicology* **73**: 176–184, <https://doi.org/10.1007/s00244-017-0380-2>.

5.4 Microplastic monitoring in the coastal area of the Peter the Great Bay (Sea of Japan), Russia

Yana Blinovskaya¹ and Nikolai Kozlovskii²

¹ Sea Protection and Shelf Development Institute, Maritime State University of G.I. Nevelskoy, Vladivostok, Russia

² Pacific Geographical Institute, Far Eastern Branch, Russian Academy of Sciences, Vladivostok, Russia

Microplastic pollution (particles smaller than 5 mm in size) has been recognized as a major threat to marine ecosystems in Russia (Blinovskaya *et al.*, 2020). Analysis of microplastics was initiated in Peter the Great Bay in 2014 by the Sea Protection and Shelf Development Institute (Maritime State University of G.I. Nevelskoy). Here we report the results of microplastic analysis in nearshore surface water in Peter the Great Bay (Sea of Japan/East Sea) conducted from 2014 to 2017. The sites sampled during the survey program included industrialized and remote areas, as well as recreational and aquaculture sites. Also, the sites varied in terms of hydrodynamics. The results show that two areas, the Tumen River estuary and Vladivostok area, had the highest level of microplastics compared to all other areas analyzed during the study (Blinovskaya *et al.*, 2020).

In 2014, the surface water (<50 cm from the surface) was sampled in the Vladivostok area at 16 sites (Fig. 5.4.1). Field duplicates were collected and analyzed for each sample. Plankton/neuston nets with 0.1 mm mesh were used during the study; therefore, the minimum size of microplastic particles that could be captured is 0.1 mm. Analysis showed that the biggest size of fragments detected was 3,313 × 2,373 mm. The presence of microplastics was detected at five sites located at the mouth of the Vtoraya

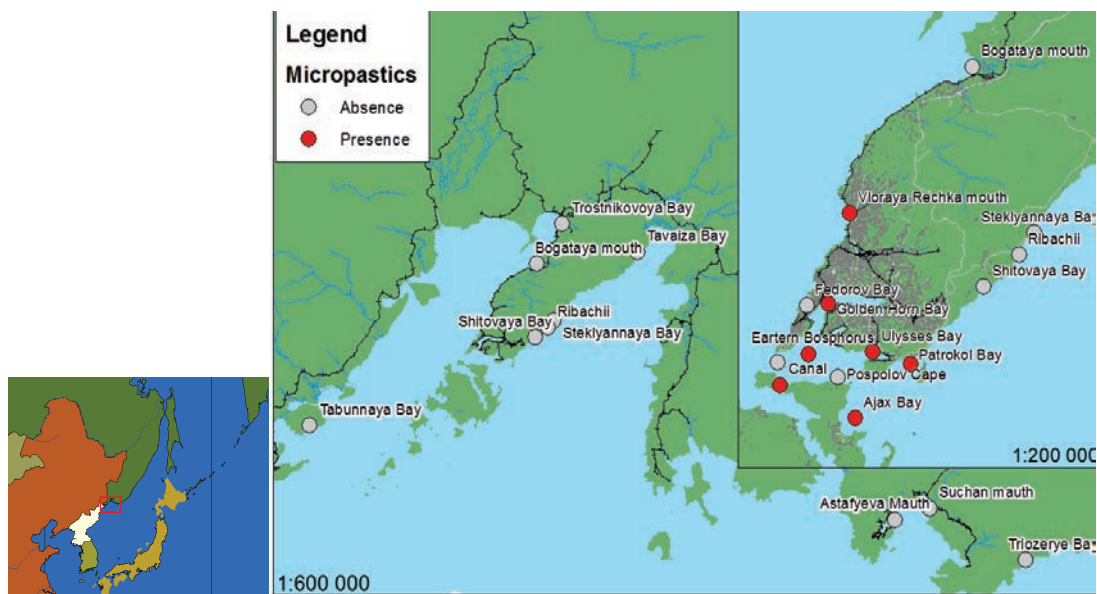


Fig. 5.4.1 Microplastic study program conducted in 2014 in the coastal area of Peter the Great Bay, Russia. Samples of surface water were collected at 16 sites, and microplastic particles were found in five of them.



Fig. 5.4.2 Sites where microplastics were analyzed in coastal surface water and intertidal sediments in 2015.

Rechka River, in the Russky Island Channel, at Ulysses Bay, Ajax Bay, and Patroclus (Patrokol) Bay (Fig. 5.4.1). In 2015, the study area was expanded. In addition to the Vladivostok area, the Hasansky region (Bezverkhovo village), Nakhodka (Astafieva Cape, Rizz Bay), and Partizansky region (Triozerie Bay) were also studied (Fig. 5.4.2). This time, in addition to surface water samples, intertidal sediments were collected at each site. Also, as of fall 2015, an additional sampling program was conducted in which 25 samples of intertidal sediments were collected.

Microplastic particles were detected at seven of the sites, located at Steklyannaya Bay, Rybachii settlement, Shitovaya Bay, Tokarevsky Cape, Fedorova Bay, Russky Island Channel, and Tabunnaya Bay. In 2016, a 2-year survey program in Peter the Great Gulf was initiated by the Pacific Geographical Institute of the Far Eastern Branch of the Russian Academy of Sciences (FEB RAS). Surface water at approximately 300 m from the coastline and intertidal sediments were collected at 12 sites along the shore from the Tumen River estuary to Cape Povorotny (southern tip of Nakhodka Bay near Kozmino), and also at two reference sites outside of the Bay (north-east of the sampling area, Fig. 5.4.3). The sampling sites were selected at urbanized and remote areas (including natural reserves), and some of the sites were located in close proximity to river estuaries. Several sites are used for summer recreational activities and for aquaculture and therefore, may experience different levels of contaminant inflow at different seasons. For example, the recreational sites at Slavyanka Bay and Lazurnaya appear to be more polluted in summer compared to autumn. Plastic contamination in the aquaculture areas were described earlier by Kozlovskii *et al.* (2017).

The combined results of this study show that the highest microplastic pollution levels (more than 20 particles per cubic meter of water) were registered in the southwestern part of the study area, near the Tumen River estuary (Fig. 5.4.3). Another hotspot of microplastic pollution was near Vladivostok city; however, the concentrations of microplastics were more than three times lower near Vladivostok compared to the Tumen River estuary. The most frequent particle types found were films, fragments, fibers and Styrofoam (EPS). The materials of the particles were identified using infrared spectroscopy and mass spectrometry, and the most frequent polymers were Polyethylene (PE), Polypropylene (PP),

and Polystyrene (PS). The types of polymers in fibers were identified only for a subset of the samples, and it was found that the most frequent polymers were nylon, polyester, PP, and PE.



Fig. 5.4.3 Approximate concentrations of microplastic particles in the coastal area of Northwest Pacific Russia (Peter the Great Bay).

Currently an additional data analysis to evaluate seasonal variations in microplastic pollution is being conducted. Also, analysis of microplastics in sediment samples is underway and will be finalized and reported in the near future.

References

- Blinovskaya, Y., Zakharenko, A., Golokhvast, K., Chernysh, O. and Zubtsova, I. 2020. Microplastic in the coastal sea waters of Russian Far East. IOP Conference Series: Earth and Environmental Science **459**(5): 052068. doi:10.1088/1755-1315/459/5/052068
- Kozlovskii, N.V., Hong, S.H., Song, Y.K. and Kachur, A.N. 2017. Distribution of beached marine litter and floating microplastics in the Minonosok Inlet of the Possiet Bay of the Peter the Great Gulf, pp. 235–239 *in*: Resources, Environment and Regional Sustainable Development in Northeast Asia, Proceedings of the III International Conference, October 10–14, 2016, Vladivostok, Russia.

6 United States of America

6.1 Temporal trends in polycyclic aromatic hydrocarbons (PAHs) in blue mussels (*Mytilus spp.*) from marine waters of Washington State

Jennifer Lanksbury¹, Alan Mearns², James West³ and Gina Ylitalo⁴

¹ Washington State Department of Fish and Wildlife, Olympia, Washington, USA

² Emergency Response Division, National Ocean Service, National Oceanic and Atmospheric Administration, Seattle, Washington, USA

³ Washington State Department of Fish and Wildlife, Olympia, Washington, USA

⁴ Northwest Fisheries Science Center, National Marine Fisheries Service, National Oceanic and Atmospheric Administration, Seattle, Washington, USA

Polycyclic aromatic hydrocarbons (PAHs) represent one of the most common and abundant groups of toxic chemicals in coastal marine waters of the United States. These chemicals are produced as a by-product of burning fossil fuels, wood combustion, natural forest and grass fires, volcanoes, and from petroleum products spilled or released into the environment (Baek *et al.*, 1991; Howsam and Jones, 1998). Although the toxicity and behavior of PAHs in the environment are wide ranging, once in aquatic systems many PAHs are highly toxic to fishes and invertebrates, and developing embryonic stages of fish are particularly susceptible to PAH exposure.

Although suspended for the past four years, the National Oceanic and Atmospheric Administration's (NOAA) Mussel Watch Program had been tracking PAHs and other chemicals in coastal marine bivalves (*i.e.*, mussels and oysters) to observe spatial and temporal trends in chemical pollutants on a national scale. Sampling in Washington State consisted of biannual collections of wild mussels from over 20 locations in inland marine waters (Puget Sound) and Pacific coastal shorelines or estuaries (Kimbrough *et al.*, 2008). Sampling locations were meant to represent "ambient" or average conditions at a state-wide scale. As such, no sampling sites were located adjacent to known major point sources.

PAH levels in mussels have remained constant and relatively high in Puget Sound compared to the Pacific Ocean coast (Fig. 6.1.1), from the inception of Mussel Watch through its most recent sampling in 2012 (Mearns, 2002; Melwani *et al.*, 2013).

Overall, Puget Sound mussels exhibited a roughly 5-fold higher sum of PAH concentration than coastal mussels (2,300 vs 430 ng/g dry weight, dw; Fig. 6.1.1), which reflects the greater urbanization of the Puget Sound Basin (greater source of PAHs) (NOAA's National Status and Trends: Mussel Watch Program, unpublished data; Washington Department of Fish and Wildlife, unpublished data).

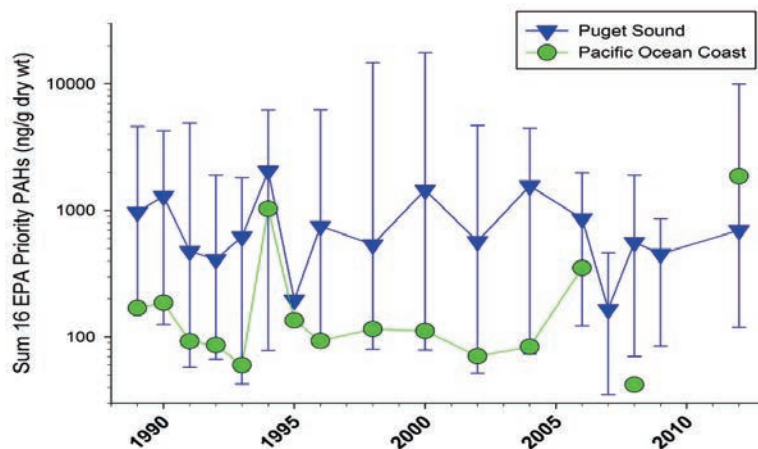


Fig. 6.1.1 Total mean PAH concentrations (ng/g dry weight, dw) in blue mussels from inland marine waters (Puget Sound; 17 sites, triangles) and Pacific coastal shorelines (three sites, circles). Data are the sum of the U.S. Environmental Protection Agency priority chemicals, in ng/g wet weight, for composites of soft tissue from whole mussels. Puget Sound sites also include 95% confidence intervals.

Data from the NOAA's Mussel Watch Program have been used by the Washington Department of Fish and Wildlife (http://wdfw.wa.gov/conservation/research/projects/marine_toxics/index.html) and the Puget Sound Ecosystem Monitoring Program (<https://sites.google.com/a/psemp.org/psemp/home>) to provide information and recommendations to policy makers for protecting, conserving, and restoring Puget Sound's natural resources (Puget Sound Action Team, 2007). Recently, data from the Mussel Watch Program were used to compare background levels of PAHs in Puget Sound mussels to those recorded in mussels exposed to diesel fuel from a sunken fishing vessel in Penn Cove, Washington (Mearns *et al.*, 2014). Though the NOAA Mussel Watch Program is currently suspended, recent mussel monitoring efforts in Puget Sound, under the new Regional Stormwater Monitoring Program (RSMP) (<http://www.ecy.wa.gov/programs/wq/stormwater/municipal/rsmp/status.html>), will be used to inform and support pollution control and management in the Sound's urban shoreline areas (Lanksbury and Lubliner, 2015).

References

- Baek, S.O., Field, R.A., Goldstone, M.E., Kirk, P.W., Lester, J.N. and Perry, R. 1991. A review of atmospheric polycyclic aromatic hydrocarbons: Sources, fate and behavior. *Water, Air, and Soil Pollution* **60**: 279–300.
- Howsam, M. and Jones, K.C. 1998. Sources of PAHs in the environment, pp. 137–174 *in*: PAHs and Related Compounds. The Handbook of Environmental Chemistry, Vol. 3 *edited by* A.H. Neilson. Springer, Berlin.
- Kimbrough, K.L., Lauenstein, G.G., Christensen, J.D. and Apeti, D.A. 2008. An assessment of two decades of contaminant monitoring in the Nation's Coastal Zone. NOAA Technical Memorandum NOS NCCOS, 74, NOAA/National Centers for Coastal Ocean Science, Silver Spring, MD.
- Lanksbury, J.A. and Lubliner, B. 2015. Quality Assurance Project Plan for Status and Trends Monitoring of Marine Nearshore Mussels for the Regional Stormwater Monitoring Program and Pierce County. Washington Department of Fish and Wildlife Publication #FPT 15-04, 76 pp. <http://wdfw.wa.gov/publications/01760/>.

- Lanksbury, J.A., Niewolny, L.A., Carey, A.J. and West, J.E. 2014. Toxic contaminants in Puget Sound's nearshore biota: a large-scale synoptic survey using transplanted mussels (*Mytilus trossulus*). Washington Department of Fish and Wildlife Publication #FPT 14-08, 180 pp. <http://wdfw.wa.gov/publications/01643/>.
- Mearns, A.J. 2002. Long-term Contaminant Trends and Patterns in Puget Sound, the Straits of Juan de Fuca, and the Pacific Coast. Paper 5a, Proceedings of Puget Sound Research 2001: The Fifth Puget Sound Conference. Puget Sound Water Quality Action Team, Olympia, WA, USA.
http://archives.eopugetsound.org/conf/2001PS_ResearchConference/sessions/oral/5a_mearn.pdf.
- Mearns, A.J., Shigenaka, G., Meyer, B., and Drury, LTJG A. 2014. Contamination and recovery of commercially-reared mussels exposed to diesel fuel from a sunken fishing vessel. International Oil Spill Conference Proceedings, May 2014, Vol. 2014, No. 1, pp. 1686–1705.
- Melwani, A.R., Gregorio, D., Jin, Y., Stephenson, M., Ichikawa, G., Siegel, E., Crane, D., Lauenstein, G. and Davis, J. 2013. Mussel Watch update: Long-term Trends in Selected Contaminants from Coastal California, 1977 – 2010. *Marine Pollution Bulletin* **81**: 291–302.
- NOAA's National Status and Trends: Mussel Watch Program. <https://data.noaa.gov/dataset/national-status-and-trends-mussel-watch-program>.
- Puget Sound Action Team. 2007. 2007 Puget Sound Update: Ninth Report of the Puget Sound Ambient Monitoring Program. Puget Sound Action Team. Olympia, WA, 260 pp.
- Washington Department of Fish and Wildlife.
http://wdfw.wa.gov/conservation/research/projects/marine_toxics/index.html.

6.2 NOAA Marine Debris Monitoring and Assessment Project: Four years of effort in the U.S. Pacific States

Sherry Lippiatt^{1,2} and Carlie Herring^{1,3}

¹I.M. Systems Group, Rockville, Maryland, USA

²U.S. National Oceanic and Atmospheric Administration, Marine Debris Program, Oakland, California, USA

³U.S. National Oceanic and Atmospheric Administration, Marine Debris Program, Silver Spring, Maryland, USA

Background and history of marine debris monitoring in the United States

The National Oceanic and Atmospheric Administration Marine Debris Program (NOAA MDP) is legislatively mandated to identify, determine sources of, assess, prevent, reduce, and remove marine debris (33 U.S.C. 1951 *et seq.*, as amended by Title VI of Public Law 112-213). Prior to the 2006 creation of the NOAA MDP, the National Marine Debris Monitoring Program (NMDMP) operated in the U.S. from 1996 to 2006. The NMDMP was developed by an interagency working group consisting of the U.S. Environmental Protection Agency, NOAA, National Park Service, and United States Coast Guard. The NMDMP was designed to assess the magnitude of the marine debris problem in the U.S. and evaluate any regional or temporal trends in source categories according to a statistically valid design and sampling plan (Escardó-Boomsa *et al.*, 1995). The NMDMP monitoring protocols and results of this national monitoring effort can be found in Sheavly (2007) and Ribic *et al.* (2012).

The NOAA MDP initiated the development of the Marine Debris Monitoring and Assessment Project (MDMAP) in 2009, to provide widely applicable and inexpensive monitoring protocols to partner organizations, with the intention of promoting standardization of shoreline marine debris monitoring efforts. Lessons learned from NMDMP were considered in the development of the protocol and program, in addition to the monitoring guidelines published by the United Nations Environment Programme in 2009 (Cheshire *et al.*, 2009). The NOAA MDMAP standing stock shoreline survey technique was designed as a rapid, quantitative beach assessment of the distribution, types, and abundance of debris in the marine environment. More specifically, these surveys were designed to measure the total load of marine debris (larger than 2.5 cm in the longest dimension) at a 100 m length shoreline site on a monthly basis, and to assess whether the amount and composition of that debris was changing over time (Lippiatt *et al.*, 2013).

Marine debris monitoring, like other monitoring programs, requires coordinated effort and commitment by the partners involved, and can reap great benefits. Long-term marine debris monitoring studies provide important information on the types and sources of debris, and results can be applied to policy development, education and outreach, and research initiatives at various spatial scales.

MDMAP as a citizen science program

The tragic 2011 Great East Japan Earthquake and tsunami generated intense public interest and concern about the significant amount of marine debris generated and the potential for tsunami debris to find its way to U.S. shorelines. In response, the NOAA MDP shifted the original intent of MDMAP to accommodate data collection by beach cleanup groups. The standing stock protocol described in

Lippiatt *et al.* (2013) was adapted to incorporate debris removal, and a new accumulation survey technique was published (Opfer *et al.*, 2012). Both protocols require a 100 m length shoreline site, are limited to debris larger than 2.5 cm, and call for monthly (every 28 ± 3 days) surveys. New partnerships were initiated in late 2011 and early 2012, as MDMAP morphed from a program originally targeting the research community to a broader citizen science initiative. The MDMAP currently includes both accumulation and standing stock survey sites (Table 6.2.1, Fig. 6.2.1), depending on the interests, goals, and physical constraints of the monitoring team.

Table 6.2.1 Number of monitoring sites and surveys completed in U.S. Pacific States between March 1, 2012 and February 29, 2016. Verified data only*.

State	Accumulation sites/Surveys	Standing stock sites/Surveys
Alaska	11/30	9/45
Washington	26/575	N/A
Oregon	7/115	N/A
California	19/360	6/210
Hawai'i	9/122	4/19
Total	72/1202	19/274

*Verified site and survey data has been reviewed by NOAA MDP staff and affiliates. There may be a delay between survey submission to the MDMAP database and verification.

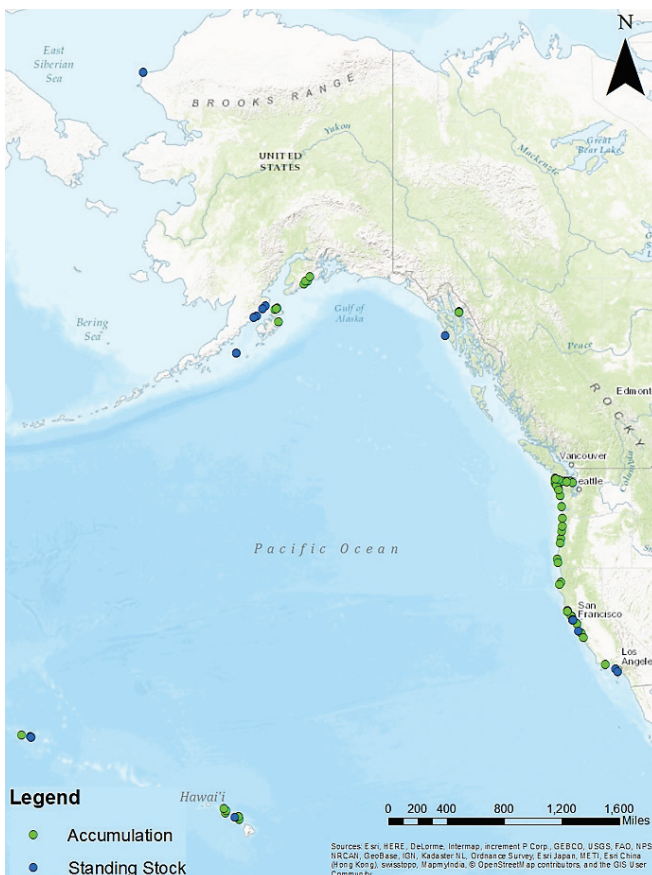


Fig. 6.2.1 Map of MDMAP accumulation and standing stock sites in U.S. Pacific States as of February 29, 2016. A more detailed map is available at <https://mdmap.orr.noaa.gov>.

Since its launch in 2012, the MDMAP has grown considerably in the number of partner organizations and shoreline sites being monitored. A total of 91 sites in U.S. Pacific States were surveyed at least once between March 2012 and February 2016. Monitoring teams come from government agencies, academia, non-profit organizations, and individual volunteers. Survey data and photos are submitted to NOAA MDP and compiled in a publicly available online database (mdmap.orr.noaa.gov). Nationally, shoreline monitoring efforts are heavily focused on the Pacific region, an area impacted by Japan tsunami marine debris.

Results to date

Across all MDMAP surveys in U.S. Pacific States, a total of 211,709 debris items were recorded between March 2012 and February 2016. Plastic debris items comprised 78% of the debris recorded (Fig. 6.2.2). Of the plastic items recorded, hard, foamed, and filmed plastic fragments dominated, accounting for 54% of the total, or 89,424 items. The remaining plastic items were identifiable items, for example, plastic bottles, bottle caps, or containers. Given that the number of sites and surveys varies across states, the relative abundance of identifiable items recorded in each state is detailed below (Fig. 6.2.3).

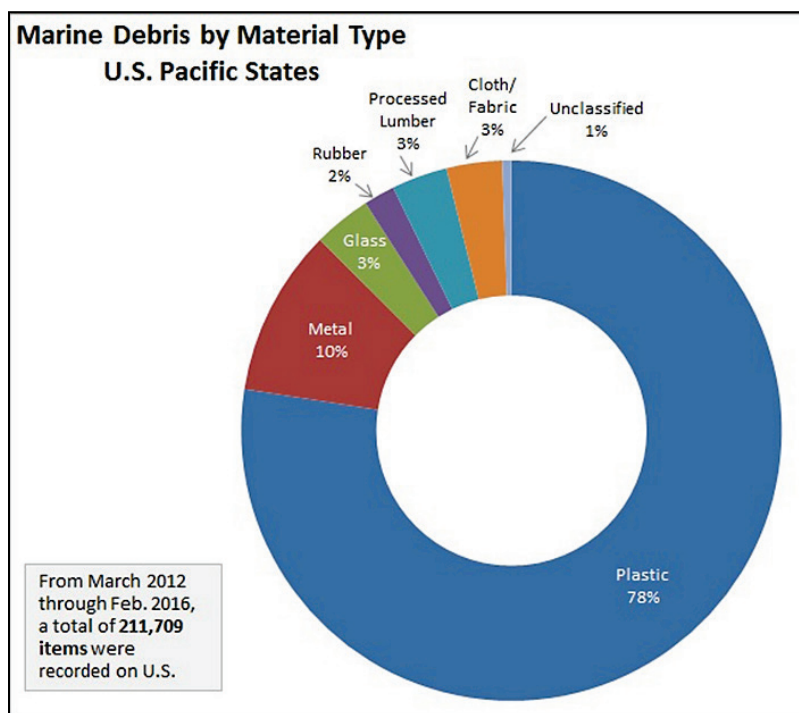


Fig. 6.2.2 Percent of marine debris items by material type recorded in the U.S. Pacific States between March 1, 2012 and February 29, 2016. Includes data from both accumulation and standing stock surveys.

For all U.S. Pacific States' data combined, the top ten identifiable items account for 29% of the total debris recorded in both accumulation and standing stock surveys. Items that appear in the top ten list for both accumulation and standing stock surveys across all states include plastic rope/small net pieces, bottle/container caps, food wrappers, and plastic beverage bottles. Notably, bottle/container caps are in

the top four most common items in all states. Oregon and California have a more varied assemblage of debris types than other states, with the most common item only accounting for 5% or less of all identifiable debris recorded in accumulation surveys (Fig. 6.2.3). Lumber/building material is the only non-plastic item to appear in the top ten list from all states except Hawai'i. Cigarettes are most common in California and Hawai'i on a per-debris item basis (Fig. 6.2.3), comprising 5% and 8% of identifiable debris items, respectively (note that only cigarette butts longer than 2.5 cm are reported, so this is likely an underestimate of the total number encountered).

Plastic debris items on the MDMAP datacard can be assigned to user group categories (Table 6.2.2). In Alaska, California, and Oregon, plastic consumer items are at least twice as common as plastic fishing-related items. In Hawai'i plastic fishing-related products are slightly more common than plastic consumer items, whereas in Washington there is about an equal split between the two user categories.

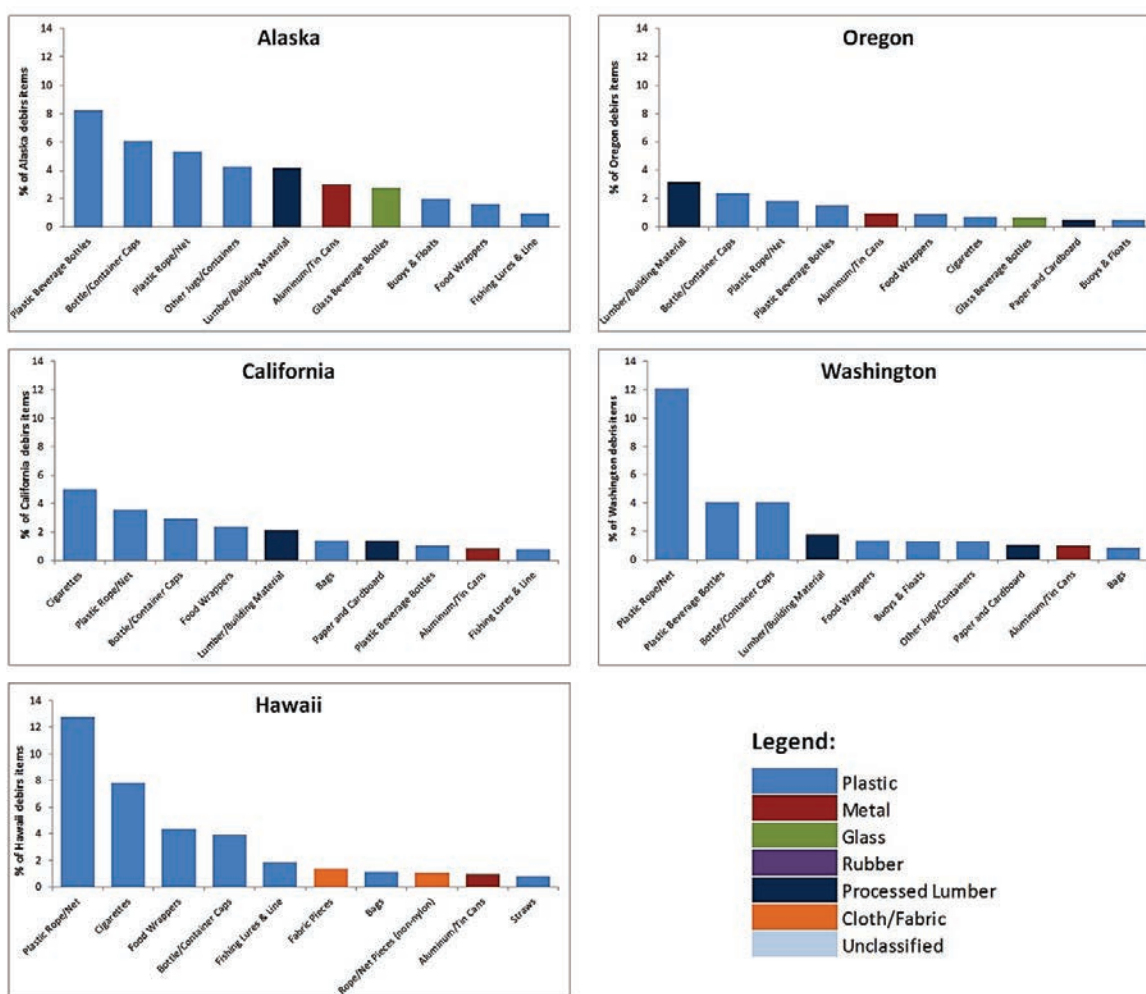


Fig. 6.2.3 The top ten identifiable debris items as a percent of total number of debris items recorded in accumulation surveys for each U.S. Pacific State between March 2012 and February 2016. Results are based on the following number of surveys for each state: Alaska (30), California (360), Hawai'i (122), Oregon (115), and Washington (575). Note that fragments of all material types are excluded from this analysis.

Table 6.2.2 Plastic debris (total number of items) by user group category for accumulation surveys for U.S. Pacific States, normalized by the number of surveys in each state.

	AK	AK per survey	CA	CA per survey	HI	HI per survey	OR	OR per survey	WA	WA per survey
Plastic consumer products	1182	39.4	4885	13.6	13575	111.3	601	5.2	4676	8.1
Plastic smoking-related products	71	2.4	2553	7.1	8423	69.0	104	0.9	223	0.4
Plastic fishing-related products	437	14.6	2065	5.7	15555	127.5	228	2.0	4909	8.5
Total	1690	56.3	9503	26.4	37553	307.8	933	8.1	9808	17.1
Number of surveys	30		360		122		115		575	

User group categories include plastic consumer products, plastic smoking products >2.5cm, and plastic fishing-related products.

Plastic consumer products include: food wrappers, plastic beverage bottles, other jugs/containers, bottle/container caps, 6-pack rings, bags, cups, plastic utensils, straws, balloons, and personal care products.

Plastic smoking products >2.5 cm include: cigar tips, cigarettes, and disposable cigarette lighters.

Plastic fishing-related products include: plastic rope/net, buoys and floats, and fishing lures and line.

Plastic rope and net alone account for about 12–13% of total identifiable debris items in both Washington and Hawai'i (Fig. 6.2.3). In California, four of the top ten most common identifiable items are plastic consumer products (Fig. 6.2.3), and plastic consumer products are twice as common as plastic smoking-related debris and 2.4 times as common as plastic fishing-related debris (Table 6.2.2). Smoking-related debris was rare in both Oregon and Washington, averaging less than one item per survey (Table 6.2.2).

On a per-survey basis, more items were recorded on shorelines in Hawai'i and Alaska compared to the other states. At accumulation survey sites, on average, 862 items were recorded per survey in Hawai'i, which is 14 times more debris than sites in Washington, 11 times more debris than sites in Oregon, seven times more debris than sites in California, and five times more debris than sites in Alaska (data not shown).

Discussion

Marine debris is generated by humans on land and at sea, and can persist in the environment and travel long distances to remote shorelines (Browne *et al.*, 2011; Bergmann *et al.*, 2017; Lavers and Bond, 2017). The preliminary MDMAP data presented here, namely the variety in debris assemblages and presumed sources of debris items found on coastlines of the U.S. Pacific States, demonstrates the long-lived and mobile nature of marine debris. For example, much of the consumer-related debris encountered at sites in Alaska was likely generated elsewhere and spent a significant amount of time at sea before being deposited onshore. Differences in debris loads and the relative abundances of debris from different user group categories occur across geographies given the various drivers of debris accumulation (Barnes *et al.*, 2009), and can provide information on debris sources and potentially effective prevention approaches.

Given the tendency of plastic and other material types to break apart when exposed to the elements, the high prevalence of fragments of larger debris items in MDMAP surveys is expected and aligns with shoreline surveys from other locales (Hardesty *et al.*, 2017; Lavers and Bond, 2017). Fragment data are not directly applicable to identifying targets for prevention, but are important in considering the relative abundance of material types and the lifecycle and degradation of debris items. Identifiable debris items are easier to link to user groups and source categories. For example, plastic bottle caps were prevalent in surveys in all U.S. Pacific States. Bottle caps are typically made of polypropylene, sourced from a single-use, disposable item, and are relatively small but recognizable on sandy beaches. Further, bottle caps were the fourth most common item found globally during the International Coastal Cleanup in 2015 (<https://oceanconservancy.org/wp-content/uploads/2017/04/2015-Ocean-Conservancy-ICC-Report.pdf>).

Processed lumber was the only non-plastic item to make the top ten list in most states. Increased abundance of processed lumber is presumed to be associated with debris generated by the 2011 Tohoku earthquake and tsunami (Clarke Murray *et al.*, 2018). As debris generated by the tsunami continues to wash ashore on U.S. shorelines in decreased amounts, it will be interesting to look for changes in the relative abundance of processed lumber compared to other debris types.

Shoreline sites in Alaska and Hawai'i had significantly greater numbers of debris items recorded per accumulation survey compared to the other states. These remote locations are notorious for high debris loads based on geography and the current-driven deposition of debris that is not locally sourced. Further, some remote shoreline sites in Alaska and Hawai'i (including the Northwestern Hawaiian Islands) are only accessed and monitored on a seasonal or annual basis, which may result in higher debris loads at each survey compared to sites with monthly survey and cleanup. Lastly, it is important to consider that across all states, without a stratified sampling strategy, the MDMAP has an inherent but undefined site-selection bias that impacts comparisons across states.

The MDMAP has garnered significant interest and participation from academia, partner organizations, and the general public. As with other environmental monitoring networks, marine debris shoreline monitoring efforts are an effective way to engage volunteers in a rewarding citizen science experience while providing for the collection of valuable scientific data (Conrad and Hilchey, 2011; Bergmann *et al.*, 2017; van der Velde *et al.*, 2017). The MDMAP network and the results of locally based shoreline monitoring studies provide tools and opportunities for outreach and raising awareness about marine debris issues, sharing the message that every individual can become a part of the solution. For example, school groups can employ the program as an experiential education and data analysis project, and simultaneously make connections between waste generated in their everyday lives and the debris that ends up in the environment. The commitment, dedication, and energy of the MDMAP partners and volunteers cannot be overlooked, and is essential to the continued success of the program. Further, the development of a public online database for accessing MDMAP data has facilitated an increasing number of login requests from students, researchers, and individuals with a general interest in the data. As of May 2016, 170 individuals have created accounts on the MDMAP database (<https://mdmap.orr.noaa.gov>).

To date, MDMAP participants have selected their own shoreline monitoring sites according to a set of criteria (Lippiatt *et al.*, 2013). Thus, site selection is usually based on proximity to partner organizations and personal interest. This makes it easier to market the program to place-based monitoring groups, but without a rigorous sample design, MDMAP is limited in data analysis that can extend beyond the individual site level. In 2015, NOAA launched a partnership with the Ocean Conservancy to develop a

model-based sampling strategy based on existing MDMAP data. The final report is available on the NOAA MDP website (<https://marinedebris.noaa.gov/reports/analysis-marine-debris-us>).

Working with many unfunded, volunteer-based organizations has allowed the MDMAP to engage a larger audience than it otherwise would have with limited resources. However, it is challenging to manage varying levels of commitment, responsiveness, and training from participating organizations. Maintaining regular communication and contact with dozens of different organizations is challenging to manage with a small staff, but essential to keeping participants engaged. Further, dissemination of research findings and results of monitoring studies is necessary for a meaningful citizen science experience and to maintain support for the program's existence. In order to assist with volunteer training, access to resources, and results sharing, the MDP recently launched a "Get Started Toolbox" for the MDMAP at <https://marinedebris.noaa.gov/research/monitoring-toolbox>.

Given increasing interest in addressing marine debris from local municipalities to the international stage, programs such as MDMAP are essential for scientifically documenting the issue, monitoring change, and tracking progress. There are many challenges inherent in standardizing monitoring techniques across the board, including different questions of interest, geographical differences, shoreline accessibility, access to technology, and target audience (Ryan *et al.*, 2009). Forward momentum toward globally harmonized marine debris monitoring practices is needed in order to make broad assessments and add value to individual efforts.

References

- Barnes, D.K.A., Galgani, F., Thompson, R.C. and Barlaz, M. 2009. Accumulation and fragmentation of plastic debris in global environments. *Philosophical Transactions of the Royal Society B* **364**: 1985–1998.
- Bergmann, M., Lutz, B., Tekman M.B. and Gutow, L. 2017. Citizen scientists reveal: Marine litter pollutes Arctic beaches and affects wild life. *Marine Pollution Bulletin* **125**: 535–540, <https://doi.org/10.1016/j.marpolbul.2017.09.055>.
- Browne, M.A., Crump, P., Niven, S.J., Teuten, E., Tonkin, A., Galloway, T. and Thompson, R. 2011. Accumulation of microplastic on shorelines worldwide: Sources and sinks. *Environmental Science and Technology* **45**: 9175–9179, doi: 10.1021/es201811s.
- Cheshire, A.C., Adler, E., Barbiere, J., Cohen, Y., Evans, S., Jarayabhand, S. and Westphalen, G. 2009. UNEP/IOC Guidelines on Survey and Monitoring of Marine Litter. UNEP Regional Seas Intergovernmental Oceanographic Commission, 120 pp.
- Clarke Murray, C., Maximenko, N. and Lippiatt, S. 2018. The influx of marine debris from the Great Japan Tsunami of 2011 to North American shorelines. *Marine Pollution Bulletin* **132**: 26–32, <https://doi.org/10.1016/j.marpolbul.2018.01.004>.
- Conrad, C.C. and Hilchey, K.G. 2011. A review of citizen science and community-based environmental monitoring: issues and opportunities. *Environmental Monitoring and Assessment* **176**: 273–291, doi: 10.1007/s10661-010-1582-5.
- Escardo-Boomsma, J., O'Hara, K. and Ribic, C.A. 1995. National Marine Debris Monitoring Program: Volume I. U.S. Environmental Protection Agency.
- Hardesty, B.D., Lawson, T., van der Velde, T., Lansdell, M. and Wilcox, C. 2017. Estimating quantities and sources of marine debris at a continental scale. *Frontiers in Ecology and the Environment* **15**: 18–25, doi:10.1002/fee.1447.

- Lavers, J.L. and Bond, A.L. 2017. Exceptional and rapid accumulation of anthropogenic debris on one of the world's most remote and pristine islands. *Proceedings of the National Academy of Sciences of the United States of America* **114**: 6052–6055, doi: 10.1073/pnas.1619818114.
- Lippiatt, S., Opfer, S. and Arthur, C. 2013. Marine Debris Monitoring and Assessment. NOAA Technical Memorandum NOS-OR&R-46.
- Opfer, S., Arthur, C. and Lippiatt, S. 2012. NOAA Marine Debris Shoreline Survey Field Guide. Marine Debris Program, NOAA, U.S. Department of Commerce.
- Ribic, C.A., Sheavly, S.B., Rugg, D.J. and Erdmann, E.S. 2012. Trends in marine debris along the U.S. Pacific Coast and Hawai'i 1998–2007. *Marine Pollution Bulletin* **64**: 994–1004. doi: 10.1016/j.marpolbul.2012.02.008
- Ryan, P.G., Moore, C.J., Van Franeker, J.A. and Moloney, C.L. 2009. Monitoring the abundance of plastic debris in the marine environment. *Philosophical Transactions of the Royal Society B* **364**: 1999–2012.
- Sheavly, S.B. 2007. National Marine Debris Monitoring Program: Final Program Report, Data Analysis and Summary *edited by* Ocean Conservancy, pp. 76. Submitted to U.S. Environmental Protection Agency.
- van der Velde, T., Milton, D.A., Lawson, T., Wilcox, C., Lansdell, M., Davis, G., Perkins, G. and Hardesty, B.D. 2017. Comparison of marine debris data collected by researchers and citizen scientist: Is citizen science data worth the effort? *Biological Conservation* **208**: 127–138, <https://doi.org/10.1016/j.biocon.2016.05.025>.

Summary and Recommendations

Pollution can adversely affect the health and abundance of marine biota, especially in densely populated coastal areas of PICES member countries. The downstream socio-economic and public health consequences can be significant, with numerous examples of consumption advisories, commercial fishery closures, commercial trade interdictions and diminished aboriginal access to food resources around the North Pacific Ocean. The protection of ecosystem health and services requires an ability to detect emerging pollutant issues before serious adverse impacts arise. Regulations, policies and other management actions resulting from marine pollution research in the past have led to dramatic declines in environmental concentrations of a number of harmful pollutants, subsequently improving the health of marine biota in several countries from the PICES community, as described in this report.

In this context, the Working Group on *Emerging Topics in Marine Pollution* recommends PICES:

- To continue the concerted and long-term characterization of emerging pollutants or pollution priorities in the North Pacific Ocean to determine the dominant anthropogenic pressures in coastal marine ecosystems and how they are changing and affecting societies depending upon marine resources;
- To keep track and ensure the compilation of pollutant data and monitoring programs conducted by its six member countries;
- To continue documenting the impacts of emerging pollutants in the North Pacific Ocean, especially in coastal environments, and in a changing ocean environment in the face of climate change forcing, including increasing sea surface temperature, sea bottom temperature, deoxygenation/hypoxia, ocean acidification, changes in primary production and sea level rise, affecting nearshore and coastal ecosystems and their interactions with offshore and terrestrial systems;
- To engage and enhance collaboration with other expert groups of PICES to document the importance of marine pollution relative to multiple anthropogenic stressors and associated interactions and cumulative effects;
- To develop approaches to pollutant indicators that account for climate variability and change to understand the consequences of projected coastal ecosystem changes as well as taking into account the predictability and uncertainty of forecasted changes by characterizing changing pollution risks as climate changes;
- To prioritize pollutant sources of contaminants of emerging concern (*e.g.*, emerging POPs, microplastics, pharmaceuticals and personal care products) and types in support of source control, regulations and best management practices.
- To enact and enforce regulations that prevent chemicals from being produced, sold and/or used commercially unless these substances are tested for and found not to exhibit bioaccumulation, toxicity and/or other harmful effects in the marine environment.

Appendix 1

WG 31 Terms of Reference

WG 31 term: 2014–2017

Parent Committee: MEQ

1. Document and profile emerging marine pollution issues in the North Pacific Ocean within the PICES community by:
 - Convening Topic Sessions and workshops on new and emerging pollutants and pollution issues;
 - Coordinating a series of special issues in international peer-reviewed journals based on Topic Sessions.
2. Compile data on pollution indicators describing spatial and temporal status, trends and impacts in the North Pacific Ocean in support of a contribution to the next edition of the PICES North Pacific Ecosystem Status Report.
3. Strengthen partnerships to deliver Topic Sessions/workshops and to publish special issues with:
 - Other PICES expert groups, especially those identified in the FUTURE Science Plan;
 - Other international organizations, including the International Council for the Exploration of the Sea (ICES), the Joint Group of Experts on the Scientific Aspects of Marine Environmental Protection (GESAMP), and the North West Pacific Action Plan (NOWPAP).
4. Contribute to FUTURE by publishing a final report summarizing results of Expert Group deliberations.

Appendix 2

WG 31 Membership

Canada

John Edward Elliott
Science and Technology Branch
Environment Canada
Pacific Wildlife Research Centre
5421 Robertson Rd.
Delta, BC, V4K 3N2
Canada
E-mail: john.elliott@ec.gc.ca

Peter S. Ross (WG 31 Co-Chair)
Ocean Pollution Research Program
Coastal Ocean Research Institute, Ocean Wise
Conservation Association/Vancouver Aquarium
P.O. Box 3232
Vancouver, BC, V6B 3X8
Canada
E-mail: peter.ross@ocean.org

Japan

Hideaki Maki
National Institute for Environmental Studies
16-2 Onogawa
Tsukuba, Ibaraki 305-8506
Japan
E-mail: hidemaki@nies.go.jp

Hideshige Takada
Laboratory of Organic Geochemistry
Tokyo University of Agriculture and Technology
3-5-8 Saiwai-cho
Fuchu, Tokyo 183-8509
Japan
E-mail: shige@cc.tuat.ac.jp

Kazuhiko Mochida
National Research Institute of Fisheries and
Environment of Inland Sea, FRA
2-17-5 Maruishi
Hatsukaichi, Hiroshima 739-0452
Japan
E-mail: kmochida@fra.affrc.go.jp

People's Republic of China

Zhengguo Cui (2017)
 Yellow Sea Fisheries Research Institute, CAFS
 106 Nanjing Rd., Shinan District
 Qingdao, Shandong 266071
 People's Republic of China
 E-mail: cuizg@ysfri.ac.cn

Haiyan Lv (2017)
 Chinese Academy of Fishery Sciences
 150 South Yongding Rd.
 Qingtacun, Fengtai District
 Beijing 100141
 People's Republic of China
 E-mail: lvhy@cafs.ac.cn

Guangshui Na
 National Marine Environmental Monitoring
 Center, SOA
 42 Linghe St., Shahekou District
 Dalian, Liaoning 116023
 People's Republic of China
 E-mail: gsna2010@163.com

Qun Wang (2017)
 Chinese Academy of Fishery Sciences
 150 South Yongding Rd.
 Qingtacun, Fengtai District
 Beijing 100141
 People's Republic of China
 E-mail: wangq@cafs.ac.cn

Zijun Xu
 North China Sea Environmental Monitoring
 Center
 North China Sea Branch of SOA
 22 Fushun Rd.
 Qingdao, Shandong 266000
 People's Republic of China
 E-mail: zjxu77@gmail.com

Republic of Korea

Dong-Woon Hwang
 Marine Environment Research Division
 National Institute of Fisheries Science, MOF
 216 Haean-ro, Gijang-eup, Gijang-gun
 Busan 46083
 Republic of Korea
 E-mail: dwhwang@korea.kr

Hyo-Bang Moon
 Department of Environmental Marine Sciences
 Hanyang University
 55 Hanyangdaehak-ro, Sangnok-gu
 Ansan, Kyeonggi-do 426-791
 Republic of Korea
 E-mail: hbmoon@hanyang.ac.kr

Wonjoon Shim (WG 31 Co-Chair)
 Oil and POPs Research Group
 Korea Institute of Ocean Science and Technology
 41 Jangmok-1-gil, Jangmok-myeon
 Geoje 656-834
 Republic of Korea
 E-mail: wjshim@kiost.ac.kr

Un Hyuk Yim
 Oil and POPs Research Group
 Korea Institute of Ocean Science and Technology
 41 Jangmok-1-gil, Jangmok-myeon
 Geoje 656-834
 Republic of Korea
 E-mail: uhyim@kiost.ac.kr

Russian Federation

Olga N. Lukyanova (WG 31 Co-Chair)
Laboratory of Fisheries Oceanography
Pacific Scientific Research Fisheries Center
(TINRO-Center)
4 Shevchenko Alley
Vladivostok, Primorsky Kray 690950
Russia
E-mail: olga.lukyanova@tinro-center.ru

Mikhail Simokon
Pacific Scientific Research Fisheries Center
(TINRO-Center)
4 Shevchenko Alley
Vladivostok, Primorsky Kray 690091
Russia
E-mail: simokon@tinro.ru

Vladimir M. Shulkin
Ex officio member, representing NOWPAP
Pacific Geographical Institute, FEB RAS
7 Radio St.
Vladivostok, Primorsky Kray 690041
Russia
E-mail: shulkin@tig.dvo.ru

United States of America

Staci Simonich
Department of Environmental and Molecular
Toxicology
Oregon State University
1007 Agriculture and Life Science Bldg.
Corvallis, OR 97331-7301
USA
E-mail: staci.simonich@oregonstate.edu

Gina M. Ylitalo
Environmental Conservation Division
Northwest Fisheries Science Center, NMFS, NOAA
2725 Montlake Blvd E
Seattle, WA 98112-2097
USA
E-mail: Gina.Ylitalo@noaa.gov

Nancy Wallace
Marine Debris Division
NOAA Office of Response and Restoration
1305 East-West Hwy
Silver Spring, MD 20910
USA
E-mail: nancy.wallace@noaa.gov

Appendix 3

WG 31's Contribution to the North Pacific Ecosystem Status Report

WG 31 fulfilled TOR2 by making a notable contribution to the next PICES North Pacific Ecosystem Status Report (NPESR). A third in a series, NPESR3 is the first to discuss pollutants. Accordingly, WG 31 members met at PICES-2015 in Qingdao, China, to develop input for the writing group tasked with compiling the major NPESR compendium. WG 31 members from every PICES member country were asked to volunteer to compile data and write reports reflecting their expertise, dataset availability and/or topic of national interest. While ideas and data varied by member country, the ultimate hope was that some national datasets/reports might be integrated into a North Pacific report. WG 31 identified two primary matrices with good time series and commonalities across the six member countries: sediments and bivalves. Although it was not necessary to restrict all reports to these matrices, they showed promise in terms of compiling international data from different national reports. In this context, it was recognized that additional biological matrices, such as marine mammals or/and seabird eggs, offer valuable opportunities to provide an overview of trends and potential impacts on marine life.

On June 26–30, 2016, the Writing Group for the North Pacific Ecosystem Status Report, Third Edition (NPESR3) effort met at an Invitational Workshop on “Evaluation and Synthesis North Pacific Time Series Observations” in Sidney (BC, Canada) to tabulate current draft reports and move towards combining national and international chapters. During WG 31's meeting at the PICES-2016 in San Diego (CA, USA), there was consensus among the participants to draft national pollution indicator reports by contributing PICES member countries, instead of reporting contaminants categories due to the different matrices and pollutants reported. A template describing the Terms of References for participating PICES members was provided with the aim of following a common format to report the description ecosystem time series observations (ETSOs; Annex). The success of the WG 31 contribution to NPESR depended upon the input of members from all PICES member countries. We received ‘pollution indicator’ reports from all six countries. This report provides an update of all contributions submitted over the period 2018 to 2019 for a total of 34 reports (Fig. 1).

In doing so, this report provides a fairly comprehensive assessment of most pollution issues that are relevant to the PICES community by contributing new data and information on the following research fronts:

- Priority pollutants in the North Pacific Ocean as identified by scientists from PICES' six member countries;
- Promising indicator approaches, including abiotic compartments (*i.e.*, sediments, seawater) and/or ‘sentinel species’ (*e.g.*, mussels, seabirds, marine mammals) that are being used within the PICES community, or elsewhere, and could be used more widely across PICES member countries;

- Opportunities to share technical information on logistical, analytical, and/or field study methods in support of improved and/or standardized methodologies that could be used by all PICES member countries.

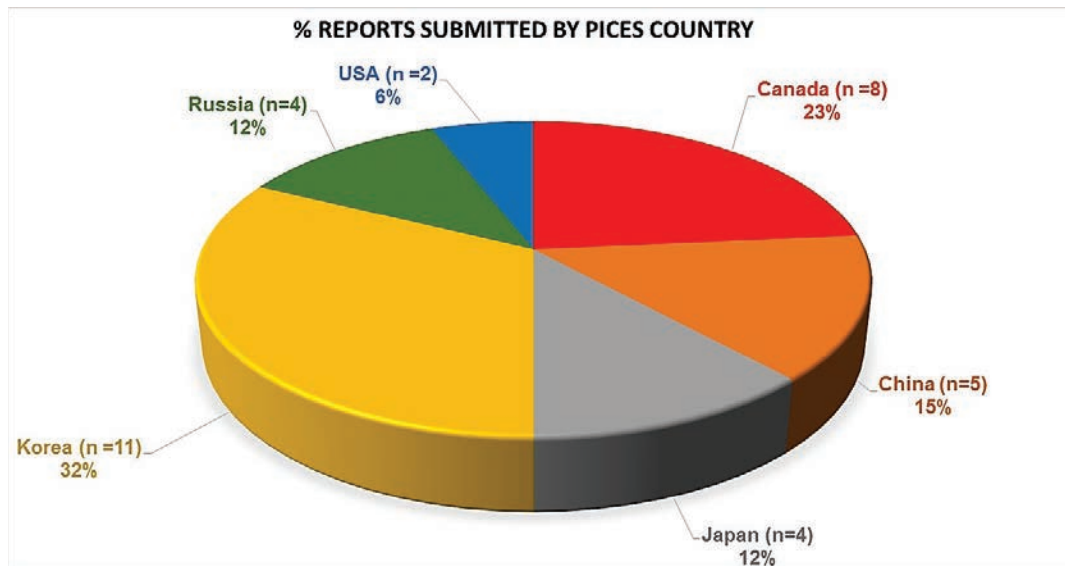


Fig. 1 Contribution (%) and number of pollution indicator reports for the North Pacific submitted by PICES member countries. Contributions were dependent on whether or not monitoring of a particular pollutant/class in different matrices was the subject of different indicator reports or included in one report.

Annex**Environmental Time Series Observations Guideline****Contribution content followed the following outline and definitions:**

1. Title: In title case (nouns, verbs capitalized; articles lowercase)
2. Contributed by: Authors of contribution
3. Contact author: Name of author to contact with questions
4. Any authors interested in involvement in synthesis report (optional)
5. Contact information: Affiliation, mailing address, email address
6. Last updated: Month, Year
7. Body of contribution: 1 page of text and 1 or 2 figures and/or tables. If possible, the text of the contribution should incorporate the following:
 - **Description of time series observation (ETSO):** a description of the ETSO including reference to methods, locating coordinates or polygon (decimal), and how the ETSO is useful for understanding climate change or its impacts. Describe length of time series and expected future status. Indicate level of quality control (preliminary or final).
 - **Status and trends:** the historical trends and current status of the ETSO
 - **Factors influencing observed trends:** potential causes for observed trends and current status
 - **Implications:** Briefly answer these questions: What are the implications or impacts of the observed trends on the ecosystem or ecosystem components? What do the trends mean? Why are they important? How can this information be used to inform policy makers' decisions?
8. Figure(s) and/or Table(s) that illustrate the index (indices). 1 or 2 figures and/or tables.
 - **Figures and Tables:** Send figures and tables as separate jpg, png, or pdf. You may also include them in the doc with the text, but they should also be sent separately in highest resolution possible. Format them as for journal publication; keep the final size in mind when considering readability of axis labels.
9. Literature Cited

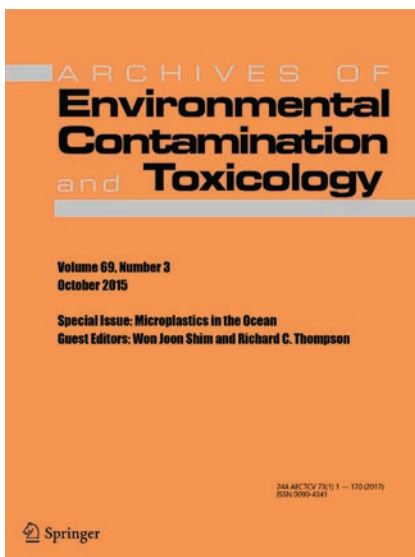
Each reference cited in text must be listed in the Citations section and vice versa. Double-check for spelling, dates, and other publication details.
10. Data contributions

You are requested to submit the data illustrated in the figures and tables, while also providing the metadata that allows another scientist to reproduce your results. If it is not possible to submit the data, please provide a statement explaining why it is not possible to do so.

Appendix 4

Journal Publications

Archives of Environmental Contamination and Toxicology, Vol. 69, Issue. 3, October 2015	
Special Issue – <i>Microplastics in the ocean</i>	126
Archives of Environmental Contamination and Toxicology, Vol. 73, Issue 1, July 2017	
Special Issue – <i>Oil spills and accidents</i>	127
Archives of Environmental Contamination and Toxicology, Vol. 73, Issue 2, August 2017	
Special Issue – <i>Indicators of ocean pollution</i>	129



Volume 69, Issue 3, October 2015

<https://link.springer.com/journal/244/69/3>

A special issue based on selected papers from the PICES-2014 Topic Session on “Marine debris in the ocean: Sources, transport, fate and effects of macro- and micro-plastic” (Issue Editors: Won Joon Shim and Richard C. Thompson). The issue highlights the high levels of contamination in the North Pacific both on shorelines and at the sea surface. The special issue also demonstrates potential for ingestion of microplastic by small planktonic organisms at the base of the food chain.

Microplastics in the Ocean

Won Joon Shim and Richard C. Thompson

Abundance and Distribution Characteristics of Microplastics in Surface Seawaters of the Incheon/Kyeonggi Coastal Region

Doo-Hyeon Chae, In-Sung Kim, Seung-Kyu Kim, Young Kyoung Song and Won Joon Shim

Occurrence and Distribution of Microplastics in the Sea Surface Microlayer in Jinhae Bay, South Korea

Young Kyoung Song, Sang Hee Hong, Mi Jang, Gi Myung Han and Won Joon Shim

Distribution and Size Relationships of Plastic Marine Debris on Beaches in South Korea

Jongmyoung Lee, Jong Su Lee, Yong Chang Jang, Su Yeon Hong, Won Joon Shim, Young Kyung Song, Sang Hee Hong, Mi Jang, Gi Myung Han, Daeseok Kang and Sunwook Hong

Factors Influencing the Spatial Variation of Microplastics on High-Tidal Coastal Beaches in Korea

In-Sung Kim, Doo-Hyeon Chae, Seung-Kyu Kim, SooBong Choi and Seung-Bum Woo

Intertidal Concentrations of Microplastics and Their Influence on Ammonium Cycling as Related to the Shellfish Industry

Melanie Cluzard, Tamara N. Kazmiruk, Vasily D. Kazmiruk & L. I. Bendell

Ingestion of Microplastics by Zooplankton in the Northeast Pacific Ocean

Jean-Pierre W. Desforges, Moira Galbraith and Peter S. Ross

Detection of Anthropogenic Particles in Fish Stomachs: An Isolation Method Adapted to Identification by Raman Spectroscopy

France Collard, Bernard Gilbert, Gauthier Eppe, Eric Parmentier and Krishna Das

Potential Threat of Microplastics to Zooplanktivores in the Surface Waters of the Southern Sea of Korea

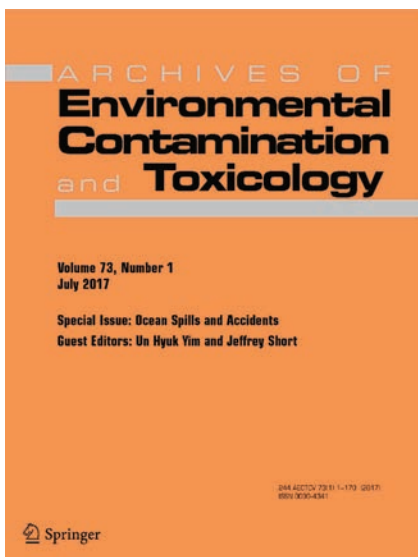
Jung-Hoon Kang, Oh-Youn Kwon and Won Joon Shim

Qualitative Analysis of Additives in Plastic Marine Debris and Its New Products

Manviri Rani, Won Joon Shim, Gi Myung Han, Mi Jang, Najat Ahmed Al-Odaini, Young Kyong Song and Sang Hee Hong

Estimation of the Environmental Load of High- and Low-Density Polyethylene From South Korea Using a Mass Balance Approach

Mijin Kim, Seunghun Hyun and Jung-Hwan Kwon



Volume 73, Issue 1, July 2017

<https://link.springer.com/journal/244/73/1/page/1>

PICES Working Group 31 on *Emerging Topics in Marine Pollution* published 14 papers and an introduction as a special issue of *Archives of Environmental Contamination and Toxicology* (Issue Editors: Un Hyuk Yim and Jeffrey Short) in July 2017. The special issue is titled *Ocean Spills and Accidents* and is based on a selection of papers from a workshop titled “Marine environmental emergencies: detection, monitoring, response and impacts” held at PICES-2015 in Qingdao. The goal of this issue is to provide an introduction to the important ways that oil spills may harm the biota, habitats and ecosystems using invited and targeted reviews complemented by original research articles.

Marine Environmental Emergencies in the North Pacific Ocean: Lessons Learned from Recent Oil Spills

Un Hyuk Yim & Jeffrey Short

Advances in Understanding the Fate and Effects of Oil from Accidental Spills in the United States Beginning with the *Exxon Valdez*

Jeffrey W. Short

The Toxicity to Fish Embryos of PAH in Crude and Refined Oils

Peter V. Hodson

Molecular Mechanisms of Crude Oil Developmental Toxicity in Fish

John P. Incardona

Challenges to Oil Spill Assessment for Seabirds in the Deep Ocean

J. Christopher Haney, Patrick G. R. Jodice, William A. Montevecchi & David C. Evers

Photoenhanced Toxicity of Petroleum to Aquatic Invertebrates and Fish

Mace G. Barron

Environmental Impacts and Recovery After the *Hebei Spirit* Oil Spill in Korea

U. H. Yim, J. S. Khim, J.-H. Jung & W. J. Shim

The Montara Oil Spill: A 2009 Well Blowout in the Timor Sea

R. B. Spies, M. Mukhtasor & K. A. Burns

Biomarkers of Aryl-hydrocarbon Receptor Activity in Gulf Killifish (*Fundulus grandis*) From Northern Gulf of Mexico Marshes Following the Deepwater Horizon Oil Spill

Benjamin Dubansky, Charles D. Rice, Lester F. Barrois & Fernando Galvez

Anomalously High Recruitment of the 2010 Gulf Menhaden (*Brevoortia patronus*) Year Class: Evidence of Indirect Effects from the *Deepwater Horizon* Blowout in the Gulf of Mexico

Jeffrey W. Short, Harold J. Geiger, J. Christopher Haney, Christine M. Voss, Maria L. Vozzo, Vincent Guillory & Charles H. Peterson

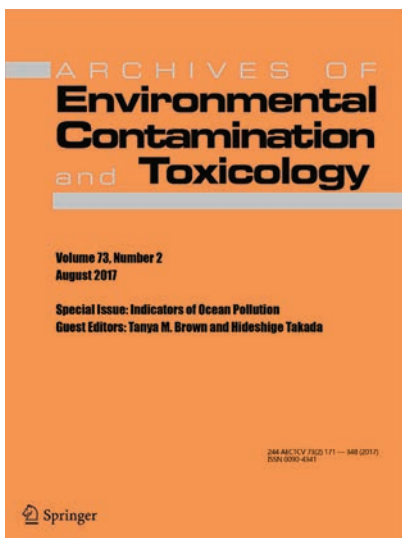
Long-Term Monitoring of PAH Contamination in Sediment and Recovery After the Hebei Spirit Oil Spill
Moonkoo Kim, Jee-Hyun Jung, Sung Yong Ha, Joon Geon An, Won Joon Shim & Un Hyuk Yim

Contamination and Human Health Risk Assessment of Polycyclic Aromatic Hydrocarbons (PAHs) in Oysters After the *Wu Yi San* Oil Spill in Korea
Andrew Loh, Un Hyuk Yim, Sung Yong Ha, Joon Geon An & Moonkoo Kim

Identification of Spilled Oil from the *MV Marathassa* (Vancouver, Canada 2015) Using Alkyl PAH Isomer Ratios
Carmen Morales-Caselles, Mark B. Yunker & Peter S. Ross

Oil Spills and Marine Mammals in British Columbia, Canada: Development and Application of a Risk-Based Conceptual Framework
Adrienne L. Jarvela Rosenberger, Misty MacDuffee, Andrew G. J. Rosenberger & Peter S. Ross

Detection and Monitoring of Oil Spills Using Moderate/High-Resolution Remote Sensing Images
Ying Li, Can Cui, Zexi Liu, Bingxin Liu, Jin Xu, Xueyuan Zhu & Yongchao Hou



Volume 73, Issue 2, August 2017

<https://link.springer.com/journal/244/73/2/page/1>

PICES Working Group 31 on *Emerging Topics in Marine Pollution* published 15 papers and an introduction as a special issue of *Archives of Environmental Contamination and Toxicology* (Issue Editors: Tanya M. Brown and Hideshige Takada) in August 2017. The special issue, titled *Indicators of Ocean Pollution in the North Pacific Ocean*, is based on a selection of papers from a scientific session on “Indicators of emerging pollution issues in the North Pacific Ocean” held at PICES-2015 in Qingdao, China. This issue provides the first compilation of research on marine pollution indicators in the North Pacific Ocean and provides guidance to inform mitigation and monitoring of contaminants in the region.

Indicators of Marine Pollution in the North Pacific Ocean

Tanya M. Brown, Hideshige Takada

Bioindicators of Organochlorine Pesticides in the Sea of Okhotsk and the Western Bering Sea

Vasilii Yu. Tsygankov, Margarita D. Boyarova, Olga N. Lukyanova and Nadezhda K. Khristoforova

Spatial Distribution and Temporal Trend of Anthropogenic Organic Compounds Derived from the 2011 East Japan Earthquake

Kaoruko Mizukawa, Yasuko Hirai, Hiroyuki Sakakibara, Satoshi Endo, Keiji Okuda, Hideshige Takada, Naoko Murakami-Sugihara, Kotaro Shirai and Hiroshi Ogawa

Polycyclic Aromatic Hydrocarbons (PAHs) and Hopanes in Plastic Resin Pellets as Markers of Oil Pollution via International Pellet Watch Monitoring

Bee Geok Yeo, Hideshige Takada, Junki Hosoda, Atsuko Kondo, Rei Yamashita, Mahua Saha and Thomas Maes

Time Trends of Persistent Organic Pollutants in Benthic and Pelagic Indicator Fishes from Puget Sound, Washington, USA

James E. West, Sandra M. O'Neill and Gina M. Ylitalo

Microplastics in Sediment Cores from Asia and Africa as Indicators of Temporal Trends in Plastic Pollution

Yukari Matsuguma, Hideshige Takada, Hidetoshi Kumata, Hirohide Kanke, Shigeaki Sakurai, Tokuma Suzuki, Maki Itoh, Yohei Okazaki, Ruchaya Boonyatumanond, Mohamad Pauzi Zakaria, Steven Weerts and Brent Newman

Dissolved Platinum Concentrations in Coastal Seawater: Boso to Sanriku Areas, Japan

Asami Suzuki Mashio, Hajime Obata and Toshitaka Gamo

The Glaucous-Winged Gull (*Larus glaucescens*) as an Indicator of Chemical Contaminants in the Canadian Pacific Marine Environment: Evidence from Stable Isotopes

M. L. Davis, J. E. Elliott and T. D. Williams

Effects of Feeding Strategy, Sediment Characteristics, and Chemical Properties on Polychlorinated Biphenyl and Polybrominated Diphenyl Ether Bioaccumulation from Marine Sediments in Two Invertebrates

H. Frouin, P. Jackman, N. D. Dangerfield and P. S. Ross

A Risk-Based Characterization of Sediment Contamination by Legacy and Emergent Contaminants of Concern in Coastal British Columbia, Canada

Carmen Morales-Caselles, Jean-Pierre W. Desforges, Neil Dangerfield & Peter S. Ross

Comparison of Toxicities of Metal Pyrithiones Including Their Degradation Compounds and Organotin Antifouling Biocides to the Japanese Killifish *Oryzias latipes*

Madoka Ohji and Hiroya Harino

Oxidative Stress in Shellfish *Sinonovacula constricta* Exposed to the Water Accommodated Fraction of Zero Sulfur Diesel Oil and Pinghu Crude Oil

Mei Jiang, Lei Li, Yingren Li, Gongming Shen and Xinqiang Shen

Imposex in *Reishia clavigera* as an Indicator to Assess Recovery of TBT Pollution After a Total Ban in South Korea

Nam Sook Kim, Sang Hee Hong, Kyung-Hoon Shin and Won Joon Shim

Polychlorinated Biphenyl-Related Alterations of the Expression of Essential Genes in Harbour Seals (*Phoca vitulina*) from Coastal Sites in Canada and the United States

Marie Noël, Neil Dangerfield, Steve Jeffries, Dyanna Lambourn, Monique Lance, Caren Helbing, Michel Lebeuf and Peter S. Ross

Sea Urchin Embryogenesis as Bioindicators of Marine Pollution in Impact Areas of the Sea of Japan/East Sea and the Sea of Okhotsk

Olga N. Lukyanova, Elena V. Zhuravel, Denis N. Chulchekov and Andrey A. Mazur

Tissue Contaminant Burdens in San Francisco Estuary White Sturgeon (*Acipenser transmontanus*): Implications for Population Recovery

Deke T. Gundersen, Steven C. Zeug, Robert B. Bringolf, Joseph Merz, Zachary Jackson and Molly A. H. Webb

Appendix 5

Topic Session/Workshop Summaries and Meeting Reports from Past Annual Meetings

PICES-2014, Yeosu, Korea	
Topic Session on “ <i>Marine debris in the ocean: Sources, Transport, fate and effects of macro- and micro-plastics</i> ”	132
Meeting Report.....	134
PICES-2015, Qingdao, China	
Topic Session on “ <i>Indicators of emerging pollution issues in the North Pacific Ocean</i> ”	139
Workshop on “ <i>Marine environment emergencies: Detection, monitoring, response, and impacts</i> ”	141
Meeting Report.....	144
PICES-2016, San Diego, USA	
Topic Session on “ <i>Source, transport and fate of hydrocarbons in the marine environment</i> ”	150
Meeting Report.....	152
PICES-2017, Vladivostok, Russia	
Topic Session on “ <i>Microplastics in marine environments: Fate and effects</i> ”	159

PICES-2014, Yeosu, Korea

October 16–26, 2014

Excerpted from:

Summary of Scientific Sessions and Workshops at PICES-2014

MEQ Topic Session (S8)

Marine debris in the Ocean: Sources, transport, fate and effects of macro- and micro-plastics

Co-sponsored by the Group of Experts on Scientific Aspects of Marine Pollution (GESAMP) the International Council for the Exploration of the Sea (ICES) and the Northwest Pacific Action Plan (NOWPAP)

Co-Convenors: *Won Joon Shim (Korea), Peter S. Ross (Canada), Olga Lukyanova (Russia), Sangjin Lee (NOWPAP), Peter Kershaw (GESAMP), Jesus Manuel Gago Piñeiro (Spain / ICES)*

Invited Speakers:

Marcus Eriksen (5 Gyres Institute, USA)

Francois Galgani (Institut Français de Recherche pour l'Exploitation de la Mer (IFREMER), France)

Sunwook Hong (Our Sea of East Asia Network (OSEAN), Korea)

Hideshige Takada (Tokyo University of Agriculture and Technology, Japan)

Background

Marine debris is increasingly recognized as a threat to biota in the ocean, which can have a range of socio-economic impacts from coastal areas to the open ocean. The majority of marine debris consists of synthetic polymers, or 'plastics', which readily float on the ocean surface or are suspended in the water column. Microplastics may be attributed to the intentional manufacture of commercial products or the fragmentation of plastic products. They can increase the bioavailable fraction of marine litter and act as a vector for the delivery of intrinsic or adsorbed toxic chemicals to exposed biota. Floating, submerged and beached debris have been documented in marginal seas and the adjacent coastal zone of the North Pacific Ocean. In addition, the North Pacific Ocean Gyre is known to accumulate floating debris in what has become known as the "Great Pacific Garbage Patch". Marine debris represents trans-boundary pollution which can also deliver associated chemicals and invasive organisms to regions far removed from source. The objective of this session is to present status and trend information for marine plastic debris pollution and its environmental consequences in the PICES region. Papers are invited that assess macro- or micro-plastic debris 1) hotspots in the PICES region, 2) source and input pathways, 3) long-range transport, 4) role as sink or source of associated toxic chemicals, and 5) biological and ecological effects. Recommendations on how to address growing problems associated with marine debris will be also considered.

List of papers*Oral presentations***Patterns of microplastic distribution in the global ocean and inland environments (Invited)**

Marcus [Eriksen](#)

Litter in the Mediterranean Sea within the European Marine Strategy Framework Directive (MSFD): Indicators for descriptor 10, GES and monitoring (Invited)

Francois [Galgani](#)

Microlitter: Recommendations for monitoring from the MSFD

Jesus [Gago](#), Richard C. Thompson, Francois Galgani and T. Maes

Results and lessons learned from joint beach debris surveys by Asian NGOs (Invited)

Yong Chang Jang, Sunwook [Hong](#), Jongmyoung Lee, Jong Su Lee, Sang Hee Hong, Won Joon Shim, Martin Thiel, Fujieda Shigeru, Tai-di Chang, Kanyarat Kosavisutte and Trieu Thuy Ha

Hazardous chemicals in plastics in marine environments and their potential effects on marine organisms (Invited)

Hideshige [Takada](#), Kosuke Tanaka, Rei Yamashita and Yutaka Watanuki

Marine litter problem at the Russian Far East and approaches for solution

Iana [Blinovskaia](#)

Distribution, sources and abundance of Marine debris in the coastal area of southern part of Primorsky Krai (Russia)

Nikolai [Kozlovskii](#)

Sequential monitoring of beach litter at multiple sites using webcams

Tomoya [Kataoka](#), Hirofumi Hinata and Shin'ichiro Kako

Inverse estimation of marine-debris outflows using webcam observation data

Shin'ichiro [Kako](#), Atsuhiko Isobe, Tomoya Kataoka and Hirofumi Hinata

Distribution and impacts of micro- and macro-plastics in coastal British Columbia, Canada

Peter S. [Ross](#), Jean-Pierre W. Desforges, Jean Fong, Moira Galbraith and Wendy Szanizlo

Producing fragmented micro- and nano-plastics from expanded polystyrene with an accelerated mechanical abrasion experiment

Won Joon [Shim](#), Young Kyoung Song, Sang Hee Hong, Mi Jang, Gi Myung Han and Seung Won Jung

Analysis of a beach as a time-invariant linear input/output system of marine litter

Tomoya Kataoka, Hirofumi [Hinata](#) and Shigeru Kato

Chemicals of concern in plastic marine debris: Hexabromocyclododecanes in expanded polystyrene Products

Sang Hee [Hong](#), Won Joon Shim, Manviri Rani, Mi Jang, Najat Ahmed Al-Odaini, Gi Myung Han and Young Kyoung Song

Leaching characteristics of Hexabromocyclododecane from expanded polystyrene buoy fragments in marine water

Manviri [Rani](#), Won Joon Shim, Mi Jang, Gi Myung Han, Young Kyoung Song and Sang Hee Hong

Persistent organic pollutants adsorbed on microplastic from two beaches in China

Weiwei [Zhang](#), Zhifeng Zhang, Xindong Ma, Yan Wang and Ling Qu

Transport of marine debris from the 2011 tsunami in Japan: Model simulations and observational evidence

Jan [Hafner](#), Nikolai Maximenko and Gisela Speidel

Selective transport of microplastics and mesoplastics by drifting in coastal waters

Atsuhiko [Isobe](#), Kenta Kubo, Yuka Tamura, Shin'ichiro Kako, Etsuko Nakashima and Naoki Fujii

*Poster presentations***Monitoring of traditional and emerging BFRs in expanded polystyrene (EPS) from various countries**

Manviri [Rani](#), Sang Hee Hong, Mi Jang, Gi Myung Han and Won Joon Shim

Expanded polystyrene buoy as a moving source of toxic chemicals to marine life: Enrichment of hexabromocyclododecanes in mussel

Mi [Jang](#), Sang Hee Hong, Manviri Rani, Gi Myung Han, Young Kyoung Song and Won Joon Shim

Compositions and distributions of microplastic in Korean beaches

Young Kyoung [Song](#), Mi Jang, Gi Myung Han, Sang Hee Hong, and Won Joon Shim

A Nile Red staining method for microplastic identification and quantification

Young Kyoung Song, Sang Hee Hong, Mi Jang, Gi Myung Han and Won Joon Shim

Potential threat of microplastics to neustonic zooplankton in surface waters of the Southern Sea of Korea

Jung-Hoon Kang, Oh-Youn Kwon, Bu-Gyeong Choi and Won Joon Shim

Estimation of used buoy debris outflow from oyster aquaculture with Material Flow Analysis

Su Yeon Hong, Yong Chang Jang, Jongmyoung Lee, Sunwook Hong and Chan Won Lee

Conceptual difference of flow and stock of marine debris and its implications to setting of policy goals and indicators

Yong Chang Jang, Sunwook Hong, Jongmyoung Lee, Hyun Woo Choi and Won Joon Shim

Abundance of packaging debris on beaches increases as non-recycled amount increases: A case in Korea

Yong Chang Jang, Jongmyoung Lee, Sunwook Hong, Jong Su Lee, Su Yeon Hong and Won Joon Shim

The distribution of floating macro- and micro-plastics in the open ocean and Large Marine Ecosystems

Peter J. Kershaw, Courtney Arthur, Marcus Eriksen, Jesús M. Gago Piñeiro, Kara Lavender Law and Laurent Lebreton

Sources, fate and effects of micro-plastics in the marine environment – A global assessment

Peter J. Kershaw, A. Andrady, C. Arthur, J. Baker, H. Boumann, S. Gall, V. Hidalgo-Ruz, A. Koehler, K.L. Law, H. Leslie, J. Potemra, P. Ryan, W.J. Shim, H. Takada, R. Thompson, A. Turra, D. Verthaa and K. Whyles

Report of Working Group 31 on *Emerging Topics in Marine Pollution*

The Working Group on *Emerging Topics in Marine Pollution* (WG 31) met from 9:00 to 18:00 h on October 18, 2014 in Yeosu, Korea, under the chairmanship of Drs. Won Joon Shim (Korea) and Olga Lukyanova (Russia). The meeting objective was to review activities during the first year (2013–2014) of WG 31, plan for activities during the second year (2014–2015), and discuss the strategy and future plan to compile marine pollution data in the Pacific region.

AGENDA ITEM 1

Welcome and adoption of agenda

WG 31 membership was finalized on January 21, 2014 and updated on May 21, 2014. Ten out of 17 WG members from all six member countries participated the meeting (*WG 31 Endnote 1*). After review by members, the draft agenda was slightly modified and adopted (*WG 31 Endnote 2*).

AGENDA ITEM 2

Introduction of WG 31 mission

It was the first meeting of WG 31. Thus, the background, mission, terms of reference, future activity and linkage to FUTURE program of the WG were reviewed by Dr. Shim.

AGENDA ITEM 3

Review of the main achievements of WG 31 in 2014

A 1-day MEQ Topic Session (S8) in 2014 meeting, entitled “*Marine debris in the ocean: Sources, transport, fate and effects of macro- and micro-plastics*” was held at PICES-2014. Seventeen oral

presentations, including four invited papers, and 10 poster presentations (total 27) were accommodated. The session was co-sponsored by ICES, NOWPAP and GESAMP.

A special issue of the scientific journal *Archives of Environmental Contamination and Toxicology* (Springer/2013 IF = 1.960) will feature selected submissions from Topic Session S8. Dr. Shim is serving as a co-Guest Editor of this special issue. Publication is set for late 2015. The submission deadline is the end of February 2015. A total of 14 papers are expected.

AGENDA ITEM 4

Topic session and workshop proposals for PICES-2015

A 1-day Topic Session proposal entitled “*Indicators of emerging pollution issues in the North Pacific Ocean*” was proposed by co-convenors, Drs. Peter Ross, Olga Lukyanova and Won Joon Shim (WG 31 *Endnote 3*), and supported by the WG members. Travel funds are requested for 3 invited speakers. Presenters will be invited to submit papers to a special issue of a scientific journal. It is expected that data and results from this session will be important as WG 31 supports the needs of FUTURE plans and reporting.

A 1-day workshop on “*Marine environment emergencies: Detection, monitoring and response*” was proposed by co-convenors, Drs. Ziwei Yao, Peter Ross, Won Joon Shim, and Olga Lukyanova, and forms part of the established 3-year plan of activities for WG 31. The workshop was supported by the WG members under the condition that it be combined with another oil spill response-related workshop proposed by NOWPAP.

AGENDA ITEM 5

Establish strategy and plan in data compilation for PICES status report

The WG spent half a day discussing and compiling emerging marine pollution data in the North Pacific region. Four target pollutants were selected based on the SG-MP study conducted by the Study Group on *Marine Pollutants* (PICES Scientific Report No. 46): (1) Persistent, bioaccumulative and toxic (PBT) substances, (2) Hydrocarbons (*e.g.*, PAHs), (3) Metals or elements of concerns, and (4) Marine debris and microplastics.

The data on the four target pollutant classes will be obtained from (i) scientific papers and reports, (ii) governments, organizations, and scientists, (iii) the existing database from regional monitoring programs.

WG members were divided into four task force groups:

- (1) PBT substances: Peter Ross (Canada), John Elliot (Canada), Guangshui Na (China), Hideshige Takada (Japan), Hyo-Bang Moon (Korea), Olga Lukyanova (Russia), Gina Ylitalo (USA);
- (2) Hydrocarbons: Kazuhiko Mochida (Japan), Hideaki Maki (Japan), Un Hyuk Yim (Korea), Staci Simonich (USA), Gina Ylitalo (USA);
- (3) Metals or elements of concerns: Don-Woon Hwang (Korea), Mikhail Simokon (Russia);
- (4) Marine debris and microplastics; Won Joon Shim (Korea), Nancy Wallace (USA).

The timeframe for data compilation was discussed and tentatively established as follows:

- December 2014: Selection of indicators for each emerging pollution issues;
- March 2015: Collection of available data for indicators;
- June 2015: first draft for data interpretation and feedback from the WG 31 members;
- September 2015: second draft for data interpretation and visualization;
- October 2015: Sharing of results at WG 31 meeting and feedback (PICES-2015).

AGENDA ITEM 6

Preparation of WG 31 report for FUTURE and MEQ meeting

The major items of the WG 31 report were discussed and finalized for presentation at the joint FUTURE Advisory Panel meeting and at the Committee meeting of its parent, MEQ.

AGENDA ITEM 7

Relations with other groups/organizations

The relationship between WG 31 activities and other groups within and outside of PICES was discussed.

AGENDA ITEM 8

Other business

There was no additional business to discuss and thus, the meeting adjourned at 18:00 h.

WG 31 Endnote 1

WG 31 participation list

Members

John Elliott (Canada)
 Olga Lukyanova (Russia, Co-Chair)
 Hideaki Maki (Japan)
 Kazuhiko Mochida (Japan)
 Guangshui Na (China)
 Won Joon Shim (Korea, Co-Chair)
 Hideshige Takada (Japan)
 Nancy Wallace (USA)
 Un Hyuk Yim (Korea)
 Gina Ylitalo (USA)

Observers

Karin Baba (Japan)
 Chuanlin Huo (China)
 K. Ishimaru (Japan)
 Peter Kerwshaw (GESAMP)
 Yutaka Watanuki (Japan)

WG 31 Endnote 2**WG 31 meeting agenda**

Saturday, October 18, 2014

[09:00 – 12:30] Morning session

1. Welcome and adoption of agenda (Co-Chairs)
Welcome and agenda outline
2. Introduction of WG 31 mission (All)
Introduce and share background, mission, terms of reference and plans for WG 31
3. Review of main achievements of WG 31-2014 (Co-Chairs)
Summarize the main achievements of WG 31 in the past 1 year
4. Proposals of new topics and direction (Co-Convenors)
Introduction of the proposed topic session and workshop for 2015 and discussion

[14:00 – 18:00] Afternoon session

5. Design strategy for data compilation plans for PICES status report (All)
6. Selection of topics (refer to SG-MP report), timeframe, and data compilation methods
7. Preparation of WG 31 report for MEQ meeting (All)
8. Relations with other groups/organizations (All)
9. Introduce the progress on their work with other groups or other organizations.
10. Other business (All)

WG 31 Endnote 3

Proposal for a 1-day MEQ Topic Session on “*Indicators of emerging pollution issues in the North Pacific Ocean*” at PICES-2015

Co-sponsors: NOWPAP

Co-Convenors: Olga Lukyanova (Russia), Peter S. Ross (Canada), Won Joon Shim (Korea)

This session led by the Working Group on *Emerging Topics in Marine Pollution* (WG-ETMP) anticipates wide-ranging interest from a number of disciplines. The session aims to attract presentations on the use of sediments, shellfish, fish, seabirds, and marine mammals as indicators of marine pollution. Novel approaches and study designs will be discussed, with the aim of providing managers, regulators and scientists with timely feedback on emerging pollution threats. Depending on the study design and sample matrix, it is expected that pollutants to be discussed will include hydrocarbons, organochlorine pesticides, flame retardant chemicals, metals, pharmaceuticals, microplastics and other contaminants. Presentations that provide insight into the identification of contaminants of emerging concern, the ranking of priority pollutants from multiple sources, and the assessment of the relative importance of pollutants among other natural and anthropogenic stressors are encouraged. Presenters will be invited to contribute to a special issue of a scientific journal.

Proposal for 1-day Workshop on “*Marine environment emergencies: Detection, monitoring and response*” at PICES-2015

Co-sponsors: NOWPAP, ICES

Co-Convenors: Ziwei Yao (China), Seong-Gil Kang (Korea/NOWPAP), Peter Ross (Canada), Won Joon Shim (Korea), Olga Lukyanova (Russia)

In recent years, the importance of marine environmental emergency issues has been illustrated by oil and chemical spills, as well as by a major nuclear power plant accident. Globalization of markets has led to rapid growth of maritime transport in the North Pacific, which has become more vulnerable to ship-source incidents, including oil and hazardous and noxious substances (HNS) spills. Oil and HNS spills may be hazardous to human health, harm living resources and marine life, and can damage amenities or interfere with other legitimate uses of the sea. In 2003, the NOWPAP Regional Oil and HNS Spill Contingency Plan (RCP) provided technical and operational guidelines for regional cooperation in responding to oil and HNS spills. Marine environmental emergency issues and their strategies become an increasingly important topic for PICES member countries. However, accepted scientific and monitoring methods to document the ecological impacts of such emergencies, and post-accident recovery of the environment, are lacking. In order to better understand the interaction between the marine ecosystem and human pressures, and to formulate sustainable marine development strategies more effectively, an applied information sharing workshop for PICES is timely. The workshop on marine environmental emergencies has three objectives. The first is to summarize important examples of North Pacific marine environmental emergencies from the perspective of different nations, and to discuss the different approaches taken by PICES member countries. The second is to develop response strategies and capacities of PICES members in light of environmental emergencies. The third is to develop joint strategies to improve responsiveness and effectiveness of current national approaches to manage and mitigate such emergencies in the PICES region. The workshop will address the following three aspects: (1) oil and chemical spills and their damage on the marine environment, (2) detection methods for oil and chemical spills and (3) spill response, monitoring and mitigation strategies at the interface of science and management. Case studies will be used to illustrate this workshop and will serve to focus efforts to design a response and monitoring framework for implementation in the event of a major environmental emergency.

PICES-2015, Qingdao, China

October 15–25, 2015

Excerpted from:

Summary of Scientific Sessions and Workshops at PICES-2015

MEQ Topic Session (S4)

Indicators of emerging pollution issues in the North Pacific Ocean

Co-sponsor: Northwest Pacific Action Plan (NOWPAP)

Co-Convenors: *Peter S. Ross (Canada) and Olga Lukyanova (Russia)*

Invited Speakers:

Tomohiko Isobe (National Institute for Environmental Studies, Japan)

Hyo-Bang Moon (NOWPAP, Korea)

Vladimir Shulkin (NOWPAP, Russia)

Hideshige Takada (NOWPAP, Japan)

Background

This session, led by the Working Group on *Emerging Topics in Marine Pollution* (WG-ETMP), provided a forum for wide-ranging interests from a number of disciplines. The purpose of the session was to attract presentations on the use of sediments, shellfish, fish, seabirds, and marine mammals as indicators of marine pollution, and to discuss novel approaches and study designs, with the aim of providing managers, regulators and scientists with timely feedback on emerging pollution threats. The session was open to discussion of a wide range of pollutants including hydrocarbons, organochlorine pesticides, flame retardant chemicals, metals, pharmaceuticals, microplastics and other contaminants. Presentations that provided insight into the identification of contaminants of emerging concern, the ranking of priority pollutants from multiple sources, and the assessment of the relative importance of pollutants among other natural and anthropogenic stressors were encouraged, and presenters were invited to contribute to a special issue of a scientific journal.

List of papers

Oral presentations

Microplastic: An emerging threat to marine environment and a new tool for POP monitoring (Invited)

Hideshige Takada, Rei Yamashita, Yohei Okazaki, Bee Geok Yeo, Ryu Yoshida, Atsuko Kondo and Yu Saito

Spatial variability of trace metal concentrations in different mussels from coastal areas of the northwestern Pacific Ocean (Invited)

Vladimir M. Shulkin and Victor Ya. Kavun

Levels and temporal trends of legacy and emerging POPs in small cetacean species from Japan (Invited)

Tomohiko Isobe, Yuko Tajima, Tadasu K. Yamada, Masao Amano, Takashi Matsuishi, Tatsuya Kunisue and Shinsuke Tanabe

Monitoring of legacy and emerging contaminants in wildlife from Korea (Invited)

Yunsun Jeong, Hyun-Kyung Lee, Xiangzi Jin, Sunggyu Lee and Hyo-Bang Moon

Microplastic is the macroproblem of the world's oceans, including the Russian Far EastIana Blinovskaia**At-sea distributions reveal where Cassin's Auklets are exposed to microplastics in the fall in British Columbia 2014**Patrick D. O'Hara, Jocelyn Wood, Stephanie Avery-Gomm, Laurie Wilson, Ken H. Morgan and Peter S. Ross**Spatial and temporal mercury trends in seabird eggs from Pacific Canada 1968-2012 are due to diet: Evidence from sulfur isotopes**Kyle H. Elliott and John E. Elliott**Mercury speciation and major factors controlling the cycling of methylmercury in the Yellow Sea and Bohai Sea, China**Yanbin Li, Lufeng Chen and Yongguang Yin**Persistent organic pollutants in the food chain: Salmon, seabirds and marine mammals from the North-West Pacific (Russian Far East)**Vasiliy Yu. Tsygankov, Margarita D. Boyarova, Peter A. Tyupelev, Ilya A. Shcherbakov, Olga N. Lukyanova and Nadezhda K. Khristoforova**Emerging ocean pollution issues in the NE Pacific Ocean vary by matrix: Lessons from sediments, shellfish and marine mammals**Peter S. Ross, Carmen Morales-Caselles and Marie Noel**Bioindicators of marine pollution in impacted areas of the Sea of Okhotsk**Olga N. Lukyanova, Elena V. Zhuravel, Denis N. Chulchekov, Olga V. Podgurskaya and Andrey A. Mazur*Poster presentations***Acute effects of emamectin benzoate on the calanoid copepod *Pseudodiaptomus poplesia***Xiaoyan Yi, Yunyun Zhuang, Hongju Chen, Yousong Huang, Feifei Yang, Huan Zhang and Guangxing Liu**Phototransformation of oxytetracycline in saline waters under simulated sunlight irradiation: Kinetics, mechanism and products**Cui Zhang, Xuefeng Hu and Yongming Luo*Presented by Qian Zhou on behalf of Cui Zhang***Assessment of marine environment quality of the coastal zone of Peter the Great Bay (the Sea of Japan/East Sea)**Tatyana A. Belan, Tatyana S. Lishavskaya, Alexander V. Sevastianov, Tatyana V. Chatkina and Boris M. Borisov*Presented by Olga Lukyanova on behalf of Tatyana Belan***Metal concentrations in pink and chum salmon (Kuril Islands, the North Western Pacific)**Vasiliy Yu. Tsygankov, Nadezhda K. Khristoforova, Margarita D. Boyarova and Olga N. Lukyanova**Estimation of seawater pollution in Uglovoy Bay (Peter the Great Bay, Japan/East Sea)**Valery I. Petukhov, Oleg V. Losev and Evgeniya A. Tikhomirova*Presented by I. Blinovskaya on behalf of O. Losev*

Workshop (W4)***Marine environment emergencies: Detection, monitoring, response, and impacts***

Co-sponsors:

International Council for the Exploration of the Sea (ICES)

Northwest Pacific Action Plan (NOWPAP)

Co-Convenors: *Ziwei Yao (China), Seong-Gil Kang (Korea/NOWPAP), Peter Ross (Canada), Olga Lukyanova (Russia)*

Invited Speakers:

Seong Gil Kang (Korea/NOWPAP)

Yongge Sun (Zhejiang University, China)

Background

In recent years, the importance of marine environmental emergency issues has been illustrated by oil and chemical spills, as well as by a major nuclear power plant accident. Globalization of markets has led to rapid growth of maritime transport in the North Pacific, which has become more vulnerable to ship-source incidents, including oil and hazardous and noxious substances (HNS) spills. Oil and HNS spills may be hazardous to human health, harm living resources and marine life, and can damage amenities or interfere with other legitimate uses of the sea. In 2003, the NOWPAP Regional Oil and HNS Spill Contingency Plan (RCP) provided technical and operational guidelines for regional cooperation in responding to oil and HNS spills. Marine environmental emergency issues and their strategies become an increasingly important topic for PICES member countries. However, accepted scientific and monitoring methods to document the ecological impacts of such emergencies, and post-accident recovery of the environment, are lacking. In order to better understand the interaction between the marine ecosystem and human pressures, and to formulate sustainable marine development strategies more effectively, an applied information sharing workshop for PICES is timely. The workshop on marine environmental emergencies had three objectives:

- 1) Summarize important examples of North Pacific marine environmental emergencies from the perspective of different nations, and to discuss the different approaches taken by PICES member countries;
- 2) Develop response strategies and capacities of PICES member countries in light of environmental emergencies;
- 3) Develop joint strategies to improve responsiveness and effectiveness of current national approaches to manage and mitigate such emergencies in the PICES region.

The workshop addressed the following three aspects: (1) oil and chemical spills and their damage on the marine environment, (2) detection methods for oil and chemical spills and (3) spill response, monitoring and mitigation strategies at the interface of science and management. Case studies were used to illustrate this workshop and served to focus efforts to design a response and monitoring framework for implementation in the event of a major environmental emergency.

Summary of the workshop

This 1-day workshop was held on October 15, as part of the 2015 PICES Annual Meeting in Qingdao, China. The event was co-sponsored by the International Council for the Exploration of the Sea (ICES) and the Northwest Pacific Action Plan (NOWPAP). The meeting was co-convened by Drs. Ziwei Yao

(China), Seong-Gil Kang (Korea/NOWPAP), Peter S. Ross (Canada), and Olga Lukyanova (Russia). Invited speakers were Drs. Seong-Gil Kang (Korea/NOWPAP) and Yongge Sun (Zhejiang University, China).

More than 40 researchers attended W4, with a total of 10 oral presentations (2 invited) given. Topics included presentations on geochemical and microbial community response oil spills, marine pollution preparedness and response to oil and Hazardous and Noxious Substance (HNS) spill incidents, lessons on oil spill response, examples of a number of oil spills in the North Pacific Ocean, determination of oil fingerprints, adverse effects, and prediction models.

The workshop featured presentations from five PICES member countries (USA excepted) as well as from NOWPAP. These included talks from Yongge Sun, Seong Gil Kang, Peter S. Ross, Chuanyuan Wang, Adedayo Adeleye, Zhen Wang, Patrick O'Hara, Un Hyuk Yim, Yongliang Wei, and Yoon Young Back.

Based on the oral presentations and workshop theme, several hot topics and related questions were posed and reviewed. A constructive and forward-looking roundtable discussion followed these presentations, with salient points summarized below.

I) Data:

- 1) Data is everything. Participants recognized that high resolution (fingerprinting) data were key to identifying the source, tracking the event, documenting the impact, and monitoring the recovery following a spill.
- 2) Data ownership was recognized as a frequent point of contention. Polluters ('Responsible Parties') are typically responsible for much monitoring, but such results are owned by a private party that may well face litigation or charges. This highlighted the importance of independent, third party data that are open to all parties and comes from scientifically-defensible sampling, analysis and interpretation efforts.
- 3) Pre-spill baselines are important. In a time of diminishing support for environmental monitoring, it was recognized that having baseline signatures for pre-existing levels and profiles of a variety of contaminant classes, including hydrocarbons, is very important. *Post-hoc* evaluation can garner some insight *via* depositional sediment core records.

II) Response:

- 1) The operational spill response is a short-term emergency effort. Who leads this operational spill response? Who leads discussions /evaluation of dispersant decisions? Are samples being collected during an emergency for environmental monitoring? For enforcement?
- 2) Environmental monitoring needs to be in place for the 'long-term'. Who leads this monitoring? How many samples of what matrix will be collected over space and time? Who decides on the extent of the monitoring? Are data freely available? Who pays for this monitoring?
- 3) How does one go about establishing recovery goals following a spill? Environmental recovery goals provide guidance for those overseeing the cleanup as well as other stakeholders (users). Which Environmental Quality Guidelines should be used? Are they adequate in protecting all valued species? Should new EQGs be developed?

III) Research needs / gaps:

- 1) Impacts of dispersants on the environment;
- 2) Fate and weathering of dispersants;
- 3) Environmental Quality Guideline development for hydrocarbons;
- 4) Molecular and isotopic fingerprinting methods (*e.g.*, new techniques to distinguish between crude and fuel oils);

- 5) Toxicology of hydrocarbons and other contaminants (invertebrates, fish, birds, marine mammals);
- 6) Fate of spilled oil and chemicals;
- 7) Remediation techniques;
- 8) Prediction models;
- 9) Ecological effects assessment methods;
- 10) Rapid (onsite, *in situ*) detection methods (*e.g.*, fluorescence-based methods).

Participants agreed to propose a Special Issue for a scientific journal resulting from this workshop, including additional submissions from researchers that were unable to participate in this session. The topic would be “*Environmental emergencies in the North Pacific*”. It was recognized that the 2016 PICES Annual meeting is likely to feature a complementary topic session on “*Source, transport, fate and effects of hydrocarbons in the North Pacific Ocean*”. The distinction between the two is clear: the first relates to immediate and catastrophic spills, while the latter relates to chronic, cumulative releases from multiple non point and point sources. The latter topic session was proposed to also provide for a Special Journal Issue under the Terms of Reference for WG 31.

List of papers

Oral presentations

Geochemical and microbial community response to oil spill: A five year investigation after the Xingang oil pipeline explosion, the Dalian Bay, North China (Invited)

Yongge Sun, Kai Zhang, Bingfang Shen, Xing Liu, Ziwei Yao and Zhenmei Lu

Marine pollution preparedness and response to oil and HNS spill incidents in the Northwest Pacific Action Plan (NOWPAP) region (Invited)

Seong Gil Kang

The *MV Marathassa*: Lessons learned from the 2015 spill of bunker fuel in Vancouver Harbour

Peter S. Ross, Carmen Morales and Mark Yunker

Distribution and sources of hydrocarbons in surface sediments from tail reaches of the Yellow River Estuary

Chuanyuan Wang and Shijie Ho

Genotoxic effects of PCBs and heavy metals on marine mussels

Adedayo O. Adeleye, Yanan Di, Yi Fie Zhang, Ying Ye and Jian Fang Chen

Distinguishing crude oils from heavy fuel oils by polycyclic aromatic hydrocarbon fingerprints

Xing Liu, Zhen Wang, Xindong Ma, Hengzhen Xu and Ziwei Yao

Oil dispersants impact feather function in marine birds

Patrick D. O'Hara, Benjamin G. Fieldhouse and Lora A. Morandin

Lessons learned from the *Hebei Spirit* oil spill: Environmental perspectives

Un Hyuk Yim, Won Joon Shim, Jong Seong Khim, Moonkoo Kim and Jee-Hyun Jung

Oil spill trajectory prediction using the GNOME model and satellite images

Yongliang Wei, Zeyan Tang, Jianqiang Liu and Xiaofeng Li

The activities of Marine Environmental Emergency Preparedness and Response Regional Activity (MERRAC) for oil and HNS spills preparedness and response in the Northwest Pacific Action Plan (NOWPAP) region

Seong Gil Kang, Jeong Hwan Oh, Yoon Young Back, Jung Hyun Lim and Bo Sik Kang

Poster presentation

Exploring the potential of Geospatial Technology for oil spill detection in shallow coastal areas in the Arabian Gulf

Pavan Kumar, Swati Katiyar and J.S. Rawat

Complex toxic impacts of heavy metals and PAHs to marine mussels cells

Yifei Zhang, Adedayo Adeleye and Yanan Di

Report of Working Group 31 on *Emerging Topics in Marine Pollution*

The Working Group on *Emerging Topics in Marine Pollution* (WG 31) held its second annual meeting from 9:00 to 18:00 h on October 17, 2015, in Qingdao, China, under the chairmanship of Drs. Peter S. Ross (Canada) and Olga Lukyanova (Russia). Ten out of 17 WG members from five member countries participated the meeting (*WG 31 Endnote 1*).

AGENDA ITEM 2

Review of agenda and goals

After review by members, the draft agenda was adopted without revision (*WG 31 Endnote 2*). The main meeting objective was to review data needs and report format for the 3rd PICES North Pacific Ecosystem Status Report.

AGENDA ITEM 3

Review of WG 31 history, activities and deliverables

Background

The Working Group on *Emerging Topics in Marine Pollution* (WG 31) reports through the MEQ Committee. WG 31 was established in 2014 following the recommendations of the Study Group on *Marine Pollutants* (October 2011–December 2013) at PICES-2013 (Nanaimo, Canada). The WG first assembled at PICES-2014 in Yeosu, Korea. Under the approved Terms of Reference, WG 31 has a 3-year mandate (2014–2017). The WG will focus on convening a series of timely topic sessions and workshops, and the organization of Special Issues in international scientific journals. In addition, WG 31 will ensure the continued availability of expertise on marine pollutants within PICES, collaborate with other PICES expert groups and international partners (GESAMP, NOWPAP), and deliver guidance consistent with the expectations of the FUTURE thematic program within PICES. WG 31 will address the question identified in the FUTURE Science Plan “How do human activities affect coastal ecosystems and how are societies affected by changes in these ecosystems?”

In light of the goals of PICES and its member countries, WG 31 recognizes that:

- Pollution can adversely affect the health and abundance of marine biota, especially in densely-populated coastal areas;
- There are socio-economic consequences of coastal pollution, with consumption advisories, fishery closures, trade interdictions and diminished aboriginal access to food resources;
- Regulations, policies and other management actions resulting from marine pollution research in the past have led to declines in the concentrations of a number of harmful pollutants, improving the health of marine biota.

Past WG 31 activities and deliverables

WG 31 had its first meeting at PICES-2014 in Yeosu, Korea, providing an opportunity for members to meet. The first annual business meeting was for WG 31 to confirm the roadmap for its 3-year mandate. During this time, the proposed data sets in support of time series were discussed in light of the planned

North Pacific Ecosystem Status Report, and an initial plan was designed to carry out this task. A successful 1-day MEQ Topic Session (S8) on “*Marine debris in the Ocean: Sources, transport, fate and effects of macro- and micro-plastics*” (co-sponsored by GESAMP, ICES and NOWPAP) was held at the Annual Meeting. A Special Issue on “*Microplastics in the ocean*” (Guest Editors: Won Joon Shim (Korea) and Richard Thompson (UK)) in the international journal *Archives of Environmental Contamination and Toxicology* (2015, 69(3): 265–373) resulted from this Topic Session.

A proposal for a Topic Session on “*Source, transport and fate of hydrocarbons in the marine environment*” was supported by WG 31 for PICES-2016 (WG 31 Endnote 3).

AGENDA ITEMS 4, 5 and 6

NPESR plans, roundtable presentations, and data collation

WG 31 reviewed the data needs and report format for the 3rd North Pacific Ecosystem Status Report (NPESR3). After consultations with SG-NPESR, WG 31 members produced a matrix for priority contaminants for which time series are likely to be available in some form from each PICES member country.

Applied discussions took place at both the WG 31 business meeting and at the MEQ Topic Session (S4) on “*Indicators of emerging pollution issues in the North Pacific Ocean*” (co-sponsored by NOWPAP), during which issues related to ocean pollution research and monitoring in the North Pacific Ocean were debated. While participants agreed that ‘pollution’ represents a serious threat to ocean resources, notably in coastal regions, they also acknowledged that the experts assembled through PICES would be unable to adequately speak to all pollution issues. In this context, members felt it important to raise this issue for PICES, and emphasize that WG 31, during its 3-year term, is specifically dealing with the topic of ‘chemical pollution’, and would not be addressing ‘biological’, ‘noise’ or ‘light’ pollution, all of which represent additional threats to the health of sea life. PICES may wish to consider addressing these topics through additional activities or within another expert group.

S4 featured talks on:

- Microplastics as a monitoring tool for contaminants,
- Spatial variation in metal concentrations in Northwest Pacific mussels,
- Level and trends of persistent pollutants in small cetaceans from Japan,
- Levels of legacy and emerging contaminants from wildlife in Korea,
- Microplastics in coastal environments in the Russian Far East,
- Microplastic risk to Cassin’s auklets in coastal British Columbia,
- Spatial and temporal trends in mercury in seabird eggs from Pacific Canada,
- Mercury and methyl mercury cycling in coastal China,
- Persistent pollutants in the marine food web in the Northwest Pacific Ocean,
- Bioindicators of marine pollution in the Sea of Okhotsk, and
- Emerging pollution issues as identified in different matrices in the Northeast Pacific Ocean.

A Special Issue in a peer-reviewed journal was proposed for the results of Topic Session S4. Dr. Hideshige Takada agreed to serve as one co-Guest Editor (the second remains to be secured). The proposal was reviewed by WG 31 and has been submitted to *Archives of Environmental Contamination and Toxicology*. A proposed deadline for manuscript submissions is May 1, 2016.

The Workshop (W4) on “*Marine environment emergencies: Detection, monitoring, response, and impacts*” (co-sponsored by NOWPAP) featured 10 oral presentations, including two invited speakers, as well as several posters. NOWPAP was well represented, and provided very practical overviews of progress made among and within its member countries (China, Korea, Russia and Japan) on spill response and monitoring. An applied roundtable discussion was held after the presentations to review several ‘hot topics’, data needs, and environmental assessment and monitoring questions.

A Special Issue in a peer-reviewed journal was proposed for the results of the W4. Dr. Un Hyuk Yim and Dr. Jeffrey Short (NOAA) agreed to serve as co-Guest Editors. The proposal was reviewed by WG 31 and has been submitted to *Archives of Environmental Contamination and Toxicology*. A proposed deadline for manuscript submissions is March 31, 2016.

Pollutants of concern within the PICES realm of the North Pacific Ocean were discussed, and a priority list was proposed for consideration for inclusion in the NPESR time series submissions. These pollutants included six general categories, followed by specific constituents for which data are most likely to be available from all six member countries and for comparative purposes. These included:

- Metals (Mercury (Hg), Lead (Pb));
- Hydrocarbons (Polycyclic Aromatic Hydrocarbons (PAHs), Benzo-a-Pyrene (BaP));
- Persistent, Bioaccumulative and Toxic substances (PBTs; PCBs, PBDEs, PFOS);
- Plastics (macrodebris shoreline surveys; microplastics);
- Pharmaceuticals (triclosan);
- Antifoulants (organotins, Irgarol).

Members agreed to identify data (published, unpublished) in support of NPESR reports, and to provide at least one report for each general category of contaminants (above) from each member country. These reports will be used in the NPESR effort, and ‘pollutants’ will be included for the first time in this important North Pacific compendium.

Contaminant data are available for multiple matrices, using different study designs and different analytical methods. In order to guide report drafting in support of the NPESR, members agreed that the provision of time series from a suitable ocean ‘indicator’ would be most informative. Deadline for the first draft report from each member country is January 15, 2016. This single report (× 6 member countries) will be circulated, shared, and reviewed to ensure that all authors agree on format, content and style after which each member country will be expected to produce reports on as many of the identified contaminants as possible. It is hoped that each country will produce at least one report for each of the six categories of pollutants (*e.g.*, metals), but members are encouraged to produce more, drawing from the proposed list of specific pollutants (*e.g.*, Hg). The deadline for these reports is May 15, 2016.

A working definition was shared to ensure that different individual draft reports ended up being comparable:

Ocean pollution indicator: “A regular sampling of a species or matrix that provides insight into the state of contamination of the marine environment”.

Factors to consider when selecting, tabulating, comparing and reporting on a selected contaminant or contaminant class included:

- The indicator could be abiotic (air, water or sediments) or biotic (shellfish, fish, birds or marine mammals);
- The matrix selected should be well understood so as to ensure good ‘real world’ understanding of habitat;
- Confounding factors should be considered so as to maximize comparability (*e.g.*, stable isotopes for trophic position, lipid for condition, organic carbon in sediments);
- Availability of time series and/or spatial trends;
- QA/QC issues in the laboratories (detection limits, certified reference materials, inter-lab comparisons);
- Links to regulations, source control, species of concern, habitat, and human health.

AGENDA ITEM 7

Relations with other groups/organizations

The relationship between WG 31 activities and other groups within and outside of PICES was discussed. Of note is the ongoing support of GESAMP and NOWPAP, with these two organizations providing expert participation or travel support for speakers.

AGENDA ITEM 8

Next steps for WG 31 in 2016

WG 31 expects to publish two Special Issues in a scientific journal (one on ‘Indicators of ocean pollution’ and one on ‘oil spills and environmental emergencies’) reflecting the MEQ Topic Session S4 on “*Indicators of emerging pollution issues in the North Pacific Ocean*” and Workshop W4 on “*Marine environment emergencies: Detection, monitoring, response, and impacts*”, respectively, held PICES-2015. Another Special Issue is anticipated to come from the Topic Session on “*Hydrocarbon source, transport and fate in the marine environment*” at PICES-2016, and a series of NPES reports from WG 31 efforts.

The meeting adjourned at 18:00 h.

WG 31 Endnote 1**WG 31 participation list**Members

John Elliott (Canada)
 Dong-Woon Hwang (Korea)
 Olga Lukyanova (Russia, Co-Chair)
 Hideaki Maki (Japan)
 Kazuhiko Mochida (Japan)
 Hyo-Bang-Moon (Korea)
 Guangshui Na (China)
 Peter S. Ross (Canada, Co-Chair)
 Hideshige Takada (Japan)
 Un Hyuk Yim (Korea)

Observers

Karin Baba (Japan)
 Seong-Gil Kang (NOWPAP MERRAC)
 Peter John Kershaw (GESAMP)
 Cathryn Clarke Murray (PICES)
 Vasiliy Yu. Tsygankov (Russia)
 Xiaodong Zhong (NOWPAP RCU)

WG 31 Endnote 2**WG 31 meeting agenda**

1. Introductions
2. Review of agenda and goals for the day
3. Review of WG 31 history, activities and deliverables
 - 2014 (Korea): Microplastics (Topic Session and Special Issue in *Archives of Environmental Contamination and Toxicology*)
 - 2015 (China): Indicators (Topic Session and proposed Special Issue in a journal) and workshop on “*Marine environmental emergencies*”
 - Discussion on proposal for Topic Session at PICES-2016 (San Diego, USA; co-sponsored by GESAMP) and Special Issue in a journal: Hydrocarbon transport, fate and effects in the marine environment
4. WG 31 and FUTURE opportunities and tasks; PICES Status report plans for WG 31:
 - Persistent, bioaccumulative substances;
 - Metals or elements of concern;
 - Marine debris and microplastics.
5. Roundtable presentations on ‘What is an Indicator of ocean pollution?’
6. Breakout groups to discuss data collation in support of NPESR and four selected contaminant categories:
 - Review papers and original research;
 - Assessment leads and teams;
 - Finalise framework/conceptual approach for each category;
 - Deadline for report submission;
 - Draft manuscripts for journal submission?
7. Relationship of WG 31 to PICES groups and international organizations (*e.g.*, NOWPAP, GESAMP)
8. 2016 as the final year for WG 31: roundtable on next steps

WG 31 Endnote 3**Proposal for a 1-day MEQ Topic Session on “*Source, transport and fate of hydrocarbons in the marine environment*” at PICES-2016**

Co-sponsor: GESAMP

Co-Convenors: Hideaki Maki (Japan), Staci Simonich (USA), Geraldo Gold-Bouchot (GESAMP) Peter S. Ross (Canada)

Suggested Invited Speakers: Ken Lee (Australia Oceans Program, CSIRO, Australia), John Stein (NOAA, USA)

This session will focus on the behavior, fate and effects of hydrocarbons in the marine environment. While it is expected that some examples of oil spills (catastrophic release of hydrocarbons) will be examined, most discussions will focus on chronic, low level releases from multiple sources that are far more evasive and widespread (*e.g.*, ballast discharges, fuel release, harbor contamination). Following two successful sets of activities at PICES-2014 and PICES-2015 (‘Microplastics’ and ‘Indicators of ocean pollution’), the WG 31 (Emerging Topics Marine Pollution; ETMP) proposes to organize,

convene and facilitate the third in its planned series of Special Sessions. The topic for 2016 is to comprehensively address the science of ‘Source, transport and fate of hydrocarbons in the marine environment’. This is timely for PICES as it follows up on the 2015 workshop on short-term response workshop (“*Marine Environment Emergencies: Detection, monitoring and response*”). This topic is also timely since oil and gas exploration, development and transport is taking place to varying degrees around the North Pacific Ocean. Thousands of different hydrocarbon compounds are found in fuels, each with different physical and chemical properties. The resulting complex interactions between these compounds and components of the marine environment highlight the importance of a multidisciplinary and up-to-date sharing of knowledge. This knowledge will provide insight into the consequent risks to biota, the design of monitoring programs, the choice of analytical methods, and management responses following leaks or spills. This Topic Session will feature invited speakers from several national organizations. A Special Issue in a scientific journal will arise from the presentations on “*Source, transport and fate of hydrocarbons in the North Pacific Ocean*”. Presenters and others will be invited to submit a manuscript on the topic, with the goal of the resulting compendium being to become a useful reference work for scientists and managers.

PICES-2016, San Diego, USA

November 2–13, 2016

Excerpted from:

Summary of Scientific Sessions and Workshops at PICES-2016

MEQ Topic Session (S3)

Source, Transport and Fate of Hydrocarbons in the Marine Environment

Co-sponsor: GESAMP

Co-Convenors: *Hideaki Maki (Japan), Staci Simonich (USA), Robert Duce (GESAMP, Texas A&M University)*

Invited Speaker:

Kenneth Lee (Commonwealth Scientific and Industrial Research Organization (CSIRO), Australia)

Background

Following two successful sets of activities at PICES-2014 and PICES-2015 ('Microplastics' and 'Indicators of ocean pollution'), the Working Group on *Emerging Topics Marine Pollution* (WG 31) organized, convened and facilitated the third in its planned series of Special Sessions. The topic chosen for 2016 was to comprehensively address the science of 'Source, transport and fate of hydrocarbons in the marine environment'. This is timely for PICES as it followed up on the PICES-2015 short-term response workshop, "*Marine environment emergencies: Detection, monitoring and response*". This topic is also timely since oil and gas exploration, development and transport is taking place to varying degrees around the North Pacific Ocean. Thousands of different hydrocarbon compounds are found in fuels, each with different physical and chemical properties. The resulting complex interactions between these compounds and components of the marine environment highlight the importance of a multidisciplinary and up-to-date sharing of knowledge. This knowledge will provide insight into the consequent risks to biota, the design of monitoring programs, the choice of analytical methods, and management responses following leaks or spills.

Summary of presentations

The topic session, co-sponsored by GESAMP, focused on the behavior, fate and effects of hydrocarbons in the marine environment, as well as some logistic approaches for oil spills. While some examples of catastrophic oil spills were referred to, some discussions focused on chronic, low level releases of hydrocarbons from multiple sources that are far more evasive and widespread, and also some examples on physicochemical transformations of oil and/or hydrocarbons in marine environmental media.

Dr. Kenneth Lee, Commonwealth Scientific and Industrial Research Organization (CSIRO), Australia, gave us a comprehensive talk as invited speaker on various case studies of oil spills in terms of type of oil, environmental media, ecosystem response and counter measures. There is no universal and simple

method to mitigate influences of oil spill without any adverse effect on marine ecosystems. The best possible way should be chosen for each case on the basis of scientific knowledge that has been obtained from previous examples. This was followed by some experimental studies on the influences of oil-accommodated fraction in seawater on phytoplankton and the effect of sunlight irradiation of oil slicks on marine crustaceans. The Korean research team reported experimental formation and influences of oiled particulate matter aggregates suspended in seawater. The Canadian groups reported some logistic approaches toward possible oil spill. To simulate the trajectory of spilled oil in channels in British Columbia coastal seas, many drifters have been released and simple statistical analysis of their behaviors in channels shows that it could be a useful tool to predict drifts of spilled oil. A framework has been made to assess the vulnerability of biota to oil spill in coastal areas of Canada, which identifies the areas that should be prioritized in terms of prevention of oil stranding and clean-up to mitigate potential risk of oil spills on habitats. Although the scientific contents of each presentation were rather diverse, they remind us of the importance of promoting science and technology to predict what happens in marine ecosystem after both accidental and chronic oil spills and what kinds of countermeasures should be prepared for future releases.

List of papers

Oral presentations

Sources, behaviour and environmental impacts of petroleum hydrocarbons released into the marine environment (Invited)

Kenneth Lee

Physiological responses of marine phytoplankton to oil exposure in the context of the 2015 oil spill in the Santa Barbara Channel

Tanika M. Ladd, Jessica A. Bullington, Andrea Valdez-Schulz, Paul G. Matson, and M. Debora Iglesias-Rodriguez

Photo-oxidation of crude fuel and its toxicity to marine amphipods

Hideaki Maki, Takehiko Hiwatari and Kunio Kohata

In situ formation of oil-suspended particulate matter aggregate during flushing activities

Andrew Loh, Un Hyuk Yim, Sung Yong Ha and Joon Geon An

How far will it go? The estimation of oil spill extents from surface drifter data

Charles Hannah, Hauke Blanken, Tamás Juhász and Stephen Page

A framework to assess vulnerability of biological components to oil spilled in the marine environment

Lucie Hannah, Kate Thornborough, Candice St. Germain and Miriam Q

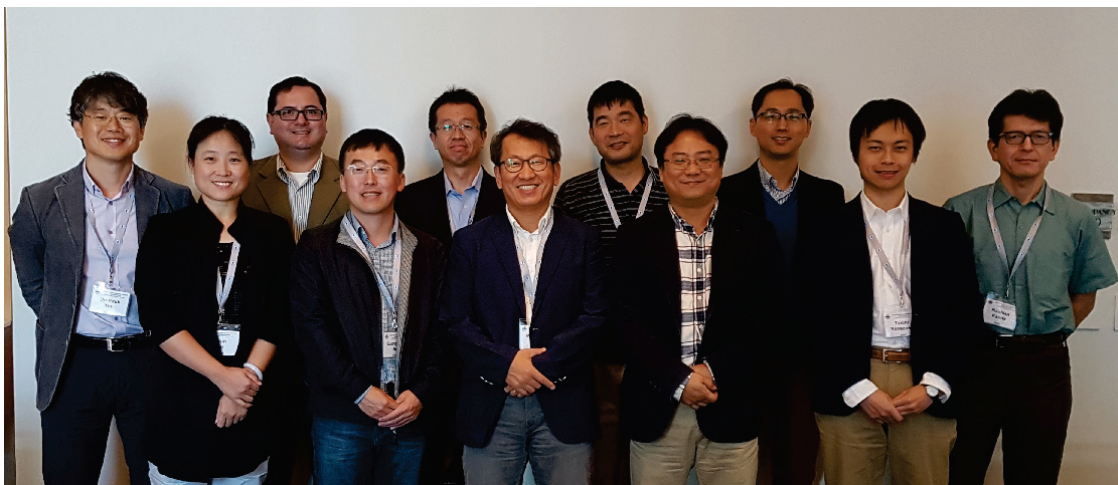
Poster presentations

Atmospheric concentration of petroleum derived polycyclic aromatic hydrocarbons after the Hebei Spirit oil spill

Joon Geon An, Un Hyuk Yim, Sung Yong Ha, Moonkoo Kim and Won Joon Shim

Report of Working Group 31 on *Emerging Topics in Marine Pollution*

WG 31 met from 9:00 to 18:00 h on November 3, 2016 in San Diego, USA, under the chairmanship of Dr. Wonjoon Shim (Korea). The meeting objective was to review activities during WG 31's third year (2016) to discuss a 1-year extension to finalize environmental time series observations (ETSO) for the third North Pacific Ecological Status Report and WG 31 final report, and to discuss the strategy and future plan post-WG 31.



WG 31 participants at PICES-2016; from left: Un Hyuk Kim, Zijun Xu, Juan José Alava, Guangshui Na, Kazuhiko Mochida, Wonjoon Shim, Hideaki Maki, Dong-Woon Hwang, Mikyu Choi, Taichi Yonezawa, Naohisa Kanda.

AGENDA ITEM 1

Welcome and adoption of agenda

Seven (out of 20 WG 31 members) representing three member countries and five observers participated in the meeting (*WG 31 Endnote 1*). The draft agenda was adopted after it was reviewed and slightly modified by the WG (*WG 31 Endnote 2*).

AGENDA ITEM 2

Review of the main achievement of WG 31 – 2016

The Co-Chair reported on the outcome of the MEQ Topic Session S3, entitled “*Source, transport and fate of hydrocarbons in the marine environment*”. Six oral presentations, including one invited speaker (Dr. Kenneth Lee, CSIRO) and two poster presentations (total 8) were accommodated in the session. The session was co-sponsored by GESAMP.

Papers were submitted from Topic Session S4 on “*Indicators of emerging pollution issues in the North Pacific Ocean*” and Workshop W4 on “*Marine environment emergencies: Detection, monitoring, response, and impacts*” (PICES-2015) to the scientific journal of *Archives of Environmental Contamination and Toxicology*. WG 31 discussed the number of papers from S4 submitted to a special

issue on “*Pollution Indicators*” (Guest Editors: T. Brown and H. Takada; $n = 15$) and W4 submitted to a special issue on “*Oil Spills*” (Guest Editors: J. Short and U.H. Yim; $n = 19$). It is expected that the two special issues will be published before PICES-2017.

AGENDA ITEM 3

Publication of special issue from the topic session at PICES-2016

Papers from PICES-2016 Topic Session S3 on “*Source, transport and fate of hydrocarbons in the marine environment*” will be submitted to a special issue of *Archives of Environmental Contamination and Toxicology*. A tentative short title is “*Hydrocarbons in Marine Environment*” and the special issue will be guest edited by Drs. S. Simonich, H. Maki and R. Duce. Publication date is anticipated to be prior to PICES-2017.

AGENDA ITEM 4

Proposals for a new topic session

A topic session entitled “*Microplastics in marine environments: Fate and effects*” was proposed by co-convenors, Drs. Wonjoon Shim, Hideshige Takada, and Peter Ross for PICES-2017 (**WG 31 Endnote 3**). The topic session proposal was supported by members. It is expected that data and results from this session will also be important as WG 31 supports the needs of FUTURE.

AGENDA ITEMS 5 and 6

NPESR-3 ETSO reports on marine pollution and schedule

Most of the business meeting was spent for this issue. Dr. Juan José Alava (Canada; proxy for Dr. Peter Ross representing WG 31 from the SG-NPESR workshop for time-series evaluation and synthesis, June 28–30, 2016, Sidney, Canada) presented the status of WG 31 ETSO reports collected and reviewed from that workshop. A total 26 ETSO reports on marine pollution were collected from Canada ($n = 7$), China ($n = 0$), Japan ($n = 5$), Korea ($n = 10$), Russia ($n = 2$) and USA ($n = 2$) (Fig. 1).

Country	Hg sediment	Hg bivalves	Cu in marine water	Pb sediments	Pb bivalves	PAHs in mussel and/or sediments	TBT, Irgarol and Diuron	PCBs in bivalves	Dioxins/Furan in sediments & bivalves	PCBs in sediment and/or marine water	POPs in marine mammals	DDT raptors/seabirds	Marine Debris	Microplastics	Total
Canada	1		1							1	1	1	1	1	7
China															0
Japan	1				1		1	1		1					5
Korea	1	1		1	1	1	1		1		2		1		10
Russia	1			1											2
USA						1							1		2
Total	4	1		3		2	2			2	3	1	3	1	26

Figure 1 List of ETSO reports on marine pollution submitted for the third version of the North Pacific Ecosystem Status Report (NPESR3) by WG 31.

China prepared reports but has not received approval from the State Oceanic Administration to release

the data. WG 31 will report this issue to MEQ. The deadline for ETSO submissions on marine pollution to NPESR3 has been extended to January 2017 to allow for the collection of more reports from PICES member countries.

AGENDA ITEM 7

Extension of WG 31

All WG members agreed that WG 31 needed to be extended for another year past its 3-year term in order to finalize special issues, ETSO reports on marine pollution, and the WG 31 final report. This request was included in the MEQ agenda to take to Science Board (see *WG 31 Endnote 4*).

AGENDA ITEM 8

Preparation and publication of WG 31 final report

The content of the final report was discussed by WG 31. Main content of the WG 31 final report will be ETSO reports on marine pollution. The publication schedule is set as follows:

- Finalize contents (30 November, 2016),
- Designate contributors (15 December, 2016),
- Collection of draft (30 April, 2017),
- Review the draft (15 May, 2017),
- Collection of revised report (30 June, 2017),
- Preparation of Executive Summary (30 July, 2017),
- Finish and submit the report to MEQ (30 August 2017).

AGENDA ITEM 9

Post-WG 31 plans

Members discussed what future activities might take place after 2017, pending approval of the WG's extension. The following ideas were proposed.

New Study Group for specific topics on

- Marine plastic debris, including microplastics,
- Pharmaceuticals and personal care products (PPCP; *e.g.* antibiotics, sunscreen),
- Impact of contaminants on the ecosystem,
- Interaction of climate change and bioaccumulation/toxic effects,
- Biological pollution (*e.g.* pathogens),
- Impact of offshore mining.

New Section for broader topics on

- Chemical pollution
- Marine Environmental Emergency (*e.g.* oil and HNS spills)

Dr. Shim instructed that the WG will correspond by email to decide on what type of expert group and future directions it will take (2018~) prior to the next PICES Annual Meeting so a proposal can be submitted to MEQ.

AGENDA ITEM 10

Preparation of WG 31 report for MEQ

Dr. Shim will prepare a brief report and requests to bring to the MEQ meeting on November 9.

AGENDA ITEM 11

Relations with other groups/organizations

No discussion.

AGENDA ITEM 12

Other business

None

WG 31 Endnote 1**WG 31 participation list**Members

Dong-Woon Hwang (Korea)
 Hideaki Maki (Japan)
 Kazuhiko Mochida (Japan)
 Guangshui Na (China)
 Wonjoon Shim (Korea, Co-Chair)
 Zijun Xu (China)
 Un Hyuk Yim (Korea)

Members unable to attend

Canada: John Elliott, Peter S. Ross (Co-Chair)
 China: Zhengguo Cui, Haiyan Lv, Qun Wang
 Japan: Hideshige Takada
 Korea: Hyo-Bang Moon
 Russia: Olga N. Lukyanova (Co-Chair), Vladimir M. Shulkin Mikhail Simokon
 USA: Staci Simonich, Nancy Wallace, Gina M. Ylitalo

Observers

Juan José Alava (Canada)
 Chuanlin Huo (China, MEQ Chair)
 Naohisa Kanda (Japan)
 Taichi Yonezawa (Japan)
 Mikyu Choi (Korea, MEQ)

WG 31 Endnote 2**WG 31 meeting agenda**

Thursday, November 3, 2016

1. Welcome and adoption of agenda (Co-Chairs)
Welcome members and introduce the agenda about the WG 31 meeting
2. Review of the main achievement of WG 31 – 2016 (Co-chairs)
Summarize the main achievements of WG 31 in the past 1 year
3. Discussion about a special issue in ‘Hydrocarbon’ (All)
4. Proposals for new topic sessions (Co-conveners)
Introduction of the proposed topic session and workshop for 2017 and discussion
5. Presentation of status and trend (Ecosystem Time Series Observation; ETSO) reports for the designated contaminants (Dr. Juan José Alava)
6. Discussion about future schedule of ETSO reports (All)
7. Discussion about 1 year extension of WG 31 (All)
8. Discussion about WG 31 final report (All)
9. Post-WG 31 plans
10. Preparation of WG 31 report for MEQ meeting (All)
11. Relations with other groups/organizations (All)
Introduce the progress on their work with other groups or other organizations.
12. Other business (All)

WG 31 Endnote 3

**Proposal for a 1-day MEQ Topic Session on
“Microplastics in Marine Environments: Fate and Effects” at PICES-2017**

Convenors: Wonjoon Shim (Korea), Hideshige Takada (Japan), Peter Ross (Canada), TBA (NOWPAP), TBA (GESAMP)

Co-sponsor: NOWPAP, GESAMP

Microplastics are now ubiquitous from the near shore to open ocean, from the sea surface to bottom, and from subtropical to polar seas. Relatively high abundance of microplastics has been reported in the North Pacific Gyre as well as coastal waters of North Pacific region among the world oceans. In addition, with decreasing size, they become more bioavailable to small aquatic organisms down to zooplankton. Ingested microplastics have been found in various taxa across trophic levels. Associated chemicals in microplastics may be transferred to an organism upon ingestion. Microplastics represent trans-boundary pollution which can also deliver associated chemicals and invasive organisms to regions far removed from source. Microplastics are increasingly recognized as a potential threat to biota in the ocean. However, because of their size detecting the presence of microplastics and adverse biological effects, if any, becomes considerably more challenging. The objective of this session is to present status and trend information for microplastic pollution and its environmental consequences in the PICES region. Papers are invited that assess microplastics 1) hotspots in the PICES region, 2) sources and input pathways, 3) fate and behaviour of microplastics, 4) role as sink or source of associated toxic chemicals, and 5) biological and ecological effects. Recommendations on how to address growing problems associated with microplastics will be also considered.

WG 31 Endnote 4**Proposal for WG 31 extension of 1 year****1. Achievements to date****1.1 Publication of peer-reviewed journal special issues**

- ‘Microplastics in the Ocean’ in *Archives of Environmental Contamination and Toxicology* (2015), Vol. 69 with eleven papers, including a review

1.2 Organizing topic sessions and workshop at PICES Annual Meetings

- A topic session on “*Marine debris in the ocean: Sources, transport, fate and effects of macro- and micro-plastics*” (co-sponsored by ICES, NOWPAP, and GESAMP), PICES-2014, Yeosu, Korea;
- A topic session on “*Indicators of emerging marine pollution issues in the North Pacific Ocean*” (co-sponsored by NOWPAP), PICES-2015, Qingdao, China;
- A topic session on “*Source, transport and fate of hydrocarbons in the marine environment*” (co-sponsored by GESAMP), PICES-2016, San Diego, USA;
- A workshop on “*Marine environmental emergencies: Detection, monitoring and response*” (co-sponsored by NOWPAP), PICES-2015, Qingdao, China.

1.3 Pollution status and trend reports for ETSO in NPESR3

- A total 26 pollution status and trend reports were collected for metals (Hg and Pb), hydrocarbons (total PAHs and B(a)P), PBT substances (PCBs, Dioxin/Frs, PBDEs, and PFOS), plastic debris (Macro-debris and microplastics), pharmaceuticals (triclosan), and anti-foulants (organotins, Irgarol and Diuron).

2. Rationale for extension

- To publish three remaining peer-reviewed special journal issues on ‘Oil Spills’, ‘Pollution Indicators’ and ‘Hydrocarbons in the Marine Environment’;
- To complete pollution status and trend reports for NPESR3;
- To complete a final report of WG 31.

3. New timeline

- February, 2017: Completion of pollution status and trend report for NPESR3;
- March, 2017: Publication of a peer-reviewed special journal issue for ‘Oil Spills’ in *Archives of Environmental Contamination and Toxicology* (twenty manuscripts were submitted);
- May, 2017: Publication of a peer-reviewed special journal issue for ‘Pollution Indicators’ in *Archives of Environmental Contamination and Toxicology* (ten manuscripts were submitted and five were in preparation);
- August, 2017: Submission of WG 31 final report (see *Appendix*);
- September, 2017: Publication of a peer-reviewed special journal issue for ‘Hydrocarbons in the Marine Environment’ in *Archives of Environmental Contamination and Toxicology*.

Appendix

Contents of WG 31 Report (tentative)

Executive Summary

1. Introduction
 2. Contaminants of concern (CoC) in North Pacific region
 3. Status and Trend of CoC in North Pacific region
 - 3.1. Canada
 - 3.1.2 Hydrocarbons
 - 3.1.3 Metals
 - 3.1.4 PBT substances
 - 3.1.5 Plastic debris
 - 3.1.6 Pharmaceuticals
 - 3.1.7 Anti-foulants
 - 3.2. China
 - 3.2.1 Hydrocarbons
 - 3.2.2
 - 3.3. Japan
 - 3.4. Korea
 - 3.5. Russia
 - 3.6. USA
 4. Data gaps
 5. Summary and Recommendations
- Appendix 1: Terms of Reference
- Appendix 2: WG31 membership
- Appendix 3: Annual Reports and Topic Session/Workshop Summaries

PICES-2017, Vladivostok, Russia

September 22–October 1, 2017

Excerpted from:

Summary of Scientific Sessions and Workshops at PICES-2017

MEQ Topic Session (S2)

Microplastics in marine environments: Fate and effects

Co-Sponsors: GESAMP, NOWPAP

Co-Convenors: *Wonjoon Shim (Korea) Hideshige Takada (Japan) Peter Ross (Canada) Peter Kershaw (GESAMP) Lev Neretin (NOWPAP)*

Invited Speakers:

Seung-Kyu Kim (Incheon National University, Korea)

Daoji Li (East China Normal University, China)

Chelsea M. Rochman (University of Toronto, Canada)

Background

Microplastics are now ubiquitous from the near shore to open ocean, from the sea surface to bottom, and from subtropical to polar seas. Relatively high abundance of microplastics has been reported in the North Pacific Gyre as well as coastal waters of North Pacific region among the world oceans. In addition, with decreasing size, they become more bioavailable to small aquatic organisms down to zooplankton. Ingested microplastics have been found in various taxa across trophic levels. Associated chemicals in microplastics may be transferred to an organism upon ingestion. Microplastics represent trans-boundary pollution which can also deliver associated chemicals and invasive organisms to regions far removed from source. Microplastics are increasingly recognized as a potential threat to biota in the ocean. However, because of their size detecting the presence of microplastics and adverse biological effects, if any, becomes considerably more challenging. The objective of this session was to present status and trend information for microplastic pollution and its environmental consequences in the PICES region. Papers are invited that assess microplastics 1) hotspots in the PICES region, 2) sources and input pathways, 3) fate and behaviour of microplastics, 4) role as sink or source of associated toxic chemicals, and 5) biological and ecological effects. Recommendations on how to address growing problems associated with microplastics will be also considered.

Summary of presentations

The session accommodated one plenary, two invited, twelve oral and seven poster presentations, including seven from early career scientists. About 50 marine scientists and geochemists, marine biologists, bird and mammal researchers, physical oceanographers, researchers from NGOs, policy makers and students attended the session.

A plenary speaker, Dr. Chelsea Rochman, provided a comprehensive overview regarding the issue of microplastics pollution and discussed its ecological and human health impacts. Of the two invited talks, Dr. Daoji Li summarized a microplastic national research project and pollution status in marine and freshwater environments of P.R. China. The other Invited Speaker, Dr. Seung-Kyu Kim, presented a microplastics monitoring study in the Arctic environment at sites along the North Pacific passage. Dr. Won Joon Shim presented a global overview based on the available literature on microplastics abundance, composition, and size distribution across the world, emphasizing especially high concentrations of microplastics in Asia and the North Pacific region, making them global 'hot spots'. Dr. Peter Kershaw from the Joint Group of Experts on the Scientific Aspects of Marine Environmental Protection (GESAMP) introduced the third phase of the microplastics Working Group 40 activity on harmonization of microplastic sampling and analysis. A vertical distribution of >20 µm microplastics from surface to bottom water which has not been addressed yet was presented by Dr. Young Kyoung Song. Microplastics pollution in two estuarine and coastal environments of several rivers in the Far Eastern part of Russia was presented by Dr. Nikolai Kozlovskii. The first data on higher microplastics abundance in the estuary of the transboundary Tumen River (P.R. China, DPRK and Russia) was especially interesting. Ms. Katerina Vassilenko presented results on the abundance of microfibers in seawater and zooplankton along the eastern Pacific coast of Canada. Trophic transfer of microplastics in zooplankton by predation of salmon and whale was estimated in the Strait of Georgia, Canada. Mr. Seongbong Seo presented modelling results on transport of microplastics with river discharge from five major rivers in the R. Korea. The study assessed microplastics transport near shore and into the marginal seas surrounding the R. Korea and the individual microplastics contribution of rivers to the marine environment were estimated. Mr. Garth Covernton gave a systematic study on microplastic residues in wild and cultured clams in various habitats. Ms. You Na Cho presented a market basket survey of microplastic contamination in four bivalve species, including an estimate of the annual uptake of microplastics through consumption of the contaminated bivalve species in the R. Korea. Dr. Patrick O'Hara showed the difference between microplastic and plastic contents in seabirds; concentrations of microplastics in Cassin's auklets' stomachs varied according to breeding and non-breeding seasons. Dr. Youn-Joo An provided an overview of the adverse biological effects of microplastics on freshwater and marine organisms, including recent data on micro- and nanoplastic ingestion by freshwater organisms based on a trophic transfer study in a laboratory. Dr. June-Woo Park presented results of the study on the accumulation and toxic effects of microfibers and the associated chemicals such as phthalates on behavior and biomarker response of sheepshead minnow. Finally, Dr. Lev Neretin from NOWPAP provided an overview of the evolution of the global marine litter policies and summarised results of international and regional activities addressing marine plastic pollution issues.

List of papers

Oral presentations

Contamination and effects of plastic debris in the marine environment (Plenary)

Chelsea M. Rochman

Microplastic in the marine environments, China (Invited)

Daoji Li, Shiye Zhao, Juying Wang, Huahong Shi and Guyu Peng

Microplastics in Arctic surface water and sea ice: 2016 ARAON Expedition (Invited)

Hee-Jee Lee and Seung-Kyu Kim

Global distribution of microplastics: An overview

Won Joon Shim, Sang Hee Hong, Soeun Eo

Monitoring and assessment of marine plastics and microplastics – Towards harmonised methodsPeter J. [Kershaw](#), Francois Galgani and Alexander Turra**Vertical distribution and composition of microplastics in Korean coastal waters**Young Kyoung [Song](#), Soeun Eo, Gi Myung Han, Sang Hee Hong, Won Joon Shim**River discharge as a source of plastic litter pollution in the Northwest Pacific Russia**Nikolai [Kozlovskii](#), Anatolii Kachur**Microfiber source characterization in the Northeastern Pacific Ocean**Katerina [Vassilenko](#), Mathew Watkins, Anahita Etamadifar, Marie Noel, Peter S. Ross**Fate of floating debris released from Korean rivers**Seongbong [Seo](#) and Young-Gyu Park**Microplastic concentrations in wild and cultured clams and their environment in British Columbia, Canada**Garth A. [Covernton](#), Sarah E. Dudas, Christopher M. Pearce, Helen J. Gurney-Smith and John F. Dower**Assessment of microplastic contamination in commercial bivalves from South Korea**You Na [Cho](#), Mi Jang, Gi Myung Han, Won Joon Shim and Sang Hee Hong**Seasonal variability in vulnerability for Cassin's Auklets exposed to plastic pollution in the Canadian Pacific region**Patrick D. [O'Hara](#), Stephanie Avery-Gomm, Jocelyn Wood, Laurie Wilson, Victoria Bowes, Jean-Pierre Desforges, Peter Ross, Sean Boyd, Ken Morgan**Ecological effect of micro-sized plastics: Research trends and research needs**Youn-Joo [An](#) and Yooeun Chae**Assessment of microplastic fibers impacts from the chronic exposure to juvenile sheepshead minnow (*Cyprinodon variegatus*)**Jin Soo Choi, Youn-Joo Jung, Yunwi Heo and June-Woo [Park](#)**Applying precautionary principle to microplastics governance framework: Solutions in the absence of “complete” scientific evidence**Lev [Neretin](#)*Poster presentations***Ecotoxicity effects of microplastic to the early life stages of large yellow croaker**Fangzhu Wu, Xiaoqun Liu, Jiangning Zeng, Qiang Liu, Wei [Huang](#)**Abundance, composition and distribution of microplastics on Korean Beaches**Soeun Eo, Young Kyung [Song](#), Sang Hee Hong, Gi Myung Han, Won Joon Shim**Changes of carbonyl and vinyl index of three plastics by outdoor exposure**Young Kyoung [Song](#), Soeun Eo, Sang Hee Hong, Won Joon Shim**Development of efficient analytical method for microplastics in bivalves**Mi Jang, You Na [Cho](#), Young Kyung Song, Won Joon Shim and Sang Hee Hong**Bioaccumulation of microplastics in sheepshead minnow (*Cyprinodon variegatus*)**Jin Soo Choi, Youn-Joo Jung, Hong gil [Yun](#) and June-Woo Park**Spatial characteristics of microplastics in the surface waters along the coast of Korea**Jung Hoon [Kang](#), Oh-Youn Kwon, Minju Kim, Sang Hee Hong and Won Joon Shim**Microplastics in freshwater river sediments in Shanghai, China: Case study of environmental risk assessment in mega cities**Guyu [Peng](#), Pei Xu, Bangshang Zhu, Daoji L