Spatial distributions of atmospheric water soluble nitrogen and phosphorus depositions to the Pacific Ocean

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1. Introduction

Atmospheric transport of anthropogenic and natural matter from continent is one of the important sources for marine biogeochemical nutrient cycles in the ocean. The atmospheric aerosols of combined nitrogen, ammonium and nitrate are transported from continental areas to the North Pacific Ocean. Duce et al. (2008) estimated from the modeling study that atmospheric deposition of total reactive nitrogen, and estimated that increased ～20 Tg N year^{-1} （～29 % was anthropogenic) in 1860 to ～67 Tg N year^{-1} （～80 % was anthropogenic) in 2000. And Jickells et al. (2017) predicted that increasing atmospheric deposition of nitrogen over the western North Pacific, even if the total net nitrogen input to the oceans may not increase, as a result, that increasing ocean primary productivity regionally. In order to understand spatial distributions of atmospheric nitrogen (N) and phosphorus (P) depositions to the Pacific Ocean were examined using aerosol (dry) and rainwater (wet) samples collected on R/V Hakuho-maru during five cruises from July 2012 to February 2015.

2. Cruises

- KH-12-3: 2012/7/6～8/14
- KH-12-4: 2012/8/23～10/3
- KH-13-7: 2013/12/11～2014/2/12
- KH-14-3: 2014/6/23～8/11
- KH-14-6: 2014/12/1～2015/2/26

R/V Hakuho-maru

Ship tracks
— : KH-12-3
— : KH-12-4
— : KH-13-7
— : KH-14-3
— : KH-14-6
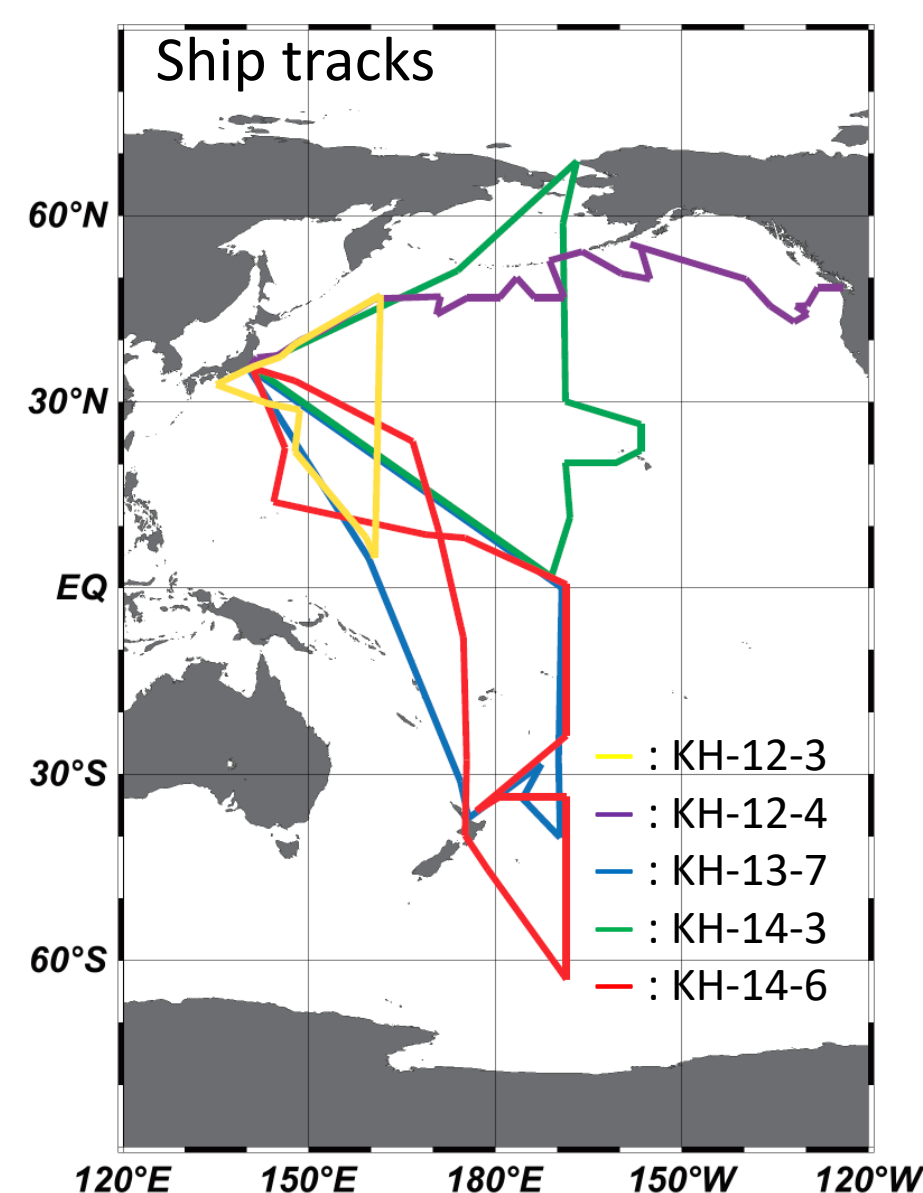
## 3. Sampling methods

【Atmospheric aerosols】
- High-volume virtual-impactor air sampler (Kimoto Electric Co., Ltd., AS-9) .
- Virtual impactor： Atmospheric aerosols were segregated into fine (<PM 2.5) and coarse (>PM 2.5) modes on the same filter.
- Teflon filter （ADVANTEC, PF040, 90mmφ）.
- Quartz fiber filter papers (Tokyo Dylec, 2500AT-UP, 90mmφ): Pre-combusted (4 h at 850 $^{\circ}$C, Nakamura et al., 2006) and used KH-12-3 only.
- Sampling intervals:
  ◆ KH-12-3, KH-13-7, KH-14-3 (Leg1), KH-14-6: 3 days
  ◆ KH-12-4, KH-14-3 (Leg2): 1 day.
- Wind-sector controller: Avoid contamination from ship's exhaust.
- Storage at 4 $^{\circ}$C.
【Rainwater】
- Polyethylene funnel: 30 cm diameter.
- Collected in a 250-ml fluorinated high-density polyethylene bottle.
- Storage at −20 $^{\circ}$C.

Fine modes
Coase modes
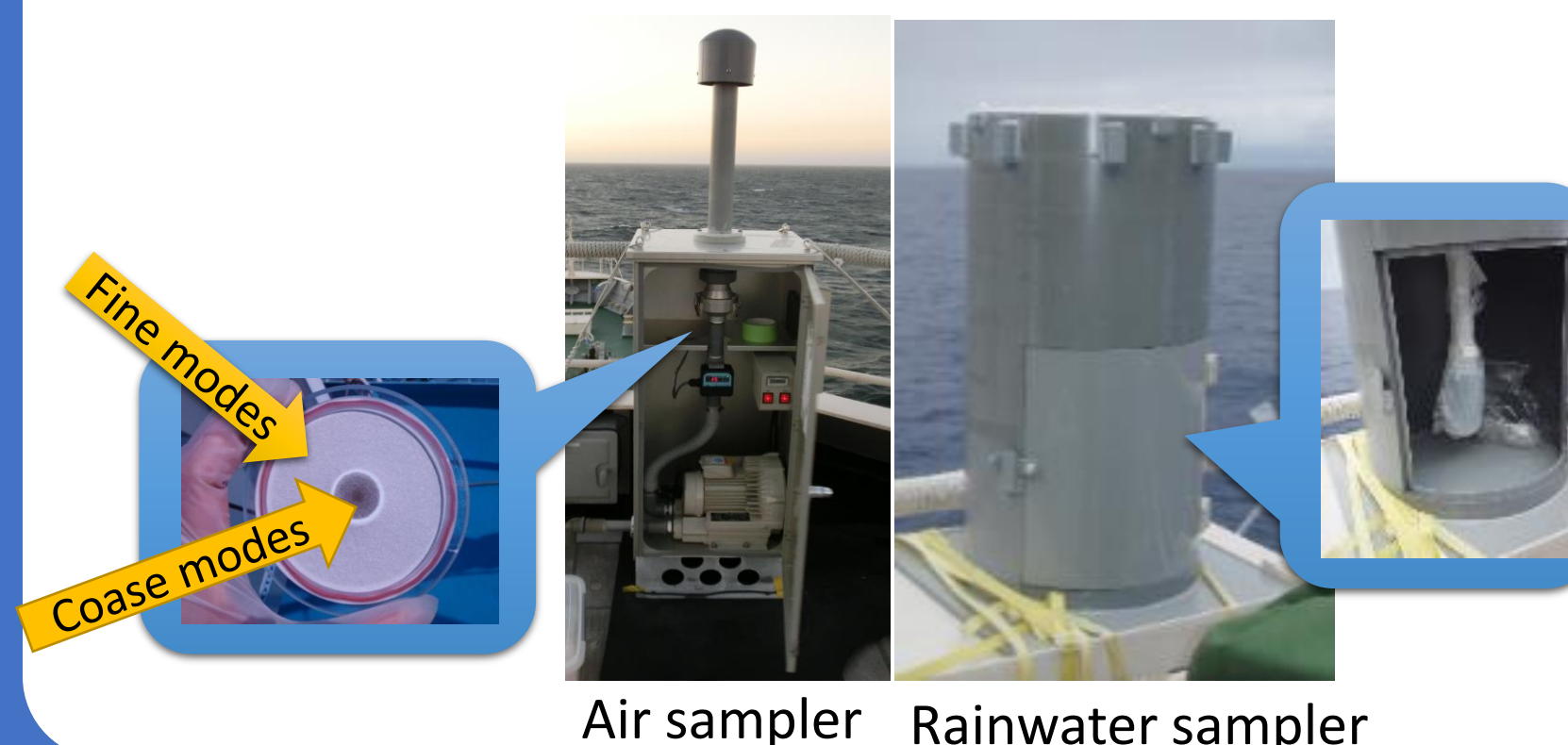
Air sampler   Rainwater sampler

4. Analytical methods

【Pretreatment】
- Atmospheric aerosols on the filter were cut into four equivalent subsamples and separated into fine and coarse mode fractions.
- Ultrasonically extracted with 25 ml of Milli-Q water for 60 min.
- The extracted solutions and rainwater samples were filtered through an acetyl-cellulose filter (0.45 μm pore size).
【Analysis】
- Auto-analyzer：NO_3^-, NH_4^+, PO_4^{3-}
- Ion chromatography：NO_3^-, NH_4^+, Na^+, Mg^{2+}, Cl^-, SO_4^{2-}, Ca^{2+}
- (DIN= NO_3^- + NH_4^+)

5. Deposition flux estimates

【Dry deposition fluxes】
  Fd = Caf x Vf + Cac x Vc
    Fd : Dry deposition fluxes
    Caf : Atmospheric aerosol concentration of fine modes
    Cac : Atmospheric aerosol concentration of coarse modes
    Vf : Dry deposition velocities of fine modes (0.1 cm s^{-1} *)
    Vc : Dry deposition velocities of coarse modes (2cm s^{-1} *)
                (*Duce et al., 1991)

【Wet deposition fluxes】
  Fr = Cr x P
    Fr : wet deposition fluxes
    Cr : rainwater concentration
    P : precipitation rate
<Precipitation rate>
- KH-12-3, KH-12-4: Tropical Rainfall Measurement Missions (TRMM, 0.25^{0}x 0.25^0 monthly average data. Huffman et al., 2007）
- KH-13-7, KH-14-3, KH-14-6: CPC Merged Analysis of Precipitation (CMAP, 0.5^{0}x 0.5^0 monthly average data)

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6. Dry deposition fluxes
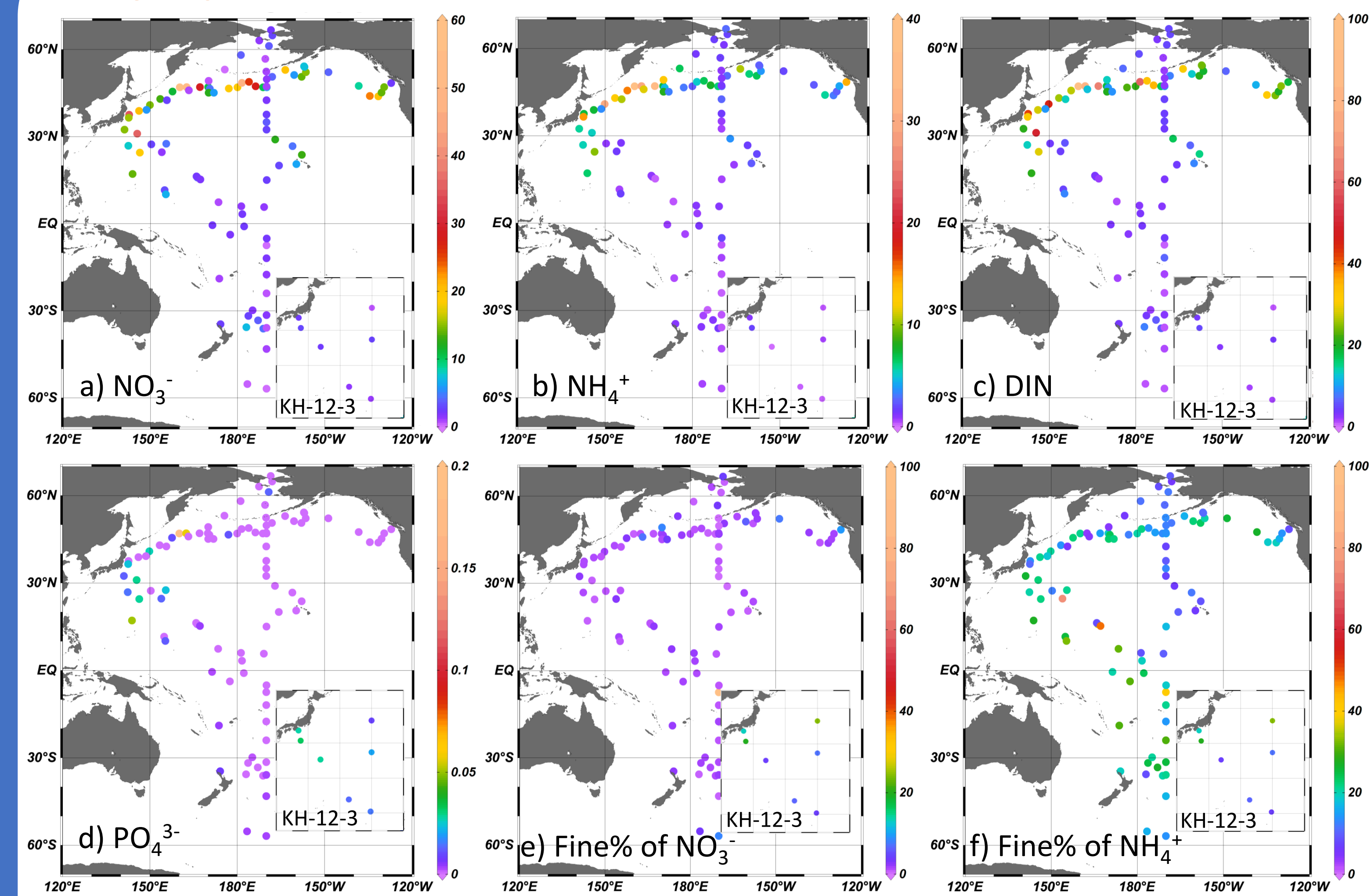
Figure 1. Distributions of dry deposition fluxes (μmol m^{-2} day^{-1}) of water soluble a) NO$_3^-$, b) NH$_4^+$, c) DIN and d) PO$_4^{3-}$, and percentages of fine modes in dry deposition for e) NO$_3^-$ and f) NH$_4^+$.
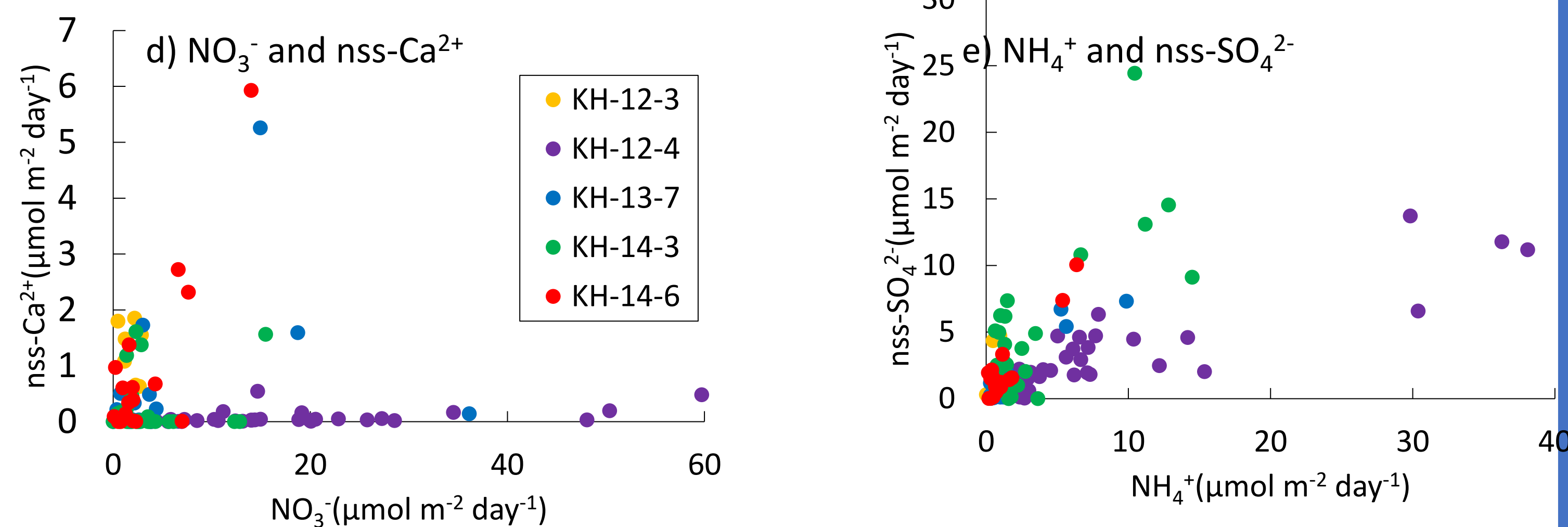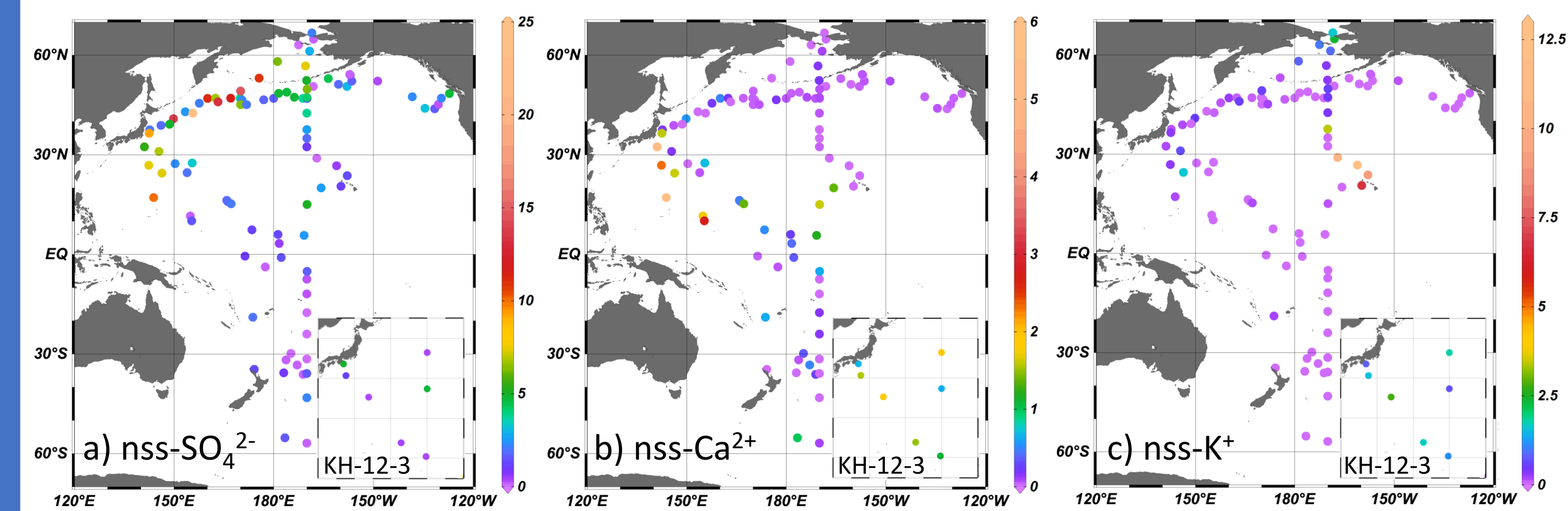　※ Data for the KH-12-3 cruise (quartz fiber filter papers were used) were shown separately.

Figure 2. Distributions of dry deposition fluxes (μmol m^{-2} day^{-1}) of water-soluble a) non-sea-salt (nss)-SO$_4^{2-}$, b) nss-Ca^{2+} and c) nss-K$^+$, and relationship between nss-ion and water-soluble d) NO$_3^-$ or e) NH$_4^+$.
　※ nss-SO$_4^{2-}$, index of anthropogenic aerosol; nss-Ca^{2+}, index of soil particles;
　　nss-K$^+$, index of biomass burning

7. Wet deposition fluxes

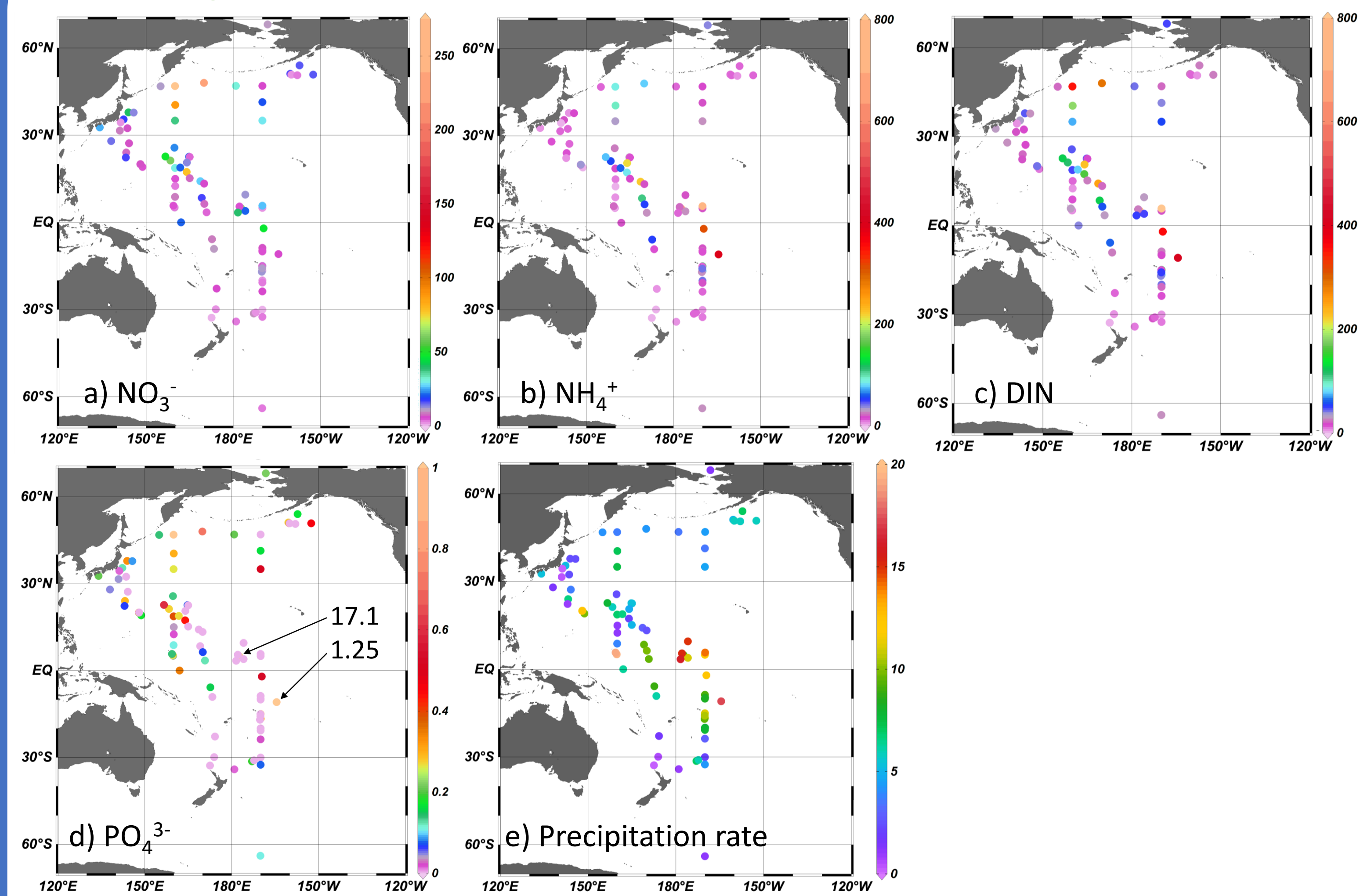Figure 3. Distributions of wet deposition fluxes (μmol m^{-2} day^{-1}) of water soluble a) NO$_3^-$, b) NH$_4^+$, c) DIN, d) PO$_4^{3-}$, and distribution of precipitation rate (mm day^{-1}).
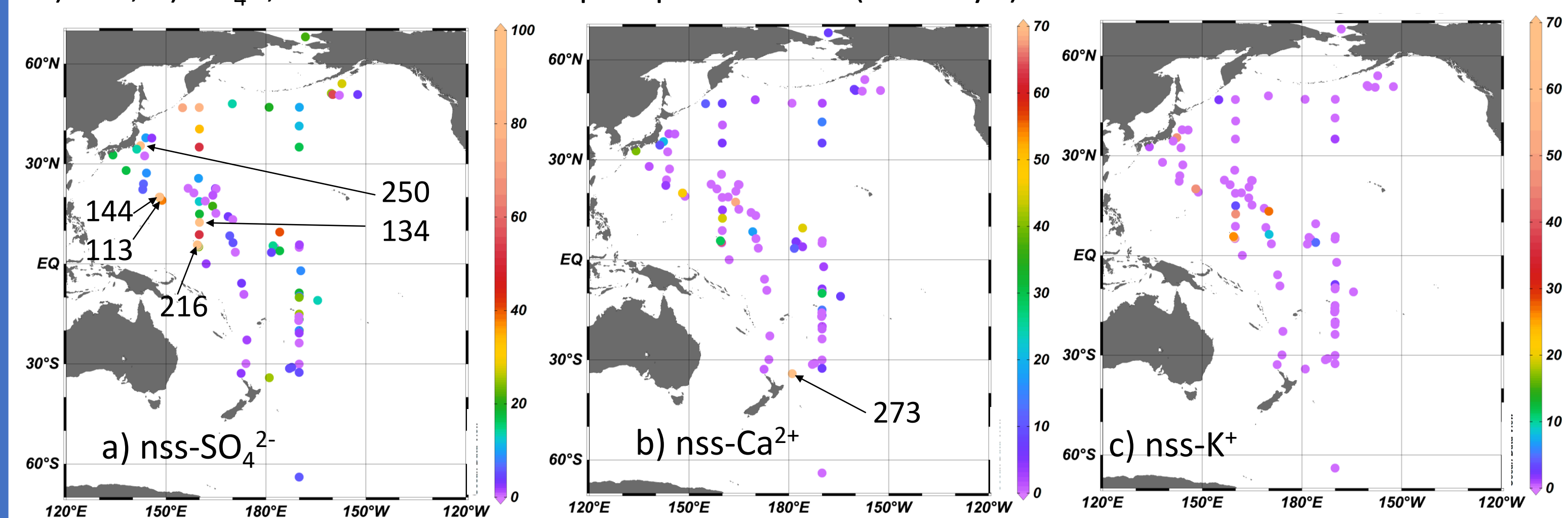
Figure 4. Distributions of wet deposition fluxes (μmol m^{-2} day^{-1}) of water-soluble a) nss-SO$_4^{2-}$, b) nss-Ca^{2+} and c) nss-K$^+$.

8. Findings

High inorganic N (IN) dry depositions were observed at the western North Pacific near the east coast of Japan and the south-east of the Kamchatka Peninsula in summer, with a mean contribution of NO3- to IN dry deposition of 62 %. Wet deposition of IN was mainly NH4+ (71 % on average over the Pacific Ocean), and NH4+ wet deposition was high in the equatorial region, which was affected by airmasses passing over relatively high chlorophyll a waters during the summer and winter. Dry depositions of P were at very low levels, but the equatorial region had high P wet deposition, about 30 times higher than average in winter 2014.

References

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[3] Duce, R.A., et al. (1991): Global Biogeochem. Cycles 5, 193–259.
[4] Huffman, G.J., et al. (2007): J. Hydrometeor. 8, 38-55, doi:10.1175/JHM560.1.