

1 Atmospheric inorganic nitrogen input via dry, wet, and sea 2 fog deposition to the subarctic western North Pacific 3 Ocean

4
5 J. Jung^{1,2}, H. Furutani¹, M. Uematsu¹, S. Kim², and S. Yoon²

6 [1]{Atmosphere and Ocean Research Institute, The University of Tokyo, Kashiwa, Japan}

7 [2]{School of Earth and Environmental Sciences, Seoul National University, Seoul, South
8 Korea}

9 Correspondence to: J. Jung (jinyoung_jung@snu.ac.kr)

10 11 Abstract

12 Aerosol, rainwater, and sea fog water samples were collected during the cruise conducted
13 over the subarctic western North Pacific Ocean in the summer of 2008, in order to estimate
14 dry, wet, and sea fog deposition fluxes of atmospheric inorganic nitrogen (N). During sea fog
15 events, mean number densities of particles with diameter larger than 0.5 μm decreased by 12–
16 78%, suggesting that particles with diameters larger than 0.5 μm could act preferentially as
17 condensation nuclei (CN) for sea fog droplets. Mean concentrations of nitrate (NO_3^-),
18 methanesulfonic acid (MSA), and non sea-salt sulfate (nss-SO_4^{2-}) in sea fog water were
19 higher than those in rainwater, whereas those of ammonium (NH_4^+) in both sea fog water and
20 rainwater were similar. These results reveal that sea fog scavenged NO_3^- and biogenic sulfur
21 species more efficiently than rain. Mean dry, wet, and sea fog deposition fluxes for
22 atmospheric total inorganic N (TIN; i.e. $\text{NH}_4^+ + \text{NO}_3^-$) over the subarctic western North
23 Pacific Ocean were estimated to be 4.9 $\mu\text{mol m}^{-2} \text{d}^{-1}$, 33 $\mu\text{mol m}^{-2} \text{d}^{-1}$, and 7.8 $\mu\text{mol m}^{-2} \text{d}^{-1}$,
24 respectively. While NO_3^- was the dominant inorganic N species in dry and sea fog deposition,
25 inorganic N supplied to surface waters by wet deposition was predominantly by NH_4^+ . The
26 contribution of dry, wet, and sea fog deposition to total deposition flux for TIN (46 $\mu\text{mol m}^{-2}$
27 d^{-1}) were 11%, 72%, and 17%, respectively, suggesting that ignoring sea fog deposition
28 would lead to underestimate of the total influx of atmospheric inorganic N into the subarctic
29 western North Pacific Ocean, especially in summer periods.

1

2 **1 Introduction**

3 Atmospheric transport of particulate matter from the continents to the oceans is well
4 recognized as a major pathway for supply of natural and anthropogenic materials to open
5 ocean surface waters, including nutrients (Duce et al., 1991; Jickells, 1995). Previous studies
6 have highlighted the significance of the atmosphere as a pathway for transport of essential
7 nutrients for biological growth such as nitrogen (N), from continents to marine surface waters
8 and its critical role in oceanic biogeochemical cycling (e.g. Duce et al., 1991; Prospero et al.,
9 1996; Paerl, 1997; Galloway et al., 2004; Dentener et al., 2006; Duce et al., 2008; Baker et al.,
10 2010; Kim et al., 2011; Jung et al., 2011).

11 While numerous studies have dealt with the input of nutrients via atmospheric deposition,
12 especially dry and wet deposition, relatively little is known about the deposition flux of
13 atmospheric constituents by fog (Lange et al., 2003). Scavenging processes of water-soluble
14 gases (e.g. HNO₃, NH₃, and SO₂) and aerosols in the atmosphere by fog events are
15 determined by the properties of ionic compositions in fog water and by the growth rate of fog
16 droplets during fog events (Aikawa et al., 2007). The chemical compositions of the particles
17 acting as condensation nuclei (CN) determine the initial compositions of the fog droplets,
18 which can be further altered by uptake of water-soluble gases and by aqueous phase chemical
19 reactions (Sasakawa et al., 2003; Raja et al., 2008). In fog, the condensation of water vapor on
20 pre-existent particles in the boundary layer shifts the aerosol size distribution towards larger
21 sizes and accelerates their removal from the atmosphere (Jacob et al., 1984; Sasakawa et al.,
22 2003; Herckes et al., 2007; Li et al., 2011). The deposition of fog can contribute significantly
23 to the hydrologic, pollutant, and nutrient cycles in coastal and mountainous regions, since it is
24 an important transfer process for water and various inorganic and organic substances from the
25 atmosphere to the biosphere (Lovett et al., 1982; Jacob et al., 1984; Collett et al., 2001; Zhang
26 and Anastasio, 2001; Klemm and Wrzesinsky, 2007). However, quantifying fog deposition
27 flux for atmospheric nutrient or pollutant and assessing its impact are still a challenge in
28 atmospheric science and ecosystem research (Klemm and Wrzesinsky, 2007).

29 Considerable effort has been devoted to investigating the chemical and physical properties of
30 fog in valleys, mountains, and urban areas (e.g. Collett et al., 2001; Burkard et al., 2002;
31 Collett et al., 2002; Moore et al., 2004; Lu et al., 2010; Li et al., 2011). However, sea fog has
32 not been extensively investigated (e.g. Sasakawa and Uematsu, 2002; Sasakawa et al., 2003;

1 Sasakawa and Uematsu, 2005); although it may stimulate phytoplankton growth over the
2 oceanic regions where sea fog occurs frequently and atmospheric nutrients derived from
3 natural and anthropogenic sources are transported and/or affected (Sasakawa et al., 2003). It is
4 therefore necessary to clarify the scavenging process of atmospheric nutrients by sea fog and
5 to quantify their deposition flux to the sea surface.

6 Rapid growth in human population and industrial activity have led to increases in the
7 concentrations of reactive N species throughout the environment (Galloway et al., 2008). In
8 particular, the increase in nitrogen oxides (NO_x) emissions in eastern Asia has been dramatic
9 over the last decade (Akimoto, 2003; Uno et al., 2007). The western North Pacific receives a
10 large influx of mineral particles and pollutants from the Asian continent through atmospheric
11 long-range transport (Uematsu et al., 1983; Gao et al., 1992; Nakamura et al., 2005; Uematsu
12 et al., 2010). Accordingly, estimating deposition flux of atmospheric N and evaluating its
13 impact on marine biogeochemical cycles over the western North Pacific have become
14 increasingly important. In addition, the subarctic western North Pacific (> 40°N) has a high
15 sea fog frequency, with a maximum of ~50% during the summertime period from June to
16 August (Wang, 1985). Nevertheless, no study has been carried out over this region to estimate
17 atmospheric N input via dry, wet, and sea fog deposition simultaneously. This study therefore
18 aims to (1) investigate general characteristics of sea fog, (2) estimate the fluxes of
19 atmospheric N via dry, wet, and sea fog deposition, (3) estimate the contribution of each
20 deposition to atmospheric N input, and (4) evaluate the impact of atmospheric N deposition
21 on the ocean marine ecosystem. In this study, we focus on ammonium (NH₄⁺) and nitrate
22 (NO₃⁻) that are dominant components for N supply to the oceans (Krishnamurthy et al., 2010).
23 The results for atmospheric inorganic N deposition from this study should be valuable for
24 filling the data gap, especially for the atmospheric inorganic N input by sea fog deposition to
25 the subarctic western North Pacific Ocean.

26

27 **2 Methods**

28 Aerosol (n = 11), rain (n = 7), and sea fog (n = 15) samples were collected over the subarctic
29 western North Pacific Ocean during Leg 1 of the KH-08-2 cruise (29 July–19 August 2008)
30 aboard R/V *Hakuho Maru* (Fig. 1).

1 **2.1 Aerosol collection**

2 A high-volume virtual impactor air sampler (AS-9, Kimoto Electric Co., Ltd.) was used to
3 collect marine aerosols on a Teflon filter (PF040, 90 mm in diameter, Advantec) (Nakamura
4 et al., 2005; Jung et al., 2011). The virtual impactor air sampler utilizes an acceleration nozzle
5 to divide the intake air into the major flow carrying the fine particles ($D < 2.5 \mu\text{m}$) and the
6 minor flow carrying the coarse particles ($D > 2.5 \mu\text{m}$). The major air flow is diverted laterally
7 after passing through the acceleration nozzle and the inertia of the coarse particles carries
8 them into a collection probe aligned with the acceleration nozzle. Both particle fractions are
9 then collected on the same filter. Details of the virtual impactor are described in Loo and Cork
10 (1988). The aerosol sampler was put on the front of the upper deck (17 m a.s.l.) of the ship. A
11 wind-sector controller was used to avoid contamination from the ship's exhaust during
12 aerosol sampling. The wind-sector controller system was configured to allow collection of
13 marine aerosol samples only when the relative wind directions were within plus or minus
14 100° relative to the ship's bow and the relative wind speeds were over 1 m s^{-1} during the
15 cruise. The flow rate was approximately $13 \text{ m}^3 \text{ h}^{-1}$ and the total sampling time was 1–3 days,
16 representing a total sampling air volume of $310\text{--}930 \text{ m}^3$. After sampling, the filter was stored
17 frozen at -24°C prior to chemical analysis. Deployment blanks ($n = 3$) were obtained by
18 placing Teflon filters in the aerosol sampler for 5 min on idle systems (i.e. no airflow through
19 the filters) and processed as other aerosol samples.

20 During the KH-08-2 cruise, number densities of ambient particles in four size fractions of $D >$
21 $0.3, 0.5, 1.0,$ and $2.0 \mu\text{m}$ were also measured continuously using an optical particle counter
22 (KC-01D, Rion Co., Inc). Meteorological variables (i.e. wind speed, wind direction, air
23 temperature, sea temperature, dew point, and relative humidity) were continuously monitored
24 by weather monitoring systems equipped on the research vessel.

25 **2.2 Rainwater collection**

26 Rainwater sampling was conducted on an event basis during the cruise. Rain samples were
27 collected with a 36.5 cm diameter polyethylene funnel fitted to a 500 ml polyethylene bottle
28 (Jung et al., 2011). Both the funnel and bottle were first soaked in detergent (Contaminon B,
29 Wako Pure Chemical Industries) for 24 h, then soaked in 1 N HCl for 24 h, before being
30 rinsed at least three times with Milli-Q water ($>18 \text{ M}\Omega \text{ cm}^{-1}$; Millipore Co.) and finally dried
31 in a drying oven (DK 600, Yamato, Japan) prior to deployment. The rain sampler was put on

1 the front of the upper deck of the ship, and was opened manually just before or as soon as
2 possible after precipitation. During rain collection, the relative wind directions were
3 monitored. If rain occurred when the relative wind directions were outside the ranges of the
4 wind-sector controller, the rainwater was not collected. After collection, the rain sampler was
5 washed thoroughly with Milli-Q water and closed. The rainwater samples were immediately
6 separated into three aliquots. Two of the aliquots were used for measurements of pH (Model
7 290A, Orion) and conductivity (Model 115, Orion), respectively. When the amount of
8 precipitation was less than 10 ml, pH and conductivity were not measured. The pH and
9 conductivity meters were calibrated before each measurement. Standard pH 4.01 and 7.00
10 buffer (Thermo Scientific) and conductivity/total dissolved solids (TDS) standard (1,413 μS
11 cm^{-1} , Thermo Scientific) solutions were used for calibrations of the pH and conductivity
12 meters, respectively. As the third aliquot, remaining rainwater was sealed in pre-cleaned 100
13 ml high-density polyethylene (HDPE) bottles, and stored in a freezer at -24°C prior to
14 chemical analyses. Procedural blanks ($n = 3$) for rainwater were collected by pouring 100 ml
15 of Milli-Q water through the clean funnel-bottle assembly. The procedural blanks were also
16 treated as other rainwater samples.

17 **2.3 Sea fog water collection**

18 Sea fog water sampling was conducted on an event basis during the cruise. A fog water
19 sampler (FWG-400, Usui Co. Inc.) was used to collect sea fog water and put on the front of
20 the upper deck of the ship. The fog water sampler is composed of a net of Teflon strings (0.5
21 mm in diameter), a net holder and a 500 ml low-density polyethylene (LDPE) bottle. Prior to
22 deployment, both the net and the bottle were first soaked in the detergent for 24 h, then
23 soaked in a 1N HCl for 24 h, then rinsed at least three times with Milli-Q water and finally
24 dried. The net of Teflon strings and the LDPE bottle were set only during the sea fog
25 occurrence and removed once sea fog event ceased. When the ship sails the sea fog
26 occurrence area, sea fog droplets collide with the strings and drop along the strings into the
27 500 ml LDPE bottle beneath the strings (Sasakawa et al., 2003). Likewise rain sampling, the
28 relative wind directions were monitored during sea fog water collection, and sea fog water
29 samples were collected only when the relative wind directions were within the ranges of the
30 wind-sector controller. If the relative wind directions changed from within to outside the
31 ranges during sea fog water sampling, the sea fog water was considered contaminated and
32 discarded. After collection, sea fog water samples were immediately separated into three

1 aliquots. Two of the aliquots were used for measurements of pH and conductivity,
2 respectively. The third aliquot was sealed in pre-cleaned 100 ml high-density polyethylene
3 (HDPE) bottles, and stored in a freezer at -24°C prior to chemical analyses. Procedural blanks
4 ($n = 5$) for sea fog water samples were collected by pouring 100 ml of Milli-Q water through
5 the clean net-bottle assembly. The procedural blanks were also treated as sea fog water
6 samples.

7 Size distributions of sea fog droplets were measured with a fog monitor (FM-100, Droplet
8 Measurement Technologies). The fog monitor detects the number and size of individual fog
9 droplets with a diameter from roughly $2\ \mu\text{m}$ up to $50\ \mu\text{m}$ by the forward scattering principle,
10 and can classify droplets in up to 40 size classes (Klemm et al., 2005). Liquid water content
11 (LWC) for each of the 40 droplet size classes was computed based on an idealized mean
12 volume of spherical droplets with aerodynamic diameter. Total LWC was obtained from the
13 sum of LWC for all size. Until now, the uncertainties of droplet measurements for the fog
14 monitor have not been considered in the most studies. Recently, Spiegel et al. (2012)
15 evaluated the influence of Mie scattering on the droplet size spectra collected with the fog
16 monitor and the droplet losses during sampling with the fog monitor. They also recommended
17 choosing the 40 channel thresholds to reduce the error from Mie scattering, and doing loss
18 calculations for the droplet measurements using an ultrasonic anemometer and other
19 instrument (e.g. Particulate Volume Monitor) for a reference. In this study, the 40 channel
20 thresholds were used for the measurement of sea fog droplets, indicating that it is sufficient
21 for the determination of the total droplet number concentration or the total liquid water
22 content (Spiegel et al., 2012). However, the droplet losses during sampling with the fog
23 monitor were not calculated since an ultrasonic anemometer and other instrument for a
24 reference were not used in this study.

25 **2.4 Chemical analysis**

26 Aerosol, rainwater, and sea fog water samples were analyzed for major ionic and inorganic N
27 species (NH_4^+ and NO_3^-) using the method described in detail elsewhere (Jung et al., 2011).
28 Briefly, aerosol samples were ultrasonically extracted using 50 ml of Milli-Q water. The
29 extraction solution was then filtered, as were the rainwater and sea fog water samples, through
30 a 13-mm diameter, $0.45\text{-}\mu\text{m}$ pore-size membrane filter (PTFE syringe filter, Millipore Co.).
31 The filtrates of aerosol extracts, rainwater, and sea fog water samples were analyzed by ion
32 chromatography (IC; Dionex-320, Thermo Scientific Dionex) for anions (Cl^- , MSA, NO_3^- ,

1 and SO_4^{2-}) and cations (Na^+ , NH_4^+ , K^+ , Mg^{2+} , and Ca^{2+}). The instrumental detection limits
2 were: Cl^- 0.035 μM , MSA 0.031 μM , NO_3^- 0.1 μM , SO_4^{2-} 0.065 μM , Na^+ 0.11 μM , NH_4^+
3 0.17 μM , K^+ 0.16 μM , Mg^{2+} 0.15 μM , and Ca^{2+} 0.16 μM .

4 Non sea-salt (nss-) concentrations of some ionic components were calculated by subtracting
5 the component's sea-salt-derived (ss-) concentration from its total concentration. In this study,
6 it was assumed that all Na^+ in aerosols, rainwater and sea fog water were derived from sea-
7 salt. Contributions from sea-salt were calculated from the Na^+ concentration in aerosols,
8 rainwater or sea fog water using the mole ratio of the component of interest to Na^+ in seawater
9 (Keene et al., 1986).

10 **2.5 Backward trajectory analysis**

11 Air mass backward trajectories (AMBTs) provide a better understanding of air flow and long-
12 range transport of aerosols. In particular, AMBTs have been used to identify the origin of
13 primary aerosols collected far away from their source region (Chiapello et al., 1997). In this
14 study, 7-day AMBTs were calculated from the National Oceanic and Atmospheric
15 Administration (NOAA) GDAS (Global Data Assimilation System) database using the
16 Hybrid Single-Particle Lagrangian Integrated Trajectories (HY-SPLIT) model (NOAA Air
17 Resources Laboratory, <http://www.arl.noaa.gov/ready/hysplit4.html>). AMBTs were
18 performed at 500, 1000, and 1500 m above ground level. Chen and Siefert (2004) reported
19 that atmospheric aerosols may not follow the resulting trajectories because of scavenging
20 processes and gravitational settling; however, the AMBTs provide useful background data on
21 airstreams and the potential origins of the source of the sampled air mass.

22 **2.6 Deposition flux estimates**

23 **2.6.1 Dry deposition**

24 Dry deposition fluxes (F_d) were calculated from aerosol concentrations (C_a) in the coarse (c)
25 and fine (f) modes and dry deposition velocities (V_d) for each size mode (Duce et al., 1991;
26 Baker et al., 2007):

$$27 \quad F_d = C_a^c \times V_d^c + C_a^f \times V_d^f \quad (1)$$

28 Here, dry deposition velocities of 2 cm s^{-1} for coarse mode and 0.1 cm s^{-1} for fine mode were
29 used since these two values are known to be best estimates based on experimental and model

1 studies (e.g. Duce et al., 1991; Baker et al., 2003; Nakamura et al., 2005). This estimate
2 results in an uncertainty of a factor of 2–3 in the calculated flux, since deposition velocity
3 includes terms for gravitational settling, impaction and diffusion of particles, all of which
4 vary in complex functions of particle size and meteorological conditions (e.g. wind speed and
5 relative humidity) (Duce et al., 1991).

6 **2.6.2 Wet deposition**

7 Wet deposition fluxes (F_w) were estimated from the concentration of the species of interest in
8 rainwater (C_r) and the precipitation rate (P) (Baker et al., 2010):

$$9 \quad F_w = C_r \times P \quad (2)$$

10 The precipitation rate was calculated from the monthly averaged precipitation rate (mm d^{-1})
11 using the CMAP model output (<http://www.cdc.noaa.gov/cdc/data.cmap.html>) (Xie and Arkin
12 1997). Similar to estimates for dry deposition flux, the choice of precipitation rates based on
13 limited data causes the greatest uncertainty in wet deposition flux estimates, particularly in the
14 open ocean (Spokes et al., 2000). However, Baker et al. (2010 and references therein), who
15 used the same method for estimating wet deposition flux, argued that the uncertainty arising
16 from selection of precipitation rate is minor since the precipitation rate data agreed relatively
17 well with other studies in terms of total rainfall amount.

18 **2.6.3 Sea fog deposition**

19 Sea fog deposition fluxes (F_f) were estimated by multiplying sea fog water flux (F_{fw}) of each
20 event by the concentration of the species of interest in each sea fog water sample (C_f)
21 (Thalmann et al., 2002):

$$22 \quad F_f = F_{fw} \times C_f \quad (3)$$

23 The F_{fw} was estimated by Eq. 4, where $LWC(D_p)$ represents the mean LWC for each of the 40
24 droplet size classes during each sea fog event and $V(D_p)$ indicates the deposition velocity for
25 sea fog droplets with an aerodynamic diameter (Klemm et al., 2005). The sum of sea fog
26 water flux for all size classes then yielded the F_{fw} .

$$27 \quad F_{fw} = \sum LWC(D_p) \times V(D_p) \quad (4)$$

28 For $V(D_p)$, the modeled values reported by Matsumoto et al. (2011), who calculated the
29 deposition velocities for the particles with diameters larger than 3 μm , were used (i.e. 4.69 cm

1 s^{-1} for $3 < D < 5 \mu\text{m}$, 10.1 cm s^{-1} for $5 < D < 10 \mu\text{m}$, 13.5 cm s^{-1} for $10 < D < 20 \mu\text{m}$, 15.6 cm
2 s^{-1} for $20 < D < 30 \mu\text{m}$ and 19.0 cm s^{-1} for $30 < D < 50 \mu\text{m}$). This ambiguity in values used
3 for sea fog deposition velocities leads to the greatest uncertainty in sea fog flux estimates.

4 In this study, it was assumed that all sea fog droplets measured with the fog monitor were
5 deposited to the sea surface without changes in size distribution of LWC; however, due to
6 evaporation and coalescence of sea fog droplets, their size and deposition velocities are
7 subject to change, suggesting that the deposition velocities used here leads to the uncertainty
8 in sea fog deposition flux estimates. The fog water sampler used in this study has a 50%
9 efficiency collection diameter of $6 \mu\text{m}$ under flow rate 3 m s^{-1} (Minami and Ishizaka, 1996). It
10 is difficult, however, to calculate the precise collection efficiency of this fog sampler in this
11 study, because the relative wind directions and the relative wind speeds change extremely
12 with the movements of the ship (Sasakawa and Uematsu, 2005). Hence, the estimates of sea
13 fog deposition fluxes in this study contain the uncertainties that are related to the changes in
14 size distribution of LWC and the collection efficiency.

15 **3 Results and discussion**

16 **3.1 Meteorological conditions associated with sea fog occurrences**

17 Sea fog typically occurs as a result of cooling of humid air over a cold ocean surface (Lewis
18 et al., 2004; Eugster, 2008). During the sampling period, sea fog occurred predominantly
19 when the dominant wind direction was southerly and/or southwesterly and air temperature
20 dropped to its dew point (Fig. 2). This result indicates that the warm and humid air masses
21 from the low and middle latitudes of the North Pacific Ocean passed over the cold sea surface
22 of the subarctic western North Pacific Ocean and they were cooled down to a saturation
23 temperature. Sea fog events, however, did not occur when sea surface temperature was higher
24 than air temperature, although the dominant wind direction was southerly, suggesting that the
25 difference between the air and sea surface temperatures is a key factor controlling sea fog
26 formation (Cho et al., 2000).

27 According to previous studies (Cho et al., 2000; Fu et al., 2006; Tokinaga and Xie, 2009), the
28 difference between air and sea surface temperatures is often observed to be positive in
29 frequent sea fog occurrence regions, since the relatively cold sea surface temperature
30 stabilizes the lower atmosphere, making a favorable condition for sea fog formation.
31 Therefore, the meteorological conditions during the sea fog sampling period show that

1 advection of warm and humid air masses from the subtropical North Pacific Ocean and the
2 positive difference between air and sea surface temperatures make favorable conditions for
3 sea fog occurrence over the subarctic western North Pacific Ocean.

4 **3.2 Effect of sea fog on particle number density**

5 Temporal variations of total LWC, particle number densities for aerosols in the range of 0.3–
6 2.0 μm , and fog droplet size distribution for each of the 40 droplet size classes during the
7 cruise are shown in Fig. 3. The total LWC varied from < 0.2 – 140 mg m^{-3} . Mean particle
8 number densities during non sea fog events were 25 cm^{-3} for aerosols in the range of $0.3 < D$
9 $< 0.5 \mu\text{m}$, 2.6 cm^{-3} for $0.5 < D < 1.0 \mu\text{m}$, 0.53 cm^{-3} for $1.0 < D < 2.0 \mu\text{m}$, and 0.17 cm^{-3} for D
10 $> 2.0 \mu\text{m}$. In comparison, the mean particle number densities during sea fog events decreased
11 by 4% (mean particle number density 24 cm^{-3}) for aerosols in the range of $0.3 < D < 0.5 \mu\text{m}$,
12 12% (2.3 cm^{-3}) for $0.5 < D < 1.0 \mu\text{m}$, 55% (0.24 cm^{-3}) for $1.0 < D < 2.0 \mu\text{m}$, and 78% (0.038
13 cm^{-3}) for $D > 2.0 \mu\text{m}$. The differences of mean particle number densities between two periods
14 were statistically significant (Wilcoxon Signed Rank Test, $p = 0.0005$), except that for
15 aerosols in the range of $0.3 < D < 0.5 \mu\text{m}$. In addition, the sea fog droplet size distributions
16 during sea fog events were shifted towards the larger droplet sizes. These results suggest that
17 particles with diameters larger than $0.5 \mu\text{m}$ could act preferentially as CN for sea fog droplets
18 (Sasakawa et al., 2003), and that the formation of liquid droplets by condensation of water
19 vapor on pre-existed particles acting as CN leads to the acceleration of particle removal from
20 the atmosphere (Pandis and Seinfeld, 1990).

21 **3.3 Chemical composition of aerosols, rainwater, and sea fog water**

22 Sea-salts (Na^+ , Cl^- , and ss-ions) were the dominant components in aerosols, rainwater, and
23 sea fog water, representing approximately 72%, 61%, and 86% of total ionic concentration,
24 respectively (Fig. 4 and Table 1). In aerosols and sea fog water, the mole equivalent ratios of
25 Cl^-/Na^+ were slightly lower than that in seawater (Table 2), suggesting that Cl^- depletion
26 occurred through the volatilization of hydrogen chloride (HCl) from sea-salt particles that
27 became acidified by the incorporation of nitric (HNO_3) and/or sulfuric (H_2SO_4) acids in the
28 marine atmosphere (Graedel and Keene 1995; Andreae and Crutzen 1997), and that the
29 acidified sea-salt particles acted as CN of sea fog droplets (Sasakawa and Uematsu, 2002;
30 Raja et al., 2008). Furthermore, the $\text{Mg}^{2+}/\text{Na}^+$, K^+/Na^+ , and $\text{Ca}^{2+}/\text{Na}^+$ ratios in aerosols and sea

1 fog water were similar or slightly higher than those in seawater, suggesting that most of Mg^{2+} ,
2 K^+ and Ca^{2+} in aerosols and sea fog water were derived from sea-salt particles.

3 Two volcanoes on the Aleutians erupted during the cruise. The eruption at Okmok volcano
4 (53.40°N , 168.17°W) started on 12 July 2008 and ended in late August 2008 (Larsen et al.,
5 2009; Lu and Dzurisin, 2010), and Kasatochi volcano (52.18°N , 175.51°W) became active on
6 7 August 2008 (Schmale et al., 2010). During the sampling period, air masses originated from
7 the Asian continent and the Kamchatka Peninsula, indicating that these air masses were likely
8 affected by anthropogenic and crustal sources as well as the eruptions of two volcanoes (Figs.
9 5a–5c).

10 Unlike aerosol and sea fog water samples, the Cl^-/Na^+ , $\text{Mg}^{2+}/\text{Na}^+$, K^+/Na^+ , and $\text{Ca}^{2+}/\text{Na}^+$
11 ratios in rainwater were much higher than those in seawater (Table 2). Atmospheric HCl are
12 derived from sea-salt particles, volcanoes, and anthropogenic activities (e.g. fossil fuel
13 combustion and incineration) (Graedel and Keene, 1995). Gioda et al. (2011) observed high
14 Cl^-/Na^+ ratios in rainwater (2.2) and cloud water (3.2), collected in Puerto Rico from
15 December 2004 to March 2007, when ash from the Soufriere Hills volcano reached the
16 sampling site. During the collection of rainwater samples, air masses originated from the
17 Asian continent or the subtropical western North Pacific Ocean and thereafter swept over
18 large regions of the Korean Peninsula and/or the Japanese Islands, indicating that these air
19 masses were most likely affected by strong anthropogenic and crustal sources rather than by
20 the influences of two volcanoes (Fig. 5b). The high Cl^-/Na^+ ratio in rainwater thus is likely
21 due to scavenging of HCl derived from sea-salt particles and/or anthropogenic source by
22 rainwater. Sasakawa and Uematsu (2002) reported that $\text{NH}_4^+/\text{nss-Ca}^{2+}$ ratio in rainwater
23 (0.53) collected over the northwestern North Pacific (34°N – 48°N , 137°E – 155°E) from 15–29
24 July 1998, was two orders of magnitude lower than that in sea fog water (11), reflecting that
25 mineral particles, such as calcium carbonate (CaCO_3), mainly existed over higher altitudes
26 than those where sea fog appeared. While sea fog occurs near the sea surface (Fu et al., 2006)
27 and scavenges only lower atmospheric substances (Ali et al., 2004), precipitation removes the
28 substances existing in the whole air column in and below clouds (Deboudt et al., 2004). The
29 high $\text{Mg}^{2+}/\text{Na}^+$, K^+/Na^+ and $\text{Ca}^{2+}/\text{Na}^+$ ratios in rainwater therefore suggest that most of these
30 ionic species in rainwater were derived from other sources, such as crustal materials and
31 biomass burning, and that scavenging processes of aerosols by sea fog are different to those
32 by rain.

1 3.4 The pH of rainwater and sea fog water

2 The pH values of rainwater and sea fog water varied from 3.5–4.5 and 3.4–5.9, with averages
3 of 4.1 and 4.2, respectively (Table 1). Several compounds, such as H₂SO₄, HNO₃, HCl, NH₃,
4 and CaCO₃, contribute to the acid-base balance of rainwater and fog water (Millet et al.,
5 1996).

6 The mean nss-Cl⁻ concentration in rainwater was two orders of magnitude greater than those
7 of nss-SO₄²⁻ and NO₃⁻ (Table 1), suggesting that nss-Cl⁻ exerted a larger influence on acidity
8 of rainwater collected over the subarctic western North Pacific Ocean. Assuming the nss-
9 SO₄²⁻, NO₃⁻, and nss-Cl⁻ in rainwater existed in the form of free acids, the expected pH of
10 rainwater was 3.4, whereas the mean pH value measured in rainwater was 4.1. This
11 discrepancy indicates that rainwater had experienced some neutralization. From the difference
12 between the sum of nss-SO₄²⁻, NO₃⁻, nss-Cl⁻, and the mean H⁺ concentration obtained from
13 the mean pH value, it was estimated that approximately 61% of these acidic substances was in
14 neutralized forms. The mean concentrations of nss-Ca²⁺ and nss-Mg²⁺ in rainwater were 15
15 times and 3.8 times higher than that of NH₄⁺, respectively (Table 1). Moreover, the mole
16 equivalent ratios of NH₄⁺/nss-Ca²⁺ and NH₄⁺/nss-Mg²⁺ in rainwater were lower than 1,
17 suggesting that nss-Ca²⁺ and nss-Mg²⁺ played key roles in neutralization of rainwater acidity.

18 In sea fog water, nss-SO₄²⁻ and NO₃⁻ were the dominant acidic species (Fig. 4 and Table 1).
19 The mole equivalent ratio of NO₃⁻/nss-SO₄²⁻ in sea fog water was 0.57 (Table 2). This result
20 suggests that nss-SO₄²⁻ was the major component to lower the pH of sea fog, and that the pH
21 of sea fog water was controlled by nss-SO₄²⁻ derived mainly from marine biological activity
22 than that of rainwater since mean concentrations of methanesulfonic acid (MSA) and nss-
23 SO₄²⁻ in sea fog water were an order of magnitude greater than those in rainwater (Table 1).
24 For sea fog water, it was estimated that approximately 48% of nss-SO₄²⁻ and NO₃⁻ was in
25 neutralized forms. While nss-Ca²⁺ and nss-Mg²⁺ were the dominant neutralization substances
26 in rainwater, NH₄⁺ was the major basic component in sea fog water (Fig. 4 and Table 2),
27 suggesting that neutralization of sea fog water was predominantly caused by NH₄⁺, and that
28 nss-SO₄²⁻ and NO₃⁻ in sea fog water were probably in fully or partially neutralized forms,
29 such as ammonium sulfate ((NH₄)₂SO₄), ammonium bisulfate (NH₄HSO₄) and ammonium
30 nitrate (NH₄NO₃).

1 **3.5 Concentrations of NH_4^+ and NO_3^- in aerosols, rainwater, and sea fog water**

2 **3.5.1 Aerosols**

3 Total concentrations of NH_4^+ and NO_3^- in bulk (fine + coarse) aerosols ranged from 2.9–9.8
4 neq m^{-3} and 0.64–5.6 neq m^{-3} , respectively (Figs. 6a and 6b). Mean concentrations of aerosol
5 inorganic N species were 5.6 neq m^{-3} for NH_4^+ and 2.5 neq m^{-3} for NO_3^- , accounting for
6 ~70% by NH_4^+ and ~30% by NO_3^- of aerosol total inorganic N (i.e. $\text{TIN} = \text{NH}_4^+ + \text{NO}_3^-$)
7 (Table 1). Ammonium is primarily associated with fine mode aerosol and produced by
8 heterogeneous reactions involving NH_3 derived from intensive agricultural activity (Aneja et
9 al., 2001), biomass burning (Andreae and Merlet, 2001) and a relatively weak marine source
10 (Jickells et al., 2003). It is also known that NO_3^- in the marine atmosphere is predominantly
11 associated with coarse mode aerosol as a result of a chemical reaction between HNO_3 derived
12 primarily from NO_x emissions from combustion processes and sea-salt (Andreae and Crutzen,
13 1997). Mean percentages of total aerosol concentration in the fine mode for NH_4^+ and NO_3^-
14 were ~84% and ~36%, respectively. These values were similar to the results of Nakamura et
15 al. (2005), who reported the size distributions of NH_4^+ and NO_3^- in aerosols collected over the
16 East China Sea.

17 During the sampling period, over a dozen sea fog events occurred and aerosol samples, A03–
18 A06 and A08–A09, were largely affected by these fog appearance. As shown in Figs. 6a and
19 6b, NO_3^- , which mainly existed in coarse mode aerosols, was more efficiently scavenged by
20 sea fog than NH_4^+ , showing that coarse particles act predominantly as CN of sea fog droplets
21 rather than the fine particles (see section 3.6). Sasakawa and Uematsu (2002) reported that
22 mean concentrations of NH_4^+ and NO_3^- in aerosols collected over the northwestern North
23 Pacific Ocean (34°N–48°N, 137°E–155°E) from 15–29 July 1998, were $11 \pm 2.9 \text{ neq m}^{-3}$ and
24 $3.7 \pm 2.2 \text{ neq m}^{-3}$, respectively. In comparison, the mean aerosol NH_4^+ and NO_3^-
25 concentrations in this study were a factor of 2 and 1.5 lower than their results, respectively.
26 The low mean NH_4^+ and NO_3^- concentrations in aerosols thus are likely due to strong
27 influences of sea fog.

28 **3.5.2 Rainwater**

29 When sea fog and rain events occurred simultaneously, only rainwater was collected (Fig. 3).
30 That sample was then considered as a rainwater sample (i.e. rainwater sample number 3 and
31 4), although the rainwater sample contains sea fog water as well as rainwater, since sea fog

1 water is deposited by rainwater during that time. Concentrations of NH_4^+ and NO_3^- in
2 rainwater ranged from 4.1–55 $\mu\text{eq L}^{-1}$ and 1.2–18 $\mu\text{eq L}^{-1}$, respectively (Figs. 6c and 6d).
3 Mean concentrations of inorganic N species were 25 $\mu\text{eq L}^{-1}$ for NH_4^+ and 7.9 $\mu\text{eq L}^{-1}$ for
4 NO_3^- (Table 1). These values were in the range of the observed NH_4^+ (1.7–67 $\mu\text{eq L}^{-1}$) and
5 NO_3^- (2.4–26 $\mu\text{eq L}^{-1}$) concentrations in rainwater collected over the northwestern North
6 Pacific Ocean (34°N–48°N, 137°E–155°E) from 15–29 July 1998 (Sasakawa and Uematsu,
7 2002). Inorganic N in rainwater was composed of ~77% NH_4^+ and ~23% NO_3^- (mean values),
8 suggesting that NH_4^+ is more abundant in rainwater collected over the subarctic western North
9 Pacific Ocean, and that it is a more important inorganic N species supplied by wet deposition.

10 **3.5.3 Sea fog water**

11 Concentrations of NH_4^+ and NO_3^- in sea fog water ranged from 4.2–45 $\mu\text{eq L}^{-1}$ and 1.8–139
12 $\mu\text{eq L}^{-1}$, respectively (Figs. 6e and 6f). Contributions of NH_4^+ and NO_3^- to TIN in sea fog
13 water were found to represent ~39% (mean concentration 22 $\mu\text{eq L}^{-1}$) and ~61% (mean
14 concentration 50 $\mu\text{eq L}^{-1}$), respectively (Table 1). Sasakawa and Uematsu (2002) reported that
15 mean NH_4^+ and NO_3^- concentrations in sea fog water collected over the northwestern North
16 Pacific Ocean (34°N–48°N, 137°E–155°E) from 15–29 July 1998, were $25 \pm 17 \mu\text{eq L}^{-1}$ and
17 $25 \pm 22 \mu\text{eq L}^{-1}$, respectively. The mean NH_4^+ concentration observed in this study was
18 comparable to their result; however, the mean concentration of NO_3^- was a factor of 2 higher
19 than their result for NO_3^- . This different concentration is likely attributable to the duration,
20 frequency of sea fog events, and changes in the quality of air mass.

21 **3.6 Difference of scavenging process between rain and sea fog**

22 Mean concentrations of NO_3^- , MSA, and nss-SO_4^{2-} in sea fog water were higher than those in
23 rainwater (Tables 1 and 3), suggesting that they were more effectively scavenged by sea fog
24 droplets with their higher surface-to-volume ratios and longer atmospheric residence times
25 (Sasakawa and Uematsu, 2002; Sasakawa and Uematsu, 2005; Gioda et al., 2011).

26 The sea fog water/rainwater ratio for NH_4^+ was 0.88 (Table 3), suggesting that NH_4^+ was not
27 scavenged by sea fog as efficiently as NO_3^- , MSA, and nss-SO_4^{2-} . The mean NO_3^-
28 concentration in sea fog water was 6.3 times higher than that in rainwater, whereas the mean
29 NH_4^+ concentration in sea fog water was similar to that in rainwater. Sasakawa et al. (2003)
30 reported that coarse particles (e.g. sea-salt particles and NaNO_3) act predominantly as CN of
31 sea fog droplets rather than NH_4^+ and nss-SO_4^{2-} particles, such as $(\text{NH}_4)_2\text{SO}_4$ and NH_4HSO_4

1 since aerosol NH_4^+ and nss-SO_4^{2-} are largely associated with the fine mode ($D < 2.5 \mu\text{m}$)
2 particles (Nakamura et al., 2005). In addition, NH_3 readily reacts with acids in the atmosphere
3 to form NH_4^+ aerosols that can act as cloud CN (Quinn et al., 1987). In this study, it was
4 observed that larger particles ($D > 0.5 \mu\text{m}$) preferentially became activated and the fog droplet
5 size distribution was shifted towards the larger droplet sizes (Fig. 3), and that NH_4^+ and NO_3^-
6 were largely associated with fine and coarse mode aerosols, respectively (Figs. 6a and 6b).
7 Therefore, higher NO_3^- concentration in sea fog water than in rainwater is likely due to
8 preferential behaviors of coarse particles as CN in sea fog.

9 While MSA is formed exclusively from dimethylsulfide (DMS) produced by phytoplankton
10 in the ocean, non sea-salt SO_4^{2-} has a variety of sources, including DMS oxidation, volcanic
11 and industrial sulfur emissions (Gondwe et al., 2003). Dimethylsulfide is emitted into the
12 atmosphere, where it undergoes chemical transformation to eventually form gaseous (e.g.
13 MSA and SO_2) and/or particulate (e.g. MSA and nss-SO_4^{2-}) sulfur species (e.g. Charlson et al.,
14 1987; Bardouki et al., 2003). Mean concentrations of MSA and nss-SO_4^{2-} in sea fog water
15 were 15 times and 13 times higher than those in rainwater, respectively (Tables 1 and 3).
16 During the sampling period, SeaWiFS satellite images revealed high chlorophyll a levels
17 (<http://oceancolor.gsfc.nasa.gov>) in the subarctic western North Pacific Ocean (see
18 supplementary material, Fig. S1). Considering sea fog occurs near the sea surface where DMS
19 is emitted, these results suggest that sea fog scavenged biogenic sulfur species more
20 effectively than rain (Sasakawa and Uematsu, 2005).

21 If aerosol particles exert the primary influence as CN of sea fog droplets, the mole equivalent
22 ratios between aerosols and sea fog water would be similar (Sasakawa and Uematsu, 2002;
23 Gioda et al., 2011). The mole equivalent ratio of $\text{NO}_3^-/\text{Na}^+$ in sea fog water was higher than
24 that in aerosols (Table 2). This result suggests that not only aerosol NO_3^- , but also gaseous
25 HNO_3 was scavenged by sea fog water. Nitric acid is highly soluble in water. Once a large
26 amount of liquid water has amassed, the gas phase HNO_3 is rapidly dissolved (Fahey et al.,
27 2005). In order to estimate the fraction of gaseous HNO_3 scavenged by sea fog water, the
28 $\text{NO}_3^-/\text{Na}^+$ ratios in aerosols was compared to those in sea fog water (Fig. 7). In 8 sea fog
29 samples, the higher $\text{NO}_3^-/\text{Na}^+$ ratios than in aerosols were observed when air masses
30 originated from the Asian continent or from the subtropical western North Pacific Ocean
31 circulated around the vicinity of the Japanese Islands and thereafter reached the sea fog
32 sampling sites (Fig. 5c). In this study, it was estimated that 25–94% (mean 74%) of NO_3^- in

1 the 8 sea fog water samples was derived from the dissolution of HNO_3 , suggesting that sea
2 fog over the subarctic western North Pacific Ocean is an important removal mechanism for
3 gas phase HNO_3 . In comparison, the $\text{NH}_4^+/\text{Na}^+$ ratios in sea fog water for all periods were
4 lower than those in aerosols (Fig. S2), indicating that sea-salt particles exerted a greater
5 influence on the $\text{NH}_4^+/\text{Na}^+$ ratios in sea fog water due to condensation occurred preferably on
6 coarse particles as mentioned above.

7 **3.7 Contributions of dry, wet, and sea fog deposition to atmospheric input of** 8 **nitrogen to the subarctic western North Pacific Ocean**

9 The measured NH_4^+ and NO_3^- concentrations in aerosols, rainwater, and sea fog water were
10 converted into fluxes using the equations described in section 2.6. For sea fog water flux, the
11 mean LWC for each of the 40 droplet size classes measured by the fog monitor during each
12 sea fog event was used (Fig. 8). Temporal variations of dry, wet, and sea fog deposition fluxes
13 for NH_4^+ and NO_3^- during the sampling period are shown in Fig. 9. The estimated dry
14 deposition fluxes for atmospheric inorganic N species ranged from $0.67\text{--}3.1 \mu\text{mol m}^{-2} \text{d}^{-1}$ for
15 NH_4^+ and from $0.62\text{--}8.6 \mu\text{mol m}^{-2} \text{d}^{-1}$ for NO_3^- , contributing $\sim 43\%$ by NH_4^+ and $\sim 57\%$ by
16 NO_3^- to the dry deposition flux for TIN. Mean dry deposition fluxes for NH_4^+ and NO_3^- were
17 estimated to be $1.9 \mu\text{mol m}^{-2} \text{d}^{-1}$ and $3.0 \mu\text{mol m}^{-2} \text{d}^{-1}$, respectively. Although the mean
18 concentration of total NH_4^+ in aerosols collected over the subarctic western North Pacific
19 Ocean was approximately 2 times higher than that of total NO_3^- (Table 1), inorganic N
20 supplied to surface waters by atmospheric dry deposition was mainly from NO_3^- that was
21 largely associated with coarse mode particles, since fluxes to the ocean are dominated by the
22 coarse mode, resulting in NO_3^- being deposited much more rapidly (Figs. 6a and 6b). In this
23 study, dry deposition fluxes of gaseous inorganic N species such as NH_3 and HNO_3 were not
24 estimated, resulting in the underestimate of dry deposition fluxes. Due to the lack of
25 observational data for gaseous inorganic N species over the western North Pacific Ocean, we
26 used the results of Zhang et al. (2010) to estimate the contributions of gaseous inorganic N to
27 dry deposition fluxes, who reported that dry deposition fluxes for NH_3 and HNO_3 were
28 estimated to be $2.0 \mu\text{mol m}^{-2} \text{d}^{-1}$ and $4.7 \mu\text{mol m}^{-2} \text{d}^{-1}$ respectively over the East China Sea in
29 July using the MM5/CMAQ model with the 2004 national emission inventory of China.
30 Assuming that the same amounts of NH_3 and HNO_3 are deposited over the sampling region of
31 this study, the contribution of dry deposition flux to mean total (dry + wet + sea fog)
32 deposition flux for TIN would increase from 11% to 23% (Table 4). However, it is worth

1 noting that the dry deposition fluxes for NH_3 and HNO_3 would decrease as the distance from
2 shore increased since land-derived gaseous inorganic N species are probably transformed to
3 aerosol particles through acid-base reaction and/or absorption on sea-salt particles during their
4 transport over the ocean.

5 Wet deposition of atmospheric inorganic N was highly variable from one event to the next
6 depending on the concentrations of NH_4^+ and NO_3^- in the precipitation as well as the
7 frequency and amount of precipitation. Wet deposition fluxes of atmospheric inorganic N
8 species ranged from 3.5 to 98 (mean 25) $\mu\text{mol m}^{-2} \text{d}^{-1}$ for NH_4^+ and from 1.0 to 32 (mean 8.0)
9 $\mu\text{mol m}^{-2} \text{d}^{-1}$ for NO_3^- , accounting for ~77% by NH_4^+ and ~23% by NO_3^- of TIN from wet
10 deposition flux. While NO_3^- was the dominant inorganic N species in dry deposition,
11 inorganic N supplied to surface waters by atmospheric wet deposition was predominantly by
12 NH_4^+ (72–89% of the wet deposition fluxes for TIN).

13 Likewise wet deposition, sea fog deposition of atmospheric inorganic N was highly variable
14 depending on the size distribution of LWC, the amount of LWC, and the duration of sea fog
15 event as well as the concentrations of NH_4^+ and NO_3^- in the sea fog water. The estimated sea
16 fog deposition fluxes for atmospheric inorganic N species ranged from 0.18–5.2 $\mu\text{mol m}^{-2} \text{d}^{-1}$
17 for NH_4^+ and from 0.13–22 $\mu\text{mol m}^{-2} \text{d}^{-1}$ for NO_3^- , contributing ~39% by NH_4^+ and ~61% by
18 NO_3^- to the sea fog deposition flux for TIN. Mean sea fog deposition fluxes for NH_4^+ and
19 NO_3^- were estimated to be 2.1 $\mu\text{mol m}^{-2} \text{d}^{-1}$ and 5.7 $\mu\text{mol m}^{-2} \text{d}^{-1}$, respectively, indicating
20 that inorganic N supplied to surface waters by sea fog deposition was mainly by NO_3^- , since
21 aerosol NO_3^- and HNO_3 were scavenged more effectively by sea fog.

22 While dry deposition is a continuous process occurring at all times over all surfaces, wet and
23 sea fog deposition are highly episodic. The relative importance of dry, wet, and sea fog
24 deposition obviously varies greatly on short time scales, and varies spatially on longer time
25 scales with global rainfall patterns (Jickells, 2006) and trends in fog frequency (Gultepe et al.,
26 2007). Mean total (dry + wet + sea fog) deposition flux of atmospheric TIN in the subarctic
27 western North Pacific Ocean was estimated to be 46 $\mu\text{mol m}^{-2} \text{d}^{-1}$, with 72% of this in the
28 form of wet deposition (Table 4). This indicates that wet deposition plays an important role in
29 the supply of atmospheric inorganic N to the subarctic western North Pacific Ocean compared
30 to dry and sea fog deposition, although the relative contributions are highly variable. The
31 estimate of the proportion of atmospheric N input via wet deposition were comparable to

1 previously published values: the Pacific 86% (Duce et al., 1991), the Atlantic 78–85% (Baker
2 et al., 2010), and the world oceans 71% (Duce et al., 1991).

3 Fog can lead to substantial N deposition if the event persists long enough (several hours) with
4 sufficient LWC (dense fog), particularly for those formed in continental air masses (Jordan
5 and Talbot, 2000). Although the mean contribution of sea fog deposition to total atmospheric
6 TIN input to the subarctic western North Pacific Ocean was ~17% (Table 4), in some cases,
7 atmospheric TIN deposition flux via sea fog exceeded the combined dry and wet deposition
8 flux of TIN (Fig. 9), suggesting that sea fog can deposit as much N as a high N deposited by
9 rain event, and that sea fog is an important transfer process for atmospheric inorganic N from
10 the marine atmosphere to the subarctic western North Pacific Ocean.

11 **3.8 Potential impact of atmospheric inorganic nitrogen deposition on primary** 12 **production over the subarctic western North Pacific Ocean**

13 The potential impact of atmospheric deposition on marine ecosystems depends on the nutrient
14 status of the receiving waters, and is related to both the total amount and ratio of
15 atmospherically supplied nutrients and to the limiting nutrient for the existing local water
16 column (Baker et al., 2006). It is known that NH_4^+ and NO_3^- can be readily utilized by a
17 variety of aquatic microorganisms (Gilbert et al., 1991). In order to evaluate the impact of
18 atmospheric N on the marine ecosystem, potential primary production was estimated using
19 the result for total deposition flux of TIN and the Redfield C/N ratio of 6.6. The average
20 elemental C/N requirement for many plankton species and for bulk organic matter in the
21 oceans has been found to be relatively constant at ~6.6 (mol/mol) (Redfield, 1958). Assuming
22 that phytoplankton can take up all the N coming from atmospheric deposition with no losses,
23 and that there is no co-limitation by other nutrients (i.e. P and Fe), total mean deposition flux
24 of atmospheric TIN over the subarctic western North Pacific Ocean ($46 \mu\text{mol m}^{-2} \text{d}^{-1}$) was
25 found to be maximally responsible for the carbon uptake of $300 \mu\text{mol C m}^{-2} \text{d}^{-1}$. Elskens et al.
26 (2008) reported that the integrated new primary production in the upper part of the euphotic
27 zone (0–50 m) at station K2 (47°N, 161°E) in the subarctic western North Pacific Ocean from
28 30 July 2005 to 18 August 2005 ranged from $67\text{--}119 \text{ mg C m}^{-2} \text{d}^{-1}$ ($5.6\text{--}9.9 \text{ mmol C m}^{-2} \text{d}^{-1}$).
29 Wong et al. (2002) estimated that the annual new production from the surface (upper 50m) of
30 the subarctic western North Pacific Ocean, which covers the sampling area of this study, to be
31 $32.8\text{--}82.8 \text{ g C m}^{-2} \text{yr}^{-1}$. To facilitate evaluation, we have converted $\text{g C m}^{-2} \text{yr}^{-1}$ units for the
32 annual new production reported by Wong et al. (2002) to the $\mu\text{mol C m}^{-2} \text{d}^{-1}$ units used in this

1 study. Based on these estimates by Elskens et al. (2008) and Wong et al. (2002), the result of
2 this study suggests that inorganic N deposited to the subarctic western North Pacific Ocean
3 from the atmosphere can support 1.6–5.3% of the new primary production. Atmospheric
4 inorganic N deposition, however, could be an important N source in the ocean where sporadic
5 atmospheric N deposition events caused by the transport of the continental dust affect and the
6 supply of deep nutrient-rich water is restricted by the stratification of the surface ocean that is
7 enhanced by global warming.

8

9 **4 Conclusions**

10 The subarctic western North Pacific Ocean ($> 40^{\circ}\text{N}$) has a high frequency of sea fog, with a
11 maximum of $\sim 50\%$ during the summertime period (June–August). The fog deposition is an
12 important transfer process for atmospheric substances from the atmosphere to the biosphere.
13 It is therefore suggested that sea fog may play a key role in supplying atmospheric nutrients to
14 this region. Nevertheless, no study has been carried out over the subarctic western North
15 Pacific Ocean to quantify sea fog deposition flux for atmospheric N. This is the first study to
16 estimate atmospheric inorganic N fluxes via dry, wet, and sea fog deposition simultaneously
17 over the subarctic western North Pacific Ocean. The mean dry, wet, and sea fog deposition
18 fluxes for TIN were estimated to be $4.9 \mu\text{mol m}^{-2} \text{d}^{-1}$, $33 \mu\text{mol m}^{-2} \text{d}^{-1}$, and $7.8 \mu\text{mol m}^{-2} \text{d}^{-1}$,
19 respectively. Wet deposition delivered more atmospheric inorganic N to the subarctic western
20 North Pacific Ocean than dry and sea fog deposition, contributing $\sim 72\%$ to total deposition
21 flux for TIN ($46 \mu\text{mol m}^{-2} \text{d}^{-1}$), although the relative contributions are highly variable.

22 The mean contribution of sea fog deposition to total deposition flux for TIN was $\sim 17\%$.
23 Despite the relatively lower contribution of sea fog deposition, in some cases, atmospheric
24 inorganic N input via sea fog deposition exceeded the combined dry and wet deposition fluxes.
25 Thus, it is suggested that sea fog can result in substantial N deposition if the event persists
26 long enough and has sufficient LWC (dense sea fog), and that ignoring sea fog deposition
27 would lead to underestimate of the total influx of atmospheric inorganic N into the subarctic
28 western North Pacific Ocean, especially in summer periods.

29 A schematic diagram of atmospheric inorganic N input to the subarctic western North Pacific
30 Ocean via dry, wet, and sea fog deposition is shown in Fig. 10. In dry deposition, NO_3^- was
31 the dominant inorganic N species, accounting for $\sim 57\%$. This reflects higher deposition
32 velocity of NO_3^- than that of NH_4^+ since NO_3^- is largely associated with coarse mode particles

1 in the marine atmosphere. In comparison, inorganic N supplied to surface waters by
2 atmospheric wet deposition was predominantly by NH_4^+ (72–89% of the wet deposition
3 fluxes for TIN), suggesting that NH_4^+ is more important inorganic N species supplied by wet
4 deposition over the subarctic western North Pacific Ocean. The contributions of NH_4^+ and
5 NO_3^- to the sea fog deposition flux for TIN were ~39% and ~61%, respectively, indicating
6 that sea fog scavenged more effectively not only coarse mode particles (e.g. sea-salt particles
7 and NaNO_3) that acted as CN of sea fog droplets, but also gaseous HNO_3 .

8 In this study, atmospheric inorganic N input via dry, wet, and sea fog deposition to the
9 subarctic western North Pacific Ocean was estimated by simplified calculation (e.g. using
10 assumption for sea fog water flux) during limited sampling period in the summer, but these
11 results contributed to the understanding of atmospheric N cycle in open ocean environment.
12 Further studies, however, are required to understand the biogeochemical cycles of N more
13 clearly and should focus on long-term monitoring of atmospheric reactive N species,
14 including organic N, in the subarctic western North Pacific Ocean.

15

16 **Acknowledgements**

17 We are grateful to the captains and crews of R/V *Hakuho Maru* for their enthusiastic
18 assistance during Leg 1 of the KH-08-2. This study was supported by funds from the Grant-
19 in-Aid for Scientific Research in Priority Areas “Western Pacific Air-Sea Interaction Study
20 (W-PASS)” under Grant No.18067005 from the Ministry of Education, Culture, Sports,
21 Science and Technology (MEXT), Japan. This work was partly supported by the Sasakawa
22 Scientific Research Grant from The Japan Science Society. This research is a contribution to
23 the Surface Ocean Lower Atmosphere Study (SOLAS) Core Project of the International
24 Geosphere-Biosphere Programme (IGBP). S. Yoon and S. Kim were supported by the Korea
25 Meteorological Administration and Research and Development Program under Grant CATER
26 2012-3020.

27

28 **References**

29 Aikawa, M., Hiraki, T., Suzuki, M., Tamaki, M., and Kasahara, M.: Separate chemical
30 characterizations of fog water, aerosol, and gas before, during, and after fog events near an
31 industrialized area in Japan, *Atmos. Environ.*, 41, 1950–1959, 2007.

1 Akimoto, H.: Global air quality and pollution, *Science*, 302, 1716–1719, doi:
2 10.1126/science.1092666, 2003.

3 Ali, K., Momin, G. A., Tiwari, S., Safai, P. D., Chate, D. M., and Rao, P. S. P.: Fog and
4 precipitation chemistry at Delhi, North India, *Atmos. Environ.*, 38, 4215–4222, 2004.

5 Andreae, M. O. and Crutzen, P. J.: Atmospheric aerosols: biogeochemical sources and role in
6 atmospheric chemistry, *Science*, 276, 1052–1058, doi: 10.1126/science.276.5315.1052, 1997.

7 Andreae, M. O. and Merlet, P.: Emission of trace gases and aerosols from biomass burning,
8 *Global Biogeochem. Cy.*, 15, 4, doi:10.1029/2000GB001382, 2001.

9 Aneja, V. P., Roelle, P. A., Murray, G. C., Southerland, J., Erisman, J. W., Fowler, D., Asman,
10 W. A. H., and Patni, N.: Atmospheric nitrogen compounds II: emissions, transport,
11 transformation, deposition and assessment, *Atmos. Environ.*, 35, 1903–1911, 2001.

12 Baker, A. R., Kelly, S. D., Biswas, K. F., Witt, M., and Jickells, T. D.: Atmospheric
13 deposition of nutrients to the Atlantic Ocean, *Geophys. Res. Lett.*, 30, 2296,
14 doi:10.1029/2003GL018518, 2003.

15 Baker, A. R., Jickells, T. D., Biswas, K. F., Weston, K., and French, M.: Nutrient in
16 atmospheric aerosol particles along the Atlantic Meridional Transect, *Deep-Sea Res. Pt. II*, 53,
17 1706–1719, 2006.

18 Baker, A. R., Weston, K., Kelly, S. D., Voss, M., Streu, P., and Cape, J. N.: Dry and wet
19 deposition of nutrients from the tropical Atlantic atmosphere: Links to primary productivity
20 and nitrogen fixation, *Deep-Sea Res. Pt. I*, 54, 1704–1720, 2007.

21 Baker, A. R., Lesworth, T., Adams, C., Jickells, T. D., and Ganzeveld, L.: Estimation of
22 atmospheric nutrient inputs to the Atlantic Ocean from 50°N to 50°S based on large-scale
23 field sampling: Fixed nitrogen and dry deposition of phosphorus, *Global Biogeochem. Cy.*, 24,
24 GB3006, doi:10.1029/2009GB003634, 2010.

25 Bardouki, H., Berresheim, H., Vrekoussis, M., Sciare, J., Kouvarakis, G., Oikonomou, K.,
26 Schneider, J., and Mihalopoulos, N.: Gaseous (DMS, MSA, SO₂, H₂SO₄ and DMSO) and
27 particulate (sulfate and methanesulfonate) sulfur species over the northeastern coast of Crete,
28 *Atmos. Chem. Phys.*, 3, 1871–1886, 2003.

29 Burkard, R., Eugster, W., Wrzesinsky, T., and Klemm, O.: Vertical divergence of fogwater
30 fluxes above a spruce forest, *Atmos. Res.*, 64, 133–145, 2002.

1 Charlson, R. J., Lovelock, J. E., Andreae, M. O., and Warren, S. G.: Oceanic phytoplankton,
2 atmospheric sulfur, cloud albedo and climate, *Nature*, 326, 655–661, 1987.

3 Chen, Y. and Siefert, R. L.: Seasonal and spatial distributions and dry deposition fluxes of
4 atmospheric total and labile iron over the tropical and subtropical North Atlantic Ocean, *J.*
5 *Geophys. Res.*, 109, D09305, doi:10.1029/2003JD003958, 2004.

6 Chiapello, I., Bergametti, G., Chatenet, B., Bousquet, P., Dulac, F., and Soares, E. S.: Origins
7 of African dust transported over the northeastern tropical Atlantic, *J. Geophys. Res.*, 102, D12,
8 doi:10.1029/97JD00259, 1997.

9 Cho, Y. -K., Kim, M. -O., and Kim, B. -C.: Sea fog around the Korean Peninsula, *J. Appl.*
10 *Meteorol.*, 39, 2473–2479, 2000.

11 Collett Jr, J. L., Sherman, D. E., Moore, K. F., Hannigan, M. P., and Lee, T.: Aerosol particle
12 processing and removal by fogs: observations in chemically heterogeneous central California
13 radiation fogs, *Water Air Soil Poll. Focus*, 1, 303–312, 2001.

14 Collett Jr, J. L., Bator, A., Sherman, D. E., Moore, K. F., Hoag, K. J., Demoz, B. B., Rao, X.,
15 and Reilly, J. E.: The chemical composition of fogs and intercepted clouds in the United
16 States, *Atmos. Res.*, 64, 29–40, 2002.

17 Deboudt, K., Flament, P., and Bertho, M. L.: Cd, Cu, Pb and Zn concentrations in
18 atmospheric wet deposition at a coastal station in Western Europe, *Water Air Soil Poll.*, 151,
19 335–359, 2004.

20 Dentener, F., Drevet, J., Lamarque, J. F., Bey, I., Eickhout, B., Fiore, A. M., Hauglustaine, D.,
21 Horowitz, L. W., Krol, M., Kulshrestha, U. C., Lawrence, M., Galy-Lacaux, C., Rast, S.,
22 Shindell, D., Stevenson, D., Van Noije, T., Atherton, C., Bell, N., Bergman, D., Butler, T.,
23 Cofala, J., Collins, B., Doherty, R., Ellingsen, K., Galloway, J., Gauss, M., Montanaro, V.,
24 Müller, J. F., Pitari, G., Rodriguez, J., Sanderson, M., Solmon, F., Strahan, S., Schultz, M.,
25 Sudo, K., Szopa, S., and Wild, O.: Nitrogen and sulfur deposition on regional and global
26 scales: A multimodel evaluation, *Global Biogeochem. Cy.*, 20, GB4003,
27 doi:10.1029/2005GB002672, 2006.

28 Duce, R. A., Liss, P. S., Merrill, J. T., Atlas, E. L., Buat-Menard, P., Hicks, B. B., Miller, J.
29 M., Prospero, J. M., Arimoto, R., Church, T. M., Ellis, W., Galloway, J. N., Hansen, L.,
30 Jickells, T. D., Knap, A. H., Reinhardt, K. H., Schneider, B., Soudine, A., Tokos, J. J.,

- 1 Tsunogai, S., Wollast, R., and Zhou, M.: The atmospheric input of trace species to the world
2 ocean, *Global Biogeochem. Cy.*, 5, 193–259, 1991.
- 3 Duce, R. A., LaRoche, J., Altieri, K., Arrigo, K. R., Baker, A. R., Capone, D. G., Cornell, S.,
4 Dentener, F., Galloway, J., Ganeshram, R. S., Geider, R. J., Jickells, T., Kuypers, M. M.,
5 Langlois, R., Liss, P. S., Liu, S. M., Middelburg, J. J., Moore, C. M., Nickovic, S., Oschlies,
6 A., Pedersen, T., Prospero, J., Schlitzer, R., Seitzinger, S., Sorensen, L. L., Uematsu, M.,
7 Ulloa, O., Voss, M., Ward, B., and Zamora, L.: Impacts of Atmospheric Anthropogenic
8 Nitrogen on the Open Ocean, *Science*, 320, 893–897, doi: 10.1126/science.1150369, 2008.
- 9 Eugster, W.: Fog research, *Die Erde*, 139, 1–10, 2008.
- 10 Elskens, M., Brion, N., Buesseler, K., Van Mooy, B. A. S., Boyd, P., Dehairs, F., Savoye, N.,
11 and Baeyens, W.: Primary, new and export production in the NW Pacific subarctic gyre
12 during the vertigo K2 experiments, *Deep-Sea Res. Pt. II*, 55, 1594–1604, 2008.
- 13 Fahey, K. M., Pandis, S. N., Collett Jr, J. L., and Herckes, P.: The influence of size-dependent
14 droplet composition on pollutant processing by fogs, *Atmos. Environ.*, 39, 4561–4574, 2005.
- 15 Fu, G., Guo, J., Xie, S. -P., Duan, Y., and Zhang, M.: Analysis and high-resolution modeling
16 of a dense sea fog event over the Yellow Sea, *Atmos. Res.*, 81, 293–303, 2006.
- 17 Galloway, J. N., Dentener, F. J., Capone, D. G., Boyer, E. W., Howarth, R. W., Seitzinger, S.
18 P., Asner, G. P., Cleveland, C. C., Green, P. A., Holland, E. A., Karl, D. M., Michaels, A. F.,
19 Porter, J. H., Townsend, A. R., and Voeroesmary, C. J.: Nitrogen cycles: past, present, and
20 future, *Biogeochemistry*, 70(2), 153–226, 2004.
- 21 Galloway, J. N., Townsend, A. R., Erisman, J. W., Bekunda, M., Cai, Z., Freney, J. R.,
22 Martinelli, L. A., Seitzinger, S. P., and Sutton, M. A.: Transformation of the Nitrogen Cycle:
23 Recent Trends, Questions, and Potential Solutions, *Science*, 320, 889–892, doi:
24 10.1126/science.1136674, 2008.
- 25 Gao, Y., Arimoto, R., Zhou, M. Y., Merrill, J. T., and Duce, R. A.: Relationships between the
26 dust concentrations over eastern Asia and the remote North Pacific, *J. Geophys. Res.*, 97(D9),
27 9867–9872, doi:10.1029/92JD00714, 1992.
- 28 Gilbert, P. M., Garside, C., Fuhrman, J. A., and Roman, M. R.: Time-dependent coupling of
29 inorganic and organic nitrogen uptake and regeneration in the plume of the Chesapeake Bay
30 estuary and its regulation by large heterotrophs, *Limnol. Oceanogr.*, 36, 895–909, 1991.

1 Gioda, A., Reyes-Rodríguez, G. J., Santos-Figueroa, G., Collett Jr, J. L., Decesari, S., Ramos,
2 M. d. C. K. V, Bezerra Netto, H. J. C., de Aquino Neto, F. R., and Mayol-Bracero, O. L.:
3 Speciation of water-soluble inorganic, organic, and total nitrogen in a background marine
4 environment: Cloud water, rainwater, and aerosol particles, *J. Geophys. Res.*, 116, D05203,
5 doi:10.1029/2010JD015010, 2011.

6 Gondwe, M., Krol, M., Gieskes, W., Klaassen, W., and de Baar, H.: The contribution of
7 ocean-leaving DMS to the global atmospheric burdens of DMS, MSA, SO₂, and NSS SO₄²⁻,
8 *Global Biogeochem. Cy.*, 17, 1056, doi:10.1029/2002GB001937, 2003.

9 Graedel, T. E. and Keene, W. C.: Tropospheric budget of reactive chlorine, *Global*
10 *Biogeochem. Cy.*, 9(1), 47–77, doi:10.1029/94GB03103, 1995.

11 Gultepe, I., Tardif, R., Michaelides, S. C., Cermak, J., Bott, A., Bendix, J., Müller, M. D.,
12 Pagowski, M., Hansen, B., Ellrod, G., Jacobs, W., Toth, G., and Cober, S. G.: Fog research: A
13 review of past achievements and future perspectives, *Pure appl. geophys.*, 164, 1121–1159,
14 2007.

15 Herckes, P., Chang, H., Lee, T., and Collett Jr, J. L.: Air pollution processing by radiation
16 fogs, *Water Air Soil Poll.*, 181, 65–75, 2007.

17 Jacob, D. J., Waldman, J. M., Munger, W., and Hoffmann, M. R.: A field investigation of
18 physical and chemical mechanisms affecting pollutant concentrations in fog droplets, *Tellus B*,
19 36, 272–285, 1984.

20 Jickells, T.: Atmospheric inputs of metals and nutrients to the oceans: their magnitude and
21 effects, *Mar. Chem.*, 48, 199–214, 1995.

22 Jickells, T.: The role of air-sea exchange in the marine nitrogen cycle, *Biogeosciences*, 3,
23 271–280, 2006.

24 Jickells, T. D., Kelly, S. D., Baker, A. R., Biswas, K., Dennis, P. F., Spokes, L. J., Witt, M.,
25 and Yeatman, S. G.: Isotopic evidence for a marine ammonia source, *Geophys. Res. Lett.*, 30,
26 1374, doi:10.1029/2002GL016728, 2003.

27 Jordan, C. E. and Talbot, R. W.: Direct atmospheric deposition of water-soluble nitrogen to
28 the Gulf of Maine, *Global Biogeochem. Cy.*, 14, 4, doi:10.1029/2000GB001266, 2000.

- 1 Jung, J., Furutani, H., and Uematsu, M.: Atmospheric inorganic nitrogen in marine aerosol
2 and precipitation and its deposition to the North and South Pacific Oceans, *J. Atmos. Chem.*,
3 68, 157–181, 2011.
- 4 Keene, W. C., Pszenny, A. A. P., Galloway, J. N., and Hawley, M. E.: Sea-salt corrections
5 and interpretation of constituent ratios in marine precipitation, *J. Geophys. Res.*, 91(D6),
6 6647–6658, doi:10.1029/JD091iD06p06647, 1986.
- 7 Kim, T. -W., Lee, K., Najjar, R. G., Jeong, H. -D., and Jeong, H. J.: Increasing N Abundance
8 in the Northwestern Pacific Ocean Due to Atmospheric Nitrogen Deposition, *Science*, 334,
9 505–509, doi: 10.1126/science.1206583, 2011.
- 10 Klemm, O. and Wrzesinsky, T.: Fog deposition fluxes of water and ions to a mountainous site
11 in Central Europe, *Tellus B*, 59, 705–714, 2007.
- 12 Klemm, O., Wrzesinsky, T., and Scheer, C.: Fog water flux at a canopy top: Direct
13 measurement versus one-dimensional model, *Atmos. Environ.*, 39, 5375–5386, 2005.
- 14 Krishnamurthy, A., Moore, J. K., Mahowald, N., Luo, C., and Zender, C. S.: Impacts of
15 atmospheric nutrient inputs on marine biogeochemistry, *J. Geophys. Res.*, 115, G01006,
16 doi:10.1029/2009JG001115, 2010.
- 17 Lange, C. A., Matschullat, J., Zimmermann, F., Sterzik, G., and Wienhaus, O.: Fog frequency
18 and chemical composition of fog water—a relevant contribution to atmospheric deposition in
19 the eastern Erzgebirge, Germany, *Atmos. Environ.*, 37, 3731–3739, 2003.
- 20 Larsen, J., Neal, C., Webley, P., Freymueller, J., Haney, M., McNutt, S., Schneider, D.,
21 Prejean, S., Schaefer, J., and Wessels, R.: Eruption of Alaska volcano breaks historic pattern,
22 *Eos Trans. AGU*, 90(20), 173, doi:10.1029/2009EO200001, 2009.
- 23 Lewis, J. M., Koračin, D., and Redmond, K. T.: Sea fog research in the United Kingdom and
24 United States: A historical essay including outlook, *B. Am. Meteorol. Soc.*, 85, 395–408,
25 2004.
- 26 Li, P., Li, X., Yang, C., Wang, X., Chen, J., and Collett Jr, J. L.: Fog water chemistry in
27 Shanghai, *Atmos. Environ.*, 45, 4034–4041, 2011.
- 28 Loo, B. W. and Cork, C. P.: Development of High Efficiency Virtual Impactors, *Aerosol Sci.*
29 *Technol.*, 9, 167–176, 1988.

- 1 Lovett, G. M., Reiners, W. A., and Olson, R. K.: Cloud Droplet Deposition in Subalpine
2 Balsam Fir Forests: Hydrological and Chemical Inputs, *Science*, 218, 1303–1304, 1982.
- 3 Lu, C., Niu, S., Tang, L., Lv, J., Zhao, L., and Zhu, B.: Chemical composition of fog water in
4 Nanjing area of China and its related fog microphysics, *Atmos. Res.*, 97, 47–69, 2010.
- 5 Lu, Z. and Dzurisin, D.: Ground surface deformation patterns, magma supply, and magma
6 storage at Okmok volcano, Alaska, from InSAR analysis: 2. Coeruptive deflation, July–
7 August 2008, *J. Geophys. Res.*, 115, B00B03, doi:10.1029/2009JB006970, 2010.
- 8 Matsumoto, K., Tominaga, S., and Igawa, M.: Measurements of atmospheric aerosols with
9 diameters greater than 10 μm and their contribution to fixed nitrogen deposition in coastal
10 urban environment, *Atmos. Environ.*, 45, 6433–6438, 2011.
- 11 Millet, M., Sanusi, A., and Wortham, H.: Chemical composition of fogwater in an urban area:
12 Strasbourg (France), *Environ. Pollut.*, 94, 345–354, 1996.
- 13 Minami, Y. and Ishizaka, Y.: Evaluation of chemical composition in fog water near the
14 summit of a high mountain in Japan, *Atmos. Environ.*, 30, 3363–3376, 1996.
- 15 Moore, K. F., Sherman, D. E., Reilly, J. E., and Collett Jr, J. L.: Drop size-dependent
16 chemical composition in clouds and fogs. Part I. Observations, *Atmos. Environ.*, 38, 1389–
17 1402, 2004.
- 18 Nakamura, T., Matsumoto, K., and Uematsu, M.: Chemical characteristics of aerosols
19 transported from Asia to the East China Sea: an evaluation of anthropogenic combined
20 nitrogen deposition in autumn, *Atmos. Environ.*, 39, 1749–1758, 2005.
- 21 Paerl, H. W.: Coastal eutrophication and harmful algal blooms: importance of atmospheric
22 deposition and groundwater as "new" nitrogen and other nutrient sources, *Limnol. Oceanogr.*,
23 42, 1154–1165, 1997.
- 24 Pandis, S. N. and Seinfeld, J. H.: The smog-fog-smog cycle and acid deposition, *J. Geophys.*
25 *Res.*, 95, D11, doi:10.1029/JD095iD11p18489, 1990.
- 26 Prospero, J. M., Barrett, K., Church, T., Dentener, F., Duce, R. A., Galloway, J. N., Levy II,
27 H., , Moody, J., and Quinn, P.: Atmospheric deposition of nutrients to the North Atlantic
28 basin, *Biogeochemistry*, 35, 27–73, 1996.
- 29 Quinn, P. K., Charlson, R. J., and Zoller, W. H.: Ammonia, the dominant base in the remote
30 marine troposphere: a review, *Tellus B*, 39B, 413–425, 1987.

1 Raja, S., Raghunathan, R., Yu, X. -Y., Lee, T., Chen, J., Kommalapati, R. R., Murugesan, K.,
2 Shen, X., Yuan, Q., Valsaraj, K. T., and Collett Jr, J. L.: Fog chemistry in the Texas–
3 Louisiana Gulf Coast corridor, *Atmos. Environ.*, 42, 2048–2061, 2008.

4 Redfield, A. C.: The biological control of chemical factors in the environment, *Am. Sci.*, 46,
5 205– 221, 1958.

6 Sasakawa, M. and Uematsu, M.: Chemical composition of aerosol, sea fog, and rainwater in
7 the marine boundary layer of the northwestern North Pacific and its marginal seas, *J. Geophys.*
8 *Res.*, 107(D24), 4783, doi:10.1029/2001JD001004, 2002.

9 Sasakawa, M. and Uematsu, M.: Relative contribution of chemical composition to
10 acidification of sea fog (stratus) over the northern North Pacific and its marginal seas, *Atmos.*
11 *Environ.*, 39, 1357–1362, 2005.

12 Sasakawa, M., Ooki, A., and Uematsu, M.: Aerosol size distribution during sea fog and its
13 scavenge process of chemical substances over the northwestern North Pacific, *J. Geophys.*
14 *Res.*, 108(D3), 4120, doi:10.1029/2002JD002329, 2003.

15 Schmale, J., Schneider, J., Jurkat, T., Voigt, C., Kalesse, H., Rautenhaus, M., Lichtenstern, M.,
16 Schlager, H., Ancellet, G., Arnold, F., Gerding, M., Mattis, I., Wendisch, M., and Borrmann,
17 S.: Aerosol layers from the 2008 eruptions of Mount Okmok and Mount Kasatochi: In situ
18 upper troposphere and lower stratosphere measurements of sulfate and organics over Europe,
19 *J. Geophys. Res.*, 115, D00L07, doi:10.1029/2009JD013628, 2010.

20 Spiegel, J. K., Zieger, P., Bukowiecki, N., Hammer, E., Weingartner, E., and Eugster, W.:
21 Evaluating the capabilities and uncertainties of droplet measurements for the fog droplet
22 spectrometer (FM-100), *Atmos. Meas. Tech.*, 5, 2237–2260, 2012.

23 Spokes, L. J., Yeatman, S. G., Cornell, S. E., and Jickells, T. D.: Nitrogen deposition to the
24 eastern Atlantic Ocean. The importance of south-easterly flow, *Tellus B*, 52, 37–49, 2000.

25 Thalmann, E., Burkard, R., Wrzesinsky, T., Eugster, W., and Klemm, O.: Ion fluxes from fog
26 and rain to an agricultural and a forest ecosystem in Europe, *Atmos. Res.*, 64, 147–158, 2002.

27 Tokinaga, H. and Xie, S. -P.: Ocean tidal cooling effect on summer sea fog over the Okhotsk
28 Sea, *J. Geophys. Res.*, 114, D14102, doi:10.1029/2008JD011477, 2009.

1 Uematsu, M., Duce, R. A., Prospero, J. M., Chen, L., Merrill, J. T., and McDonald, R. L.:
2 Transport of mineral aerosol from Asia over the North Pacific Ocean, *J. Geophys. Res.*,
3 88(C9), 5343–5352, doi:10.1029/JC088iC09p05343, 1983.

4 Uematsu, M., Hattori, H., Nakamura, T., Narita, Y., Jung, J., Matsumoto, K., Nakaguchi, Y.,
5 and Dileep Kumar, M.: Atmospheric transport and deposition of anthropogenic substances
6 from the Asia to the East China Sea, *Mar. Chem.*, 120, 108–115, 2010.

7 Uno, I., Uematsu, M., Hara, Y., He, Y. J., Ohara, T., Mori, A., Kamaya, T., Murano, K.,
8 Sadanaga, Y., and Bandow, H.: Numerical study of the atmospheric input of anthropogenic
9 total nitrate to the marginal seas in the western North Pacific region, *Geophys. Res. Lett.*, 34,
10 L17817, doi:10.1029/2007GL030338, 2007.

11 Wang, B. -H.: Distributions and variations of sea fog in the world, in: *Sea Fog*, China Ocean
12 Press, Beijing, 51–90, 1985.

13 Wong, C. S., Waser, N. A. D., Nojiri, Y., Whitney, F. A., Page, J. S., and Zeng, J.: Seasonal
14 cycles of nutrients and dissolved inorganic carbon at high and mid latitudes in the North
15 Pacific Ocean during the Skaugran cruises: determination of new production and nutrient
16 uptake ratios, *Deep-Sea Res. Pt. II*, 49, 5317–5338, 2002.

17 Xie, P. and Arkin, P. A.: Global precipitation: A 17-year monthly analysis based on gauge
18 observations, satellite estimates, and numerical model outputs, *B. Am. Meteorol. Soc.*, 78,
19 2539–2558, 1997.

20 Zhang, Q. and Anastasio, C.: Chemistry of fog waters in California's Central Valley—Part 3.
21 Concentrations and speciation of organic and inorganic nitrogen, *Atmos. Environ.*, 35, 5629–
22 5643, 2001.

23 Zhang, Y., Yu, Q., Ma, W., and Chen, L.: Atmospheric deposition of inorganic nitrogen to the
24 eastern China seas and its implications to marine biogeochemistry, *J. Geophys. Res.*, 115,
25 D00K10, doi:10.1029/2009JD012814, 2010.

1 Table 1. Mean, 25th, 50th, and 75th percentile concentrations of major ionic species in aerosols (n = 11), rainwater (n = 7), and sea fog water
 2 (n = 15) collected over the subarctic western North Pacific during the cruise^a.

	Aerosol (neq m ⁻³)					Rain (μeq l ⁻¹)					Sea fog (μeq l ⁻¹)				
	Mean	25th ^b	50th ^b	75th ^b	n ^c	Mean	25th ^b	50th ^b	75th ^b	n ^c	Mean	25th ^b	50th ^b	75th ^b	n ^c
pH	-	-	-	-	-	4.1	3.7	4.1	4.3	5	4.2	3.6	4.1	4.4	15
Na ⁺	33	13	24	47	11	580	52	220	1300	7	390	78	190	400	15
NH ₄ ⁺	5.6	4.1	5.4	6.5	11	25	8.4	18	41	7	22	8.5	22	33	15
K ⁺	0.59	0.24	0.49	0.72	11	19	3.0	7.1	32	7	9.3	3.0	5.8	9.0	15
Mg ²⁺	5.9	2.4	5.0	9.0	11	220	14	51	530	7	83	16	42	85	15
Ca ²⁺	1.8	0.95	1.4	2.4	11	400	46	150	570	7	20	5.8	10	22	15
Cl ⁻	28	7.0	21	39	11	1100	130	540	2500	7	400	80	190	420	15
NO ₃ ⁻	2.5	1.3	2.3	3.1	11	7.9	1.6	5.4	13	7	50	7.6	51	75	15
SO ₄ ²⁻	22	13	22	29	11	66	8.9	29	170	7	120	63	100	160	15
MSA	0.62	0.24	0.43	0.67	11	0.42	0.10	0.18	0.86	6	6.2	0.93	3.2	9.5	15
nss-SO ₄ ²⁻	18	9.9	17	27	11	5.5	1.9	4.9	15	5	72	39	50	100	14
nss-K ⁺	0.068	0.018	0.024	0.091	9	7.0	1.9	2.7	10	7	1.7	0.80	1.1	2.7	14

nss-Ca ²⁺	0.66	0.29	0.41	0.77	11	380	44	140	560	7	4.6	0.079	2.6	9.1	12
nss-Mg ²⁺	0.12	0	0	0	2	95	15	59	290	5	1.1	0	0	1.3	5
nss-Cl ⁻	-	-	-	-	-	420	20	190	840	7	-	-	-	-	-

1 ^aNegative values that arise for non sea-salt ionic species as a result of analytical uncertainty and samples where concentration of each ionic
2 component was below the detection limit have been included in the calculation of the average as 0.

3 ^bThese values indicate percentiles.

4 ^cSample number of each ionic component detected (or calculated) in aerosols, rainwater, and sea fog water.

1 Table 2. Mole equivalent ratios for major ionic species in aerosols, rainwater, and sea fog
 2 water, together with seawater ratios for comparison^a.

	Aerosol	Rain	Sea fog	Seawater ^b
Cl^-/Na^+	0.79	2.7	1.0	1.17
$\text{Mg}^{2+}/\text{Na}^+$	0.19	0.42	0.19	0.22
K^+/Na^+	0.021	0.051	0.031	0.021
$\text{Ca}^{2+}/\text{Na}^+$	0.073	1.2	0.069	0.044
$\text{SO}_4^{2-}/\text{Na}^+$	1.4	0.15	0.92	0.12
nss- $\text{SO}_4^{2-}/\text{Na}^+$	1.3	0.047	0.86	-
$\text{NO}_3^-/\text{Na}^+$	0.099	0.029	0.43	-
$\text{NH}_4^+/\text{Na}^+$	0.34	0.10	0.16	-
$\text{NO}_3^-/\text{nss-SO}_4^{2-}$	0.19	0.81	0.57	-
$\text{NH}_4^+/\text{nss-Ca}^{2+}$	12	0.14	28	-
$\text{NH}_4^+/\text{nss-Mg}^{2+}$	17	0.82	11	-

3 ^aThe samples with negative value of non sea-salt ionic component were excluded.

4 ^bSeawater ratios from Keene et al. (1986).

5

1 Table 3. Sea fog water/rainwater ratios for major ionic species in terms of concentrations (μeq
 2 L^{-1}).

Sea fog water/Rainwater	
Na^+	0.67
NH_4^+	0.88
ss- K^+	0.63
ss- Mg^{2+}	0.66
ss- Ca^{2+}	0.77
ss- Cl^-	0.59
NO_3^-	6.3
ss- SO_4^{2-}	0.79
MSA	15
nss- SO_4^{2-}	13
nss- K^+	0.24
nss- Ca^{2+}	0.012
nss- Mg^{2+}	0.012

3

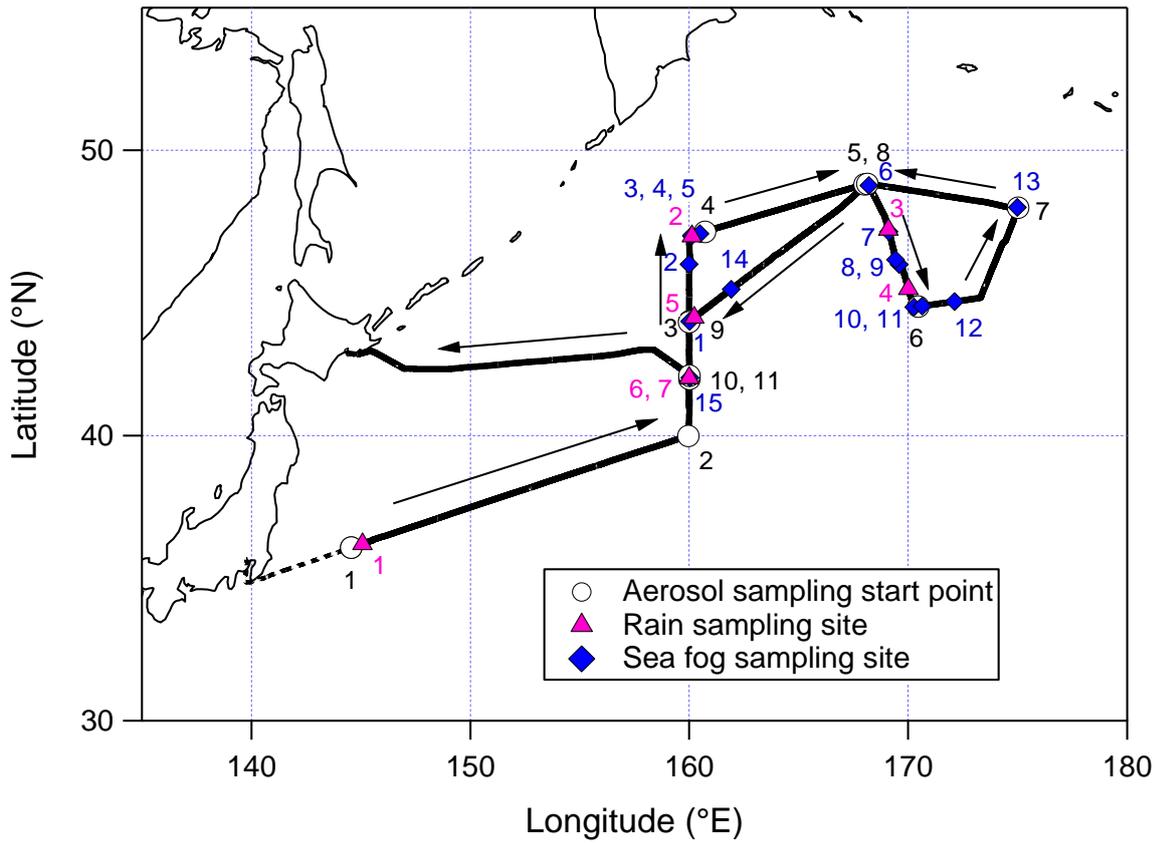
1 Table 4. Mean dry, wet, sea fog, and total deposition fluxes of NH_4^+ and NO_3^- , and the
 2 contribution of each deposition to total inorganic N input in the subarctic western North
 3 Pacific Ocean during the cruise.

	Dry deposition ($\mu\text{mol m}^{-2} \text{d}^{-1}$)	Wet deposition ($\mu\text{mol m}^{-2} \text{d}^{-1}$)	Sea fog deposition ($\mu\text{mol m}^{-2} \text{d}^{-1}$)	Total deposition ($\mu\text{mol m}^{-2} \text{d}^{-1}$)
NH_4^+	1.9 (4.1%)	25 (54%)	2.1 (4.6%)	29 (63%)
NO_3^-	3.0 (6.5%)	8.0 (17%)	5.7 (12%)	17 (37%)
TIN ^a	4.9 (11%)	33 (72%)	7.8 (17%)	46 (100%)
TIN ^b	12 (23%)	33 (62%)	7.8 (15%)	53 (100%)

4 ^aTIN represents total inorganic nitrogen. In this study, total inorganic nitrogen is defined as
 5 including NH_4^+ and NO_3^- ; i.e. $\text{TIN} = \text{NH}_4^+ + \text{NO}_3^-$.

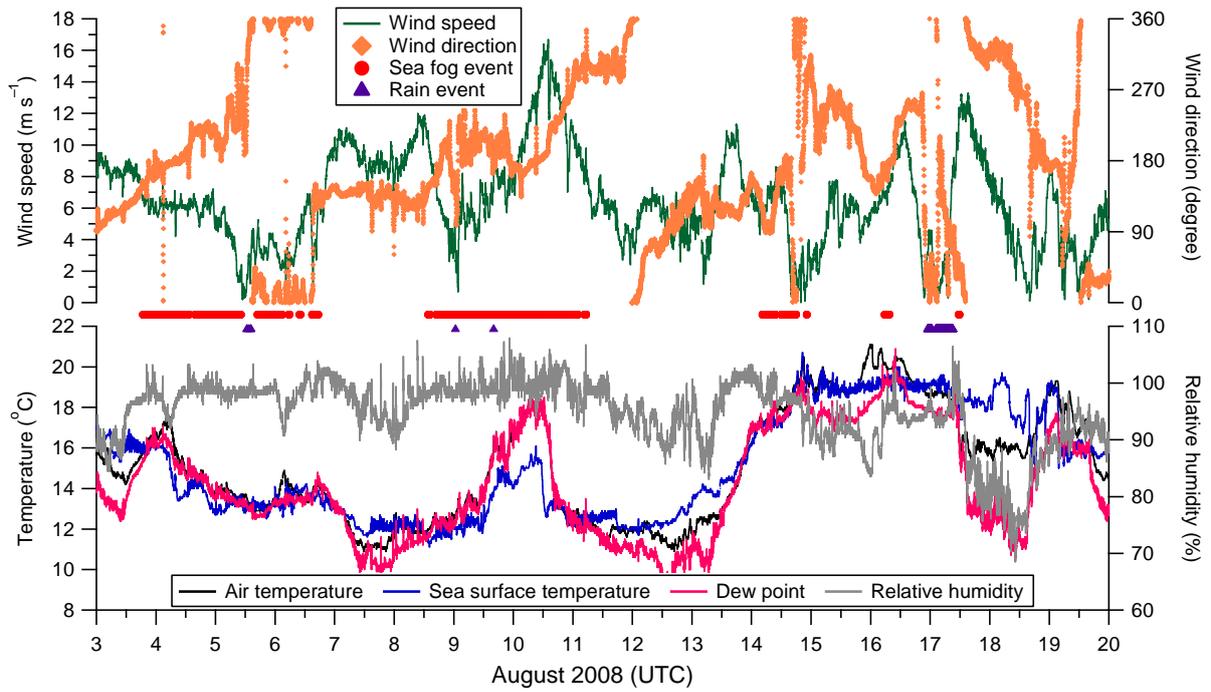
6 ^bThe mean dry, wet, and sea fog deposition fluxes for TIN and the contribution of each
 7 deposition to TIN input when the dry deposition fluxes for gaseous NH_3 ($2.0 \mu\text{mol m}^{-2} \text{d}^{-1}$)
 8 and HNO_3 ($4.7 \mu\text{mol m}^{-2} \text{d}^{-1}$) reported by Zhang et al. (2010) are included.

9



1
2
3
4
5
6
7

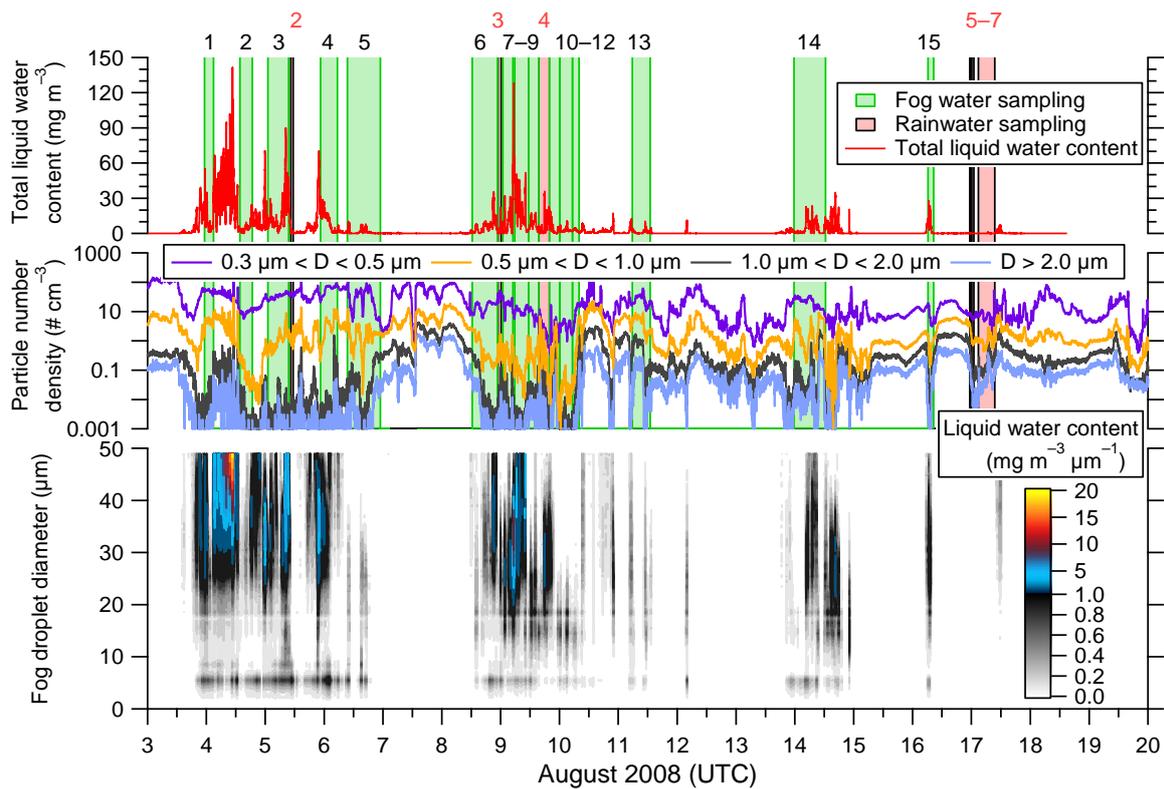
Figure 1. Cruise track of Leg 1 of the KH-08-2. White circles, pink triangles, and blue diamonds indicate aerosol, rain, and sea fog sampling locations during the cruise, respectively. Each aerosol sampling start point represents the end of the previous sampling period. Dotted line indicates that no aerosol sampling was conducted. Black, pink, and blue numbers represent the sample numbers of aerosol, rain, and sea fog, respectively.



1

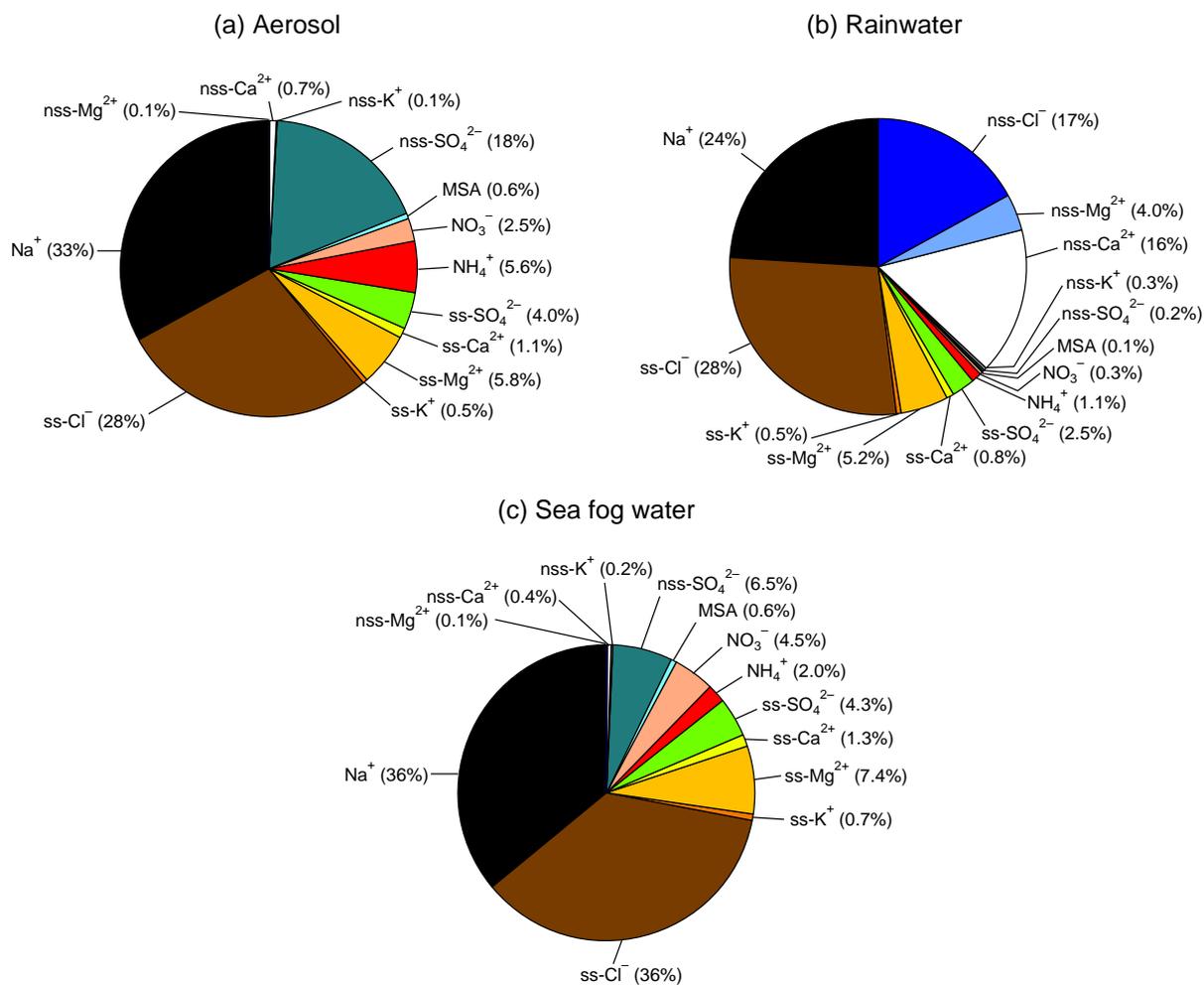
2 Figure 2. Temporal variations of meteorological variables (i.e. wind speed, wind direction, air
 3 temperature, sea surface temperature, dew point, and relative humidity) during the cruise. Sea
 4 fog (red circle) and rain (violet triangle) events indicate the occurrences of observed sea fog
 5 and rain events, respectively.

6

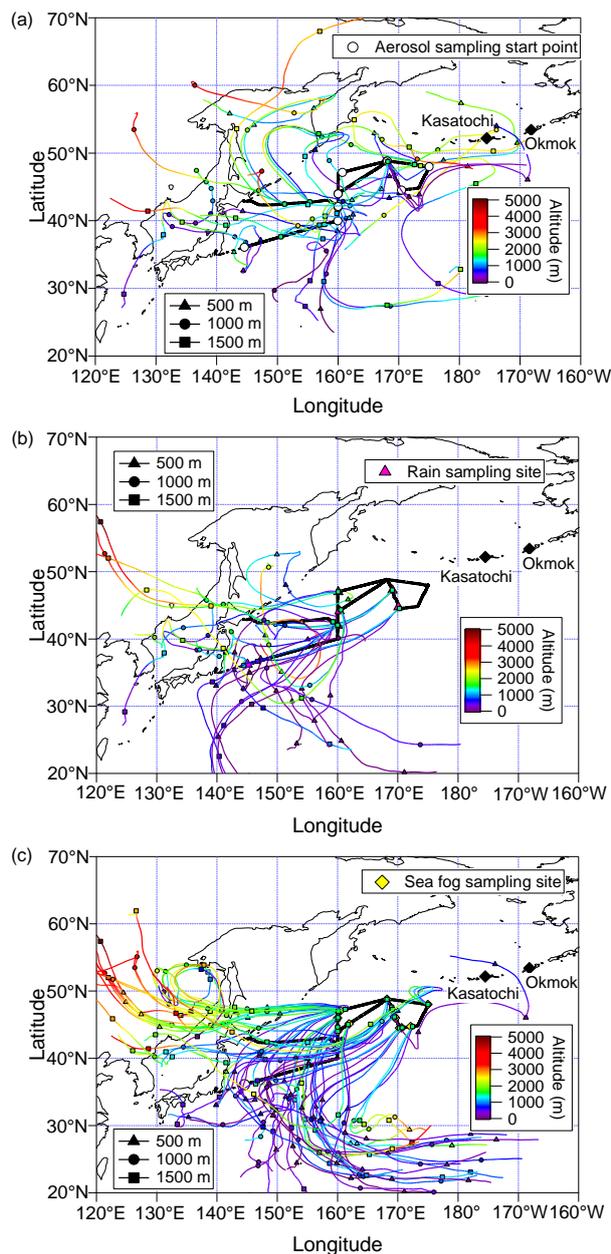


1
2
3
4
5
6
7
8
9

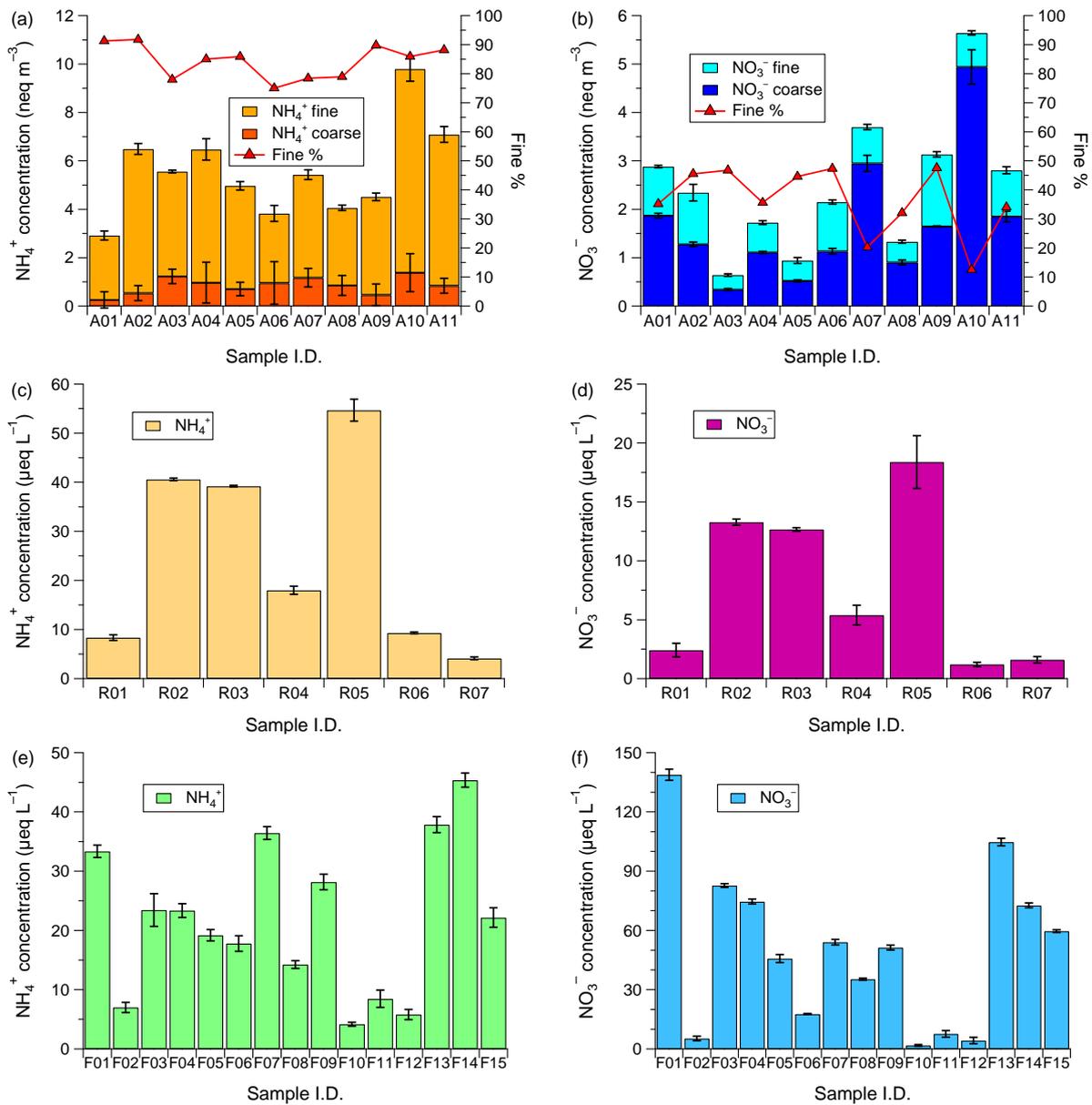
Figure 3. Temporal variations of total LWC, particle number densities for aerosols in four size groups, and LWC for each of the 40 droplet size classes during the cruise. The green and pink shaded areas indicate sea fog water and rainwater sampling durations, respectively. The black and pink numbers indicate sea fog and rainwater sample number, respectively. When sea fog and rain events occurred simultaneously, only rainwater sample was collected. Then that sample was considered as a rainwater sample (i.e. rainwater sample number 3 and 4). Since rainwater sample number 1 was collected on 30 July, it was not presented here.



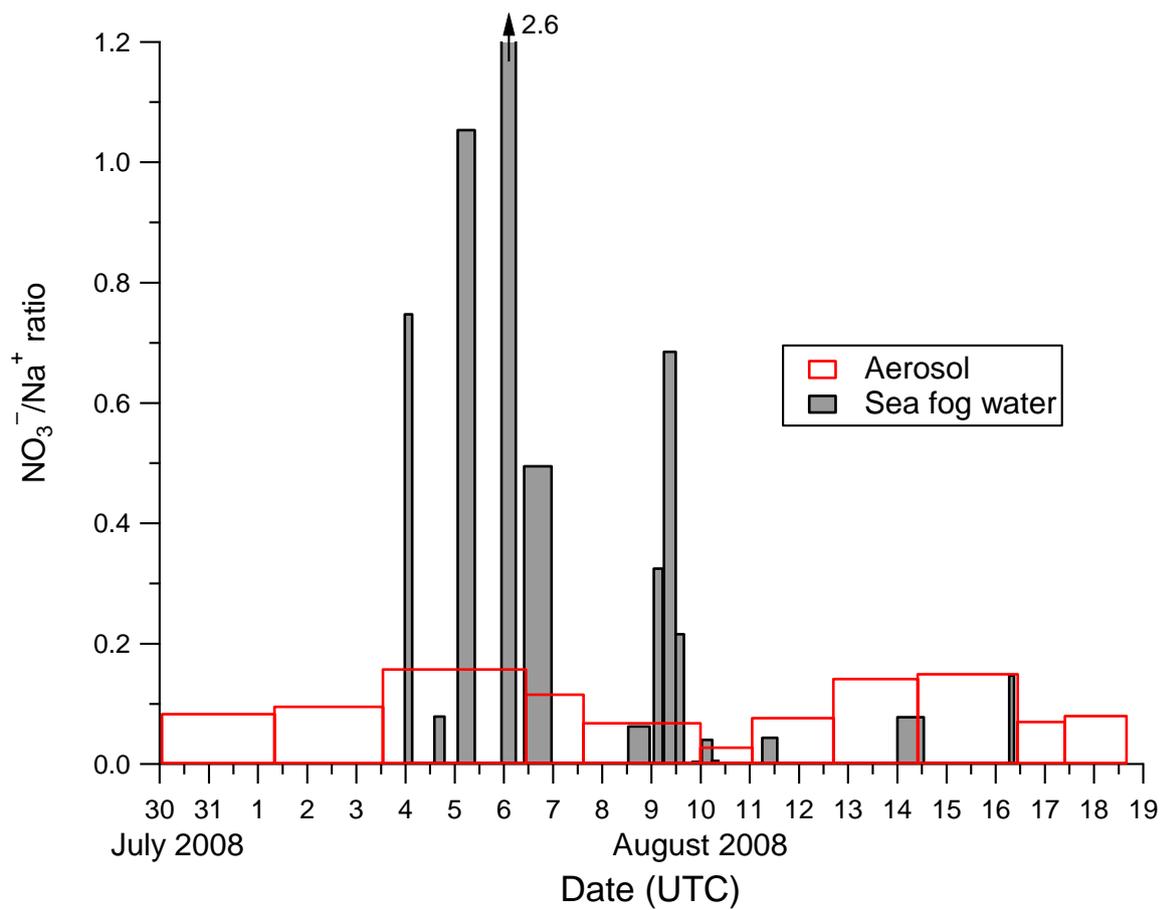
1
 2 Figure 4. Mean contributions of each major ionic component to total ionic concentration in (a)
 3 aerosols (n = 11), (b) rainwater (n = 7), and (c) sea fog water (n = 15) collected over the
 4 subarctic western North Pacific Ocean during the cruise.
 5



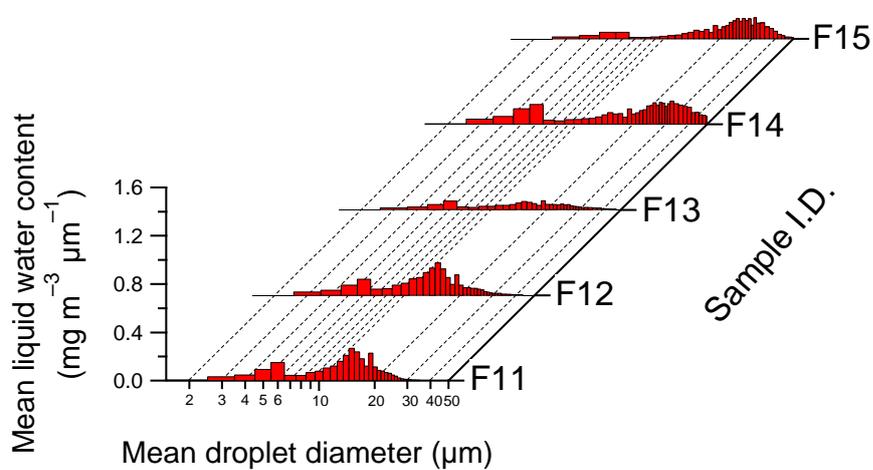
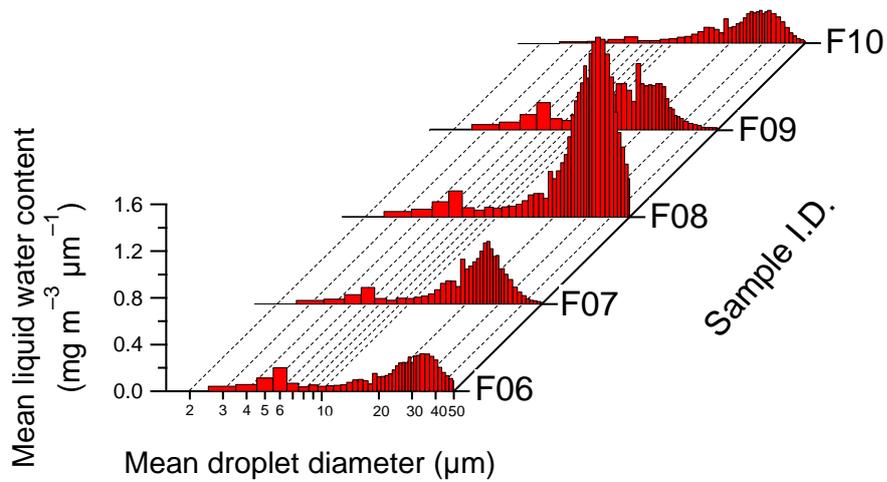
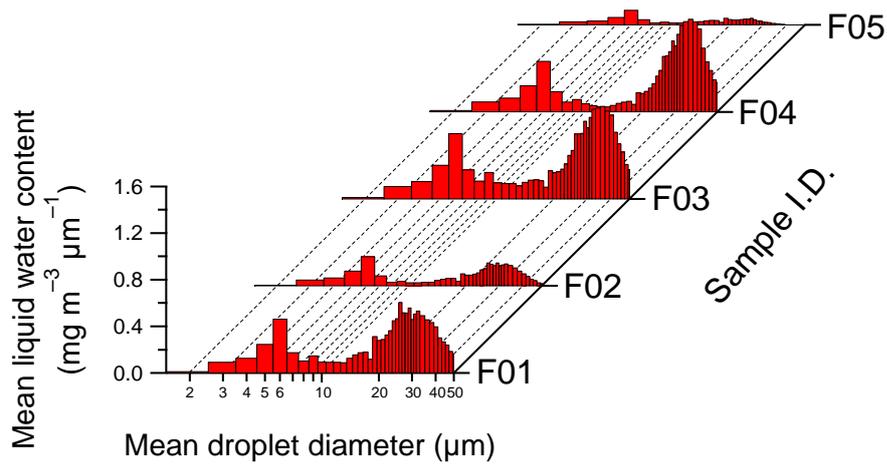
1
 2 Figure 5. The 168 h (7 days) air mass backward trajectories for starting altitudes of 500
 3 (triangle symbols), 1000 (circle symbols) and 1500 m (square symbols) above ground level
 4 (AGL) during the collections of aerosol (a), rain (b), and sea fog (c) samples were calculated
 5 from the Global Data Assimilation System (GDAS) database of the National Ocean and
 6 Atmospheric Administration (NOAA) and simulated by using the Hybrid Single-Particle
 7 Lagrangian Integrated Trajectory (HY-SPLIT) model (web site
 8 <http://www.arl.noaa.gov/ready/hysplit4.html>). White circles, pink triangles and yellow
 9 diamonds indicate aerosol, rain and sea fog sampling locations during the cruise, respectively.
 10 Kasatochi and Okmok volcanoes are black diamonds.



1
 2 Figure 6. Concentrations of NH_4^+ and NO_3^- against sample I.D. in aerosols (a, b), rainwater (c,
 3 d), and sea fog water (e, f) collected over the subarctic western North Pacific Ocean. Solid
 4 triangle lines in (a) and (b) show the percentage of NH_4^+ and NO_3^- in fine ($D < 2.5 \mu\text{m}$)
 5 aerosol particles.
 6

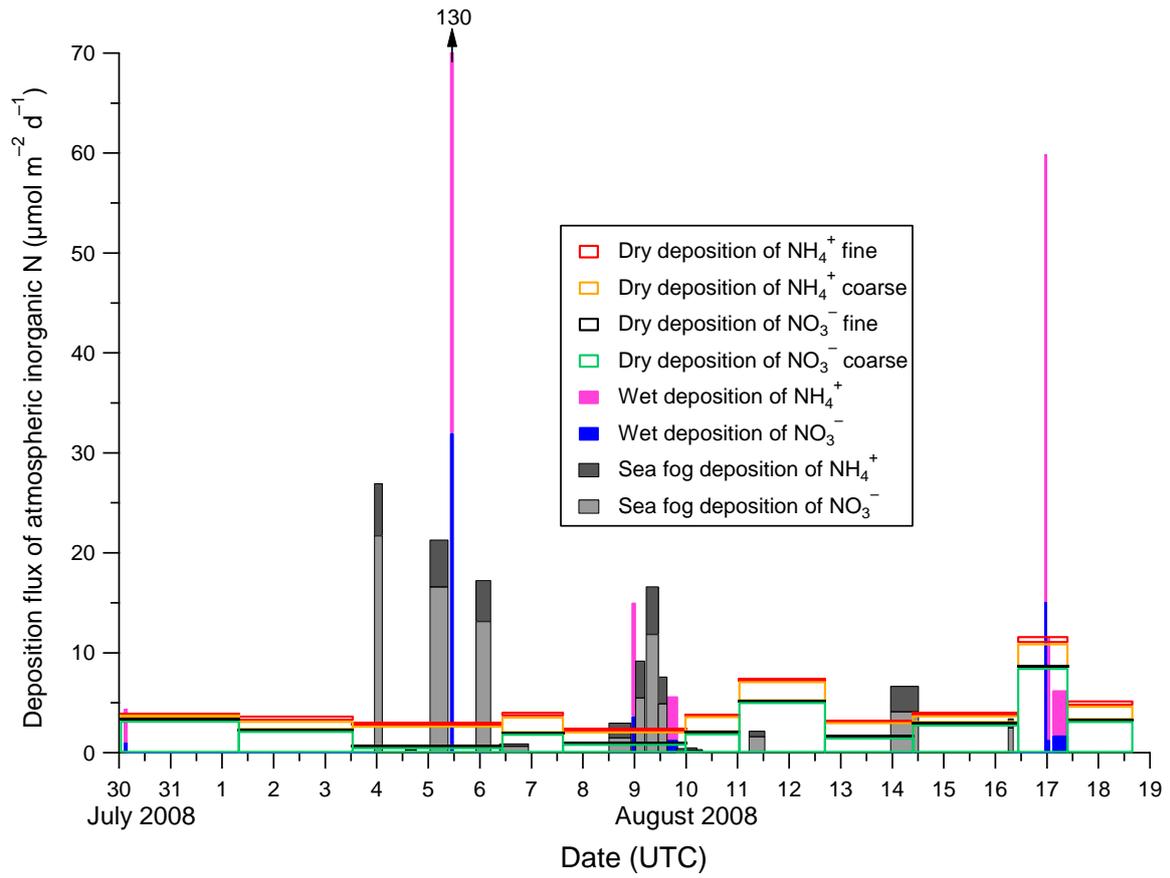


1
 2 Figure 7. The mole equivalent ratios of $\text{NO}_3^-/\text{Na}^+$ in aerosols (red open bars) and sea fog
 3 water (gray bars) during the cruise. The widths of red open and gray bars indicate the
 4 sampling duration of aerosol and sea fog water samples, respectively.
 5



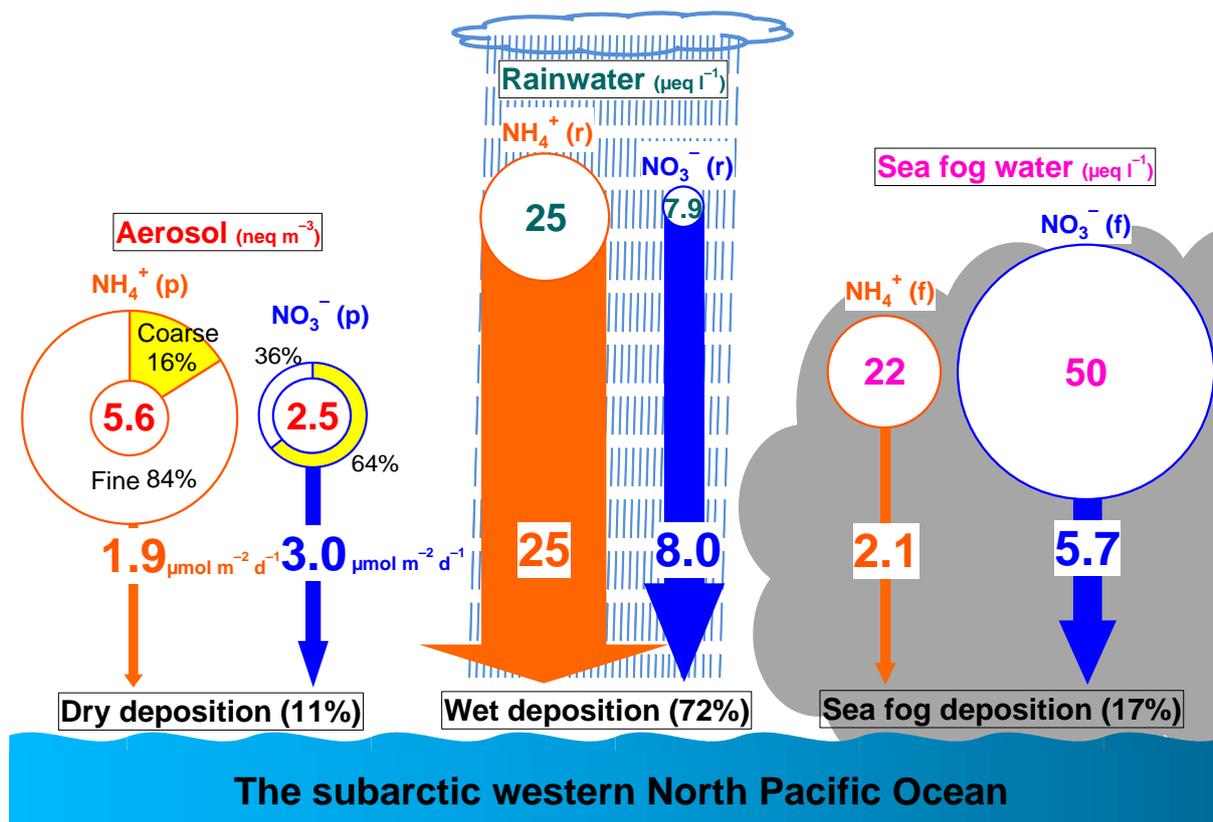
1
2
3
4

Figure 8. Variations of mean LWC for each sea fog event as a function of mean droplet diameter during the cruise.



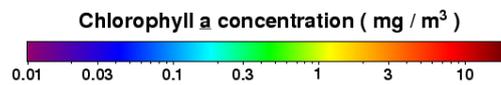
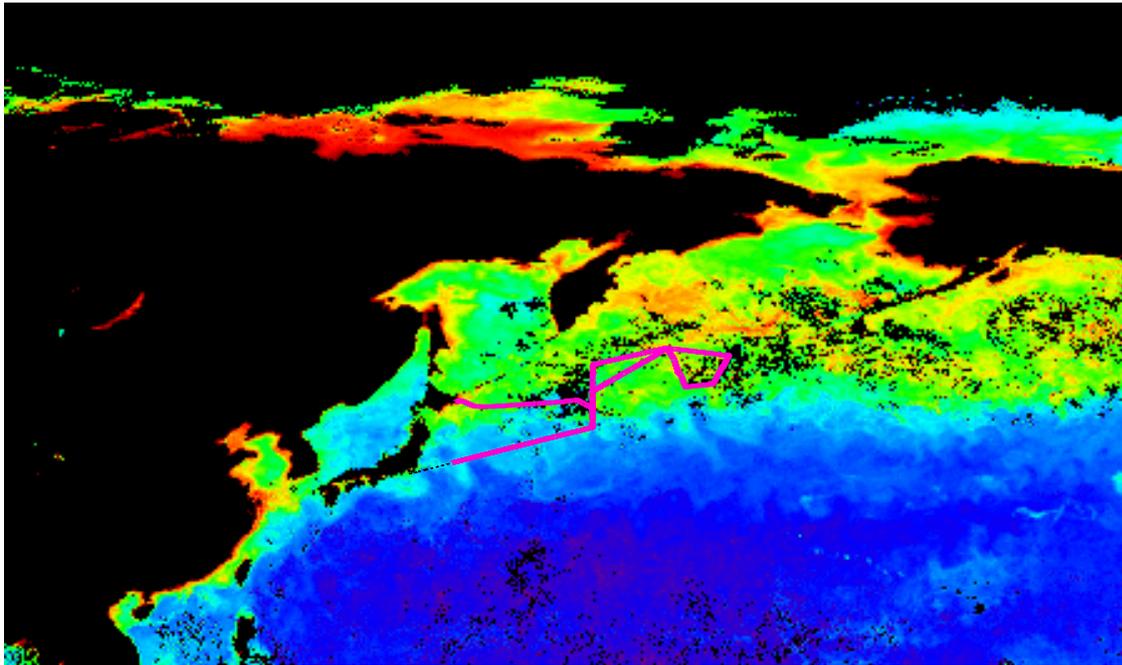
1
2
3
4

Figure 9. Temporal variations of dry, wet, and sea fog deposition fluxes for NH_4^+ and NO_3^- over the subarctic western North Pacific Ocean during the cruise.



1
 2 Figure 10. Schematic diagram of atmospheric inorganic N input to the subarctic western
 3 North Pacific Ocean during 29 July–19 August 2008. The numbers in circles represent
 4 concentrations of NH₄⁺ and NO₃⁻ in aerosols (in neq m⁻³), rainwater (in μeq l⁻¹) and sea fog
 5 water (in μeq l⁻¹). The mean percentages of total (fine + coarse) aerosol concentrations in fine
 6 (D < 2.5 μm, white) and coarse (D > 2.5 μm, yellow) modes for NH₄⁺ and NO₃⁻ are shown in
 7 the circles for aerosol. The orange and blue arrows and numbers (in μmol m⁻² d⁻¹) indicate
 8 NH₄⁺ (orange) and NO₃⁻ (blue) fluxes via dry, wet, and sea fog deposition.

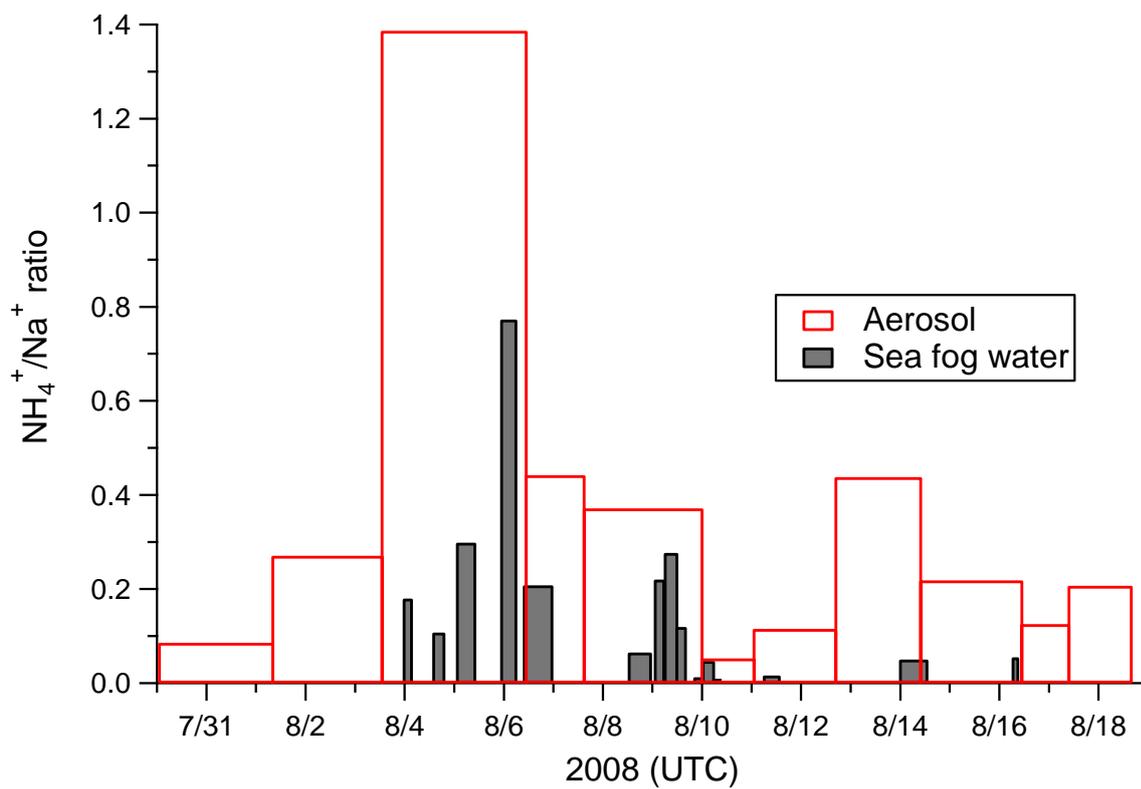
9
 10
 11
 12
 13
 14
 15
 16



1

2 Figure S1. SeaWiFS chlorophyll a image of the subarctic western North Pacific Ocean in
3 August 2008 (web site: <http://oceancolor.gsfc.nasa.gov>) and cruise track (pink line) of Leg 1
4 of the KH-08-2 cruise.

5



1
2
3
4
5
6
7
8
9
10
11
12
13
14
15

Figure S2. The ratios of NH₄⁺/Na⁺ in aerosols and sea fog water during the cruise. The widths of red open and gray bars indicate the sampling duration of aerosol and sea fog water samples, respectively.

1 **Responses to Reviewers' comments**

2 30/11/2012

3

4 Journal: Atmospheric Chemistry and Physics

5 Title: Atmospheric inorganic nitrogen input via dry, wet, and sea fog deposition to the
6 subarctic Western North Pacific Ocean

7 Authors: J. Jung, H. Furutani, M. Uematsu, S. Kim, and S. Yoon

8 Manuscript number: acp-2012-507

9 Manuscript type: Research Article

10

11 Note: Reviewers' comments are high lightened in **blue**, while our responses to reviewers are
12 in **black**.

13

14

15 Dr. Ulrich Pöschl

16 Chief Executive Editor

17 Atmospheric Chemistry and Physics

18 Max Planck Institute for Chemistry,

19 Mainz, Germany

20

21

22 Dear Doctor Pöschl,

23

24 We have attached an electronic copy of manuscript file ready to go to press entitled
25 "Atmospheric inorganic nitrogen input via dry, wet, and sea fog deposition to the subarctic
26 Western North Pacific Ocean" by J. Jung, H. Furutani, M. Uematsu, S. Kim, and S. Yoon for
27 publication in Atmospheric Chemistry and Physics (acp-2012-507). We have modified our

1 text based on the reviewers' comments. We appreciated that the comments from both
2 reviewers improved our manuscript a lot. We believe that the comments from both reviewers
3 were clearly responded in our modified manuscript.

4 We are looking forward to hearing your decision. Thank you for your consideration.

5

6 Sincerely yours,

7 Jinyoung Jung

8

9

10

11

12

13

14 **W. Eugster (Referee):**

15 Major Points:

16 1. The convention that the authors use in their definition of total deposition is not in perfect
17 agreement with conventional usage. Conventionally, dry deposition is considered the sum of
18 gaseous dry deposition plus aerosol particulate matter deposition. Here it is only the latter,
19 whereas many other studies (namely terrestrial studies) neglect aerosol deposition. My
20 suggestion is to make an estimate of gaseous dry deposition in order to obtain the correct
21 relative deposition rates in the manuscript, namely in Table 4. On terrestrial ground in the
22 mid-latitudes it is often the case that (gaseous) dry deposition is on the order of two thirds of
23 total deposition, and hence the relative relevance of fog is lowered if gaseous dry deposition is
24 also considered. It may be that it can be safely assumed that gaseous dry deposition is a small
25 component in the total deposition because of low (background) concentrations; but if it is like
26 this, then it should be stated in the manuscript. For reference our deposition estimates for
27 some Swiss localities can be found in Burkard et al. (2003) and Eugster et al. (1998).

28 (Response) We have agreed to what Dr. Eugster pointed out. In this study, dry deposition
29 fluxes of gaseous inorganic N species such as NH_3 and HNO_3 were not estimated, resulting in

1 the underestimate of dry deposition fluxes. Due to the lack of observational data for gaseous
2 inorganic N species over the western North Pacific Ocean, we used the results of Zhang et al.
3 (2010) to estimate the contributions of gaseous inorganic N to dry deposition fluxes, who
4 reported that dry deposition fluxes for NH_3 and HNO_3 were estimated to be $2.0 \mu\text{mol m}^{-2} \text{d}^{-1}$
5 and $4.7 \mu\text{mol m}^{-2} \text{d}^{-1}$ respectively over the East China Sea in July using the MM5/CMAQ
6 model with the 2004 national emission inventory of China. Assuming that the same amounts
7 of NH_3 and HNO_3 are deposited over the sampling region of this study, the contribution of
8 dry deposition flux to mean total (dry + wet + sea fog) deposition flux for TIN would increase
9 from 11% to 23%. However, it is worth noting that the dry deposition fluxes for NH_3 and
10 HNO_3 would decrease as the distance from shore increased since land-derived gaseous
11 inorganic N species are probably transformed to aerosol particles through acid-base reaction
12 and/or absorption on sea-salt particles during their transport over the ocean (page 16, line
13 number 22–page 17, line number 4).

14

15 2. Statistics. This is a flaw that is found throughout the scientific literature and hence is not
16 specific to this paper, but I feel obliged as a reviewer to make sure statistics are correctly used.
17 In this particular case it is obvious that wherever the standard deviation of reported numbers
18 is larger than the mean – notably for concentrations which cannot be negative – then this is an
19 indication that the wrong statistics are used. Mean and standard deviation are the two
20 parameters of a normal distribution; roughly 68% of the data lie within the mean \pm 1 standard
21 deviation, and 16% are smaller than mean – 1 standard deviation. This is impossible for cases
22 where the standard deviation is larger than the mean and simply indicates that the distribution
23 of the data is not following a standard normal distribution. In such cases the mean may still be
24 meaningful (as the sum divided by the number of samples), but the standard deviation is no
25 statistical parameter for the actual distribution. Hence, either an appropriate model
26 distribution for the data must be found and the respective parameters reported, or (more
27 simple) robust statistics using the empirical data distribution should be used (reporting median,
28 interquartile range, and possible 95% interval if this is relevant for the reader to know). The
29 case is very obvious on page 19,098, but also elsewhere (abstract: numbers for wet and sea
30 fog deposition, but also Tables 1 and 4). You can use a statistical test to test your data for
31 normal distribution (e.g. Shapiro-Wilks test) to convince this reviewer that he's wrong.

1 (Response) As Dr. Eugster suggested, we used a Shapiro-Wilks test to assess normality for
 2 concentrations of chemical components in aerosols, rainwater, and sea fog water using an
 3 IBM SPSS version 20. Here, the samples where each ionic component was below the
 4 detection limit have been included in the assessing of the normality as 0. The results for the
 5 Shapiro-Wilks test are given in three Tables below, revealing that most chemical components
 6 in aerosols, rainwater, and sea fog water were not normally distributed since most
 7 significance levels were lower than 0.05.

8

Tests of Normality						
	Kolmogorov-Smirnov ^a			Shapiro-Wilk		
	Statistic	df	Sig.	Statistic	df	Sig.
Aerosol_Na	.189	11	.200*	.889	11	.135
Aerosol_NH4	.135	11	.200*	.944	11	.575
Aerosol_K	.251	11	.050	.845	11	.036
Aerosol_Mg	.163	11	.200*	.936	11	.477
Aerosol_Ca	.238	11	.082	.862	11	.061
Aerosol_Cl	.176	11	.200*	.853	11	.047
Aerosol_NO3	.140	11	.200*	.941	11	.538
Aerosol_SO4	.140	11	.200*	.961	11	.788
Aerosol_MSA	.274	11	.020	.773	11	.004
Aerosol_nssSO4	.130	11	.200*	.956	11	.715
Aerosol_nssK	.291	11	.010	.643	11	.000
Aerosol_nssCa	.335	11	.001	.688	11	.000
Aerosol_nssMg	.458	11	.000	.426	11	.000

*. This is a lower bound of the true significance.

a. Lilliefors Significance Correction

9

10

Tests of Normality	
Kolmogorov-Smirnov ^a	Shapiro-Wilk

	Statistic	df	Sig.	Statistic	df	Sig.
Rain_Na	.350	7	.010	.741	7	.010
Rain_NH4	.214	7	.200*	.887	7	.257
Rain_K	.289	7	.079	.826	7	.073
Rain_Mg	.294	7	.067	.780	7	.026
Rain_Ca	.253	7	.196	.748	7	.012
Rain_Cl	.247	7	.200*	.826	7	.074
Rain_NO3	.214	7	.200*	.872	7	.192
Rain_SO4	.348	7	.011	.742	7	.011
Rain_MSA	.349	7	.010	.644	7	.001
Rain_nssSO4	.247	7	.200*	.826	7	.073
Rain_nssK	.280	7	.103	.761	7	.016
Rain_nssCa	.291	7	.075	.731	7	.008
Rain_nssMg	.304	7	.049	.688	7	.003
Rain_nssCl	.363	7	.006	.738	7	.009

*. This is a lower bound of the true significance.

a. Lilliefors Significance Correction

1
2

Tests of Normality

	Kolmogorov-Smirnov ^a			Shapiro-Wilk		
	Statistic	df	Sig.	Statistic	df	Sig.
Fog_Na	.291	15	.001	.607	15	.000
Fog_NH4	.120	15	.200*	.956	15	.629
Fog_K	.317	15	.000	.591	15	.000
Fog_Mg	.295	15	.001	.612	15	.000
Fog_Ca	.338	15	.000	.601	15	.000
Fog_Cl	.289	15	.001	.632	15	.000
Fog_NO3	.126	15	.200*	.935	15	.328
Fog_SO4	.239	15	.021	.909	15	.132

Fog_MSA	.224	15	.041	.808	15	.005
Fog_nssSO4	.215	15	.060	.905	15	.114
Fog_nssK	.273	15	.004	.794	15	.003
Fog_nssCa	.186	15	.173	.851	15	.018
Fog_nssMg	.354	15	.000	.576	15	.000

*. This is a lower bound of the true significance.

a. Lilliefors Significance Correction

1
2 Therefore, we have added 25th, 50th, and 75th percentile concentrations in Table 1 instead of
3 standard deviations, as Dr. Eugster suggested. We also have removed the standard deviations
4 for concentrations of chemical components in aerosols, rainwater, and sea fog water from
5 Table 4 and the manuscript.

6
7 3. Following up on the question of the correct distribution of your data the t-test used on page
8 19,098, 1. 8 may not be the appropriate test statistics. Please update according to your finding
9 and reply to the previous point. In many cases the t-test is rather robust to the violation of the
10 assumption of normal distribution, so it is not impossible that your results with the correct
11 statistical test are similar to what the t-test provided, but for the sake of scientific correctness I
12 encourage you to use the appropriate statistical test that applies to your empirical data
13 distribution (maybe a nonparametric test is needed if you cannot define the general statistical
14 distribution model for your data).

15 (Response) We used the Kolmogorow-Smirnov test to assess normality for the particle
16 number densities using the IBM SPSS version 20. The results for the Kolmogorow-Smirnov
17 test revealed that the particle number densities were not normally distributed. Consequently,
18 we used a Wilcoxon Signed Rank Test, which is the nonparametric test equivalent to the
19 dependent t-test, to investigate the differences of particle number densities between non sea
20 fog events and sea fog events periods. As the Wilcoxon Signed-Ranks Test does not assume
21 normality in the data, it can be used when this assumption has been violated.

22

23 **Wilcoxon Signed Ranks Test**

Ranks

		N	Mean Rank	Sum of Ranks
Fog events $0.3 < D < 0.5 \mu\text{m}$ – Non fog events $0.3 < D < 0.5 \mu\text{m}$	Negative Ranks	1775 ^a	2366.57	4200668.50
	Positive Ranks	2502 ^b	1977.55	4947834.50
	Ties	0 ^c		
	Total	4277		
Fog events $0.5 < D < 1 \mu\text{m}$ – Non fog events $0.5 < D < 1 \mu\text{m}$	Negative Ranks	2729 ^d	2080.06	5676484.00
	Positive Ranks	1547 ^e	2241.59	3467742.00
	Ties	1 ^f		
	Total	4277		
Fog events $1 < D < 2 \mu\text{m}$ – Non fog events $1 < D < 2 \mu\text{m}$	Negative Ranks	3476 ^g	2082.89	7240126.50
	Positive Ranks	799 ^h	2377.75	1899823.50
	Ties	2 ⁱ		
	Total	4277		
Fog events $D > 2 \mu\text{m}$ – Non fog events $D > 2 \mu\text{m}$	Negative Ranks	3667 ^j	2147.03	7873145.50
	Positive Ranks	600 ^k	2054.39	1232632.50
	Ties	10 ^l		
	Total	4277		

a. Fog events $0.3 < D < 0.5 \mu\text{m}$ < Non fog events $0.3 < D < 0.5 \mu\text{m}$

b. Fog events $0.3 < D < 0.5 \mu\text{m}$ > Non fog events $0.3 < D < 0.5 \mu\text{m}$

c. Fog events $0.3 < D < 0.5 \mu\text{m}$ = Non fog events $0.3 < D < 0.5 \mu\text{m}$

d. Fog events $0.5 < D < 1 \mu\text{m}$ < Non fog events $0.5 < D < 1 \mu\text{m}$

e. Fog events $0.5 < D < 1 \mu\text{m}$ > Non fog events $0.5 < D < 1 \mu\text{m}$

f. Fog events $0.5 < D < 1 \mu\text{m}$ = Non fog events $0.5 < D < 1 \mu\text{m}$

g. Fog events $1 < D < 2 \mu\text{m}$ < Non fog events $1 < D < 2 \mu\text{m}$

h. Fog events $1 < D < 2 \mu\text{m}$ > Non fog events $1 < D < 2 \mu\text{m}$

i. Fog events $1 < D < 2 \mu\text{m}$ = Non fog events $1 < D < 2 \mu\text{m}$

j. Fog events $D > 2 \mu\text{m}$ < Non fog events $D > 2 \mu\text{m}$

k. Fog events $D > 2 \mu\text{m}$ > Non fog events $D > 2 \mu\text{m}$

I. Fog events $D > 2 \mu\text{m}$ = Non fog events $D > 2 \mu\text{m}$

1
2

Test Statistics ^a				
	Fog events $0.3 < D < 0.5 \mu\text{m}$ – Non fog events $0.3 < D < 0.5 \mu\text{m}$	Fog events $0.5 < D < 1 \mu\text{m}$ – Non fog events $0.5 < D < 1 \mu\text{m}$	Fog events $1 < D < 2 \mu\text{m}$ – Non fog events $1 < D < 2 \mu\text{m}$	Fog events $D > 2 \mu\text{m}$ – Non fog events $D > 2 \mu\text{m}$
Z	-4.626 ^b	-13.680 ^c	-33.086 ^c	-41.258 ^c
Asymp. Sig. (2-tailed)	.000	.000	.000	.000

a. Wilcoxon Signed Ranks Test

b. Based on negative ranks.

c. Based on positive ranks.

3
4
5
6
7
8
9
10
11
12

The obtained results for the Wilcoxon Signed Rank Test are given in two Tables above. The results revealed that the differences of particle number densities between sea fog evens and non sea fog evens periods were statistically significant, and that the particle number densities for aerosols in the ranges of $0.5 < D < 1 \mu\text{m}$, $1 < D < 2 \mu\text{m}$, and $D > 2 \mu\text{m}$ decreased during sea fog events, except for aerosols in the range of $0.3 < D < 0.5 \mu\text{m}$. Therefore, we have revised ‘t-test, $p < 0.05$ ’ to ‘Wilcoxon Signed Rank Test, $p = 0.0005$ ’ (page 10, line number 14) and have removed the standard deviations of particle number densities in section 3.2 (page 10, line number 8, 9, 11, 12, and 13).

13
14
15
16
17
18
19
20

4. The influence of the vessel’s exhaust is minimized in their sample strategy by using conditional data selection with two criteria: (1) wind direction is from within $\pm 100^\circ$ from the bow direction, and (2) wind speed exceeds 1 m s^{-1} . This appears to be a sound approach for data selection in order to minimize the contamination from the sampling vessel. The only concern I have here is with respect to the fog droplet sampling which unfortunately is a passive system (no aspiration as e.g. a CASC would do), so there might be cases where initially the conditions were within the rejection range defined by the authors, but later they were OK and fog might have been collected on the collection mesh that has previously

1 received some contamination from soot from the vessel's engine. I assume the authors have
2 carefully avoided such conditions. They state in Section 2.3 their careful procedure to clean
3 and prepare the sampling tissue, but there is no mention what and how it was ascertained that
4 there was no contamination of the kind I could imagine as explained above. Maybe add a
5 short statement saying exactly how you did this, or provide a statement that such conditions
6 are unlikely (maybe they can be neglected since wind was never low? However, I imagine
7 that fog and low wind speeds correlate in such cases).

8
9 (Response) We have described in section 2.3 that how we avoided the contaminations from
10 ship's exhaust during sea fog water collection (page 5, line number 27–32). Likewise rain
11 sampling, the relative wind directions were monitored during sea fog water collection, and sea
12 fog water samples were collected only when the relative wind directions were within the
13 ranges of the wind-sector controller. If the relative wind directions changed from within to
14 outside the ranges during sea fog water sampling, the sea fog water was considered
15 contaminated and discarded.

16
17 5. Usage of the term “subarctic” Pacific Ocean: there is a good overview provided by Bailey
18 (1996) who defines the subarctic region as a subdivision of the polar domain, but only for
19 land surfaces. Nevertheless, in the eastern Siberian and Alaskan region the subarctic region
20 does not extend to latitudes below 50 N. On his map of the Ecoregions of the Oceans in that
21 book your cruise is entirely in the “temperate” domain (as I would have expected). So my
22 suggestion is to avoid confusion and not use the term subarctic for your study; any reader in
23 my domain would expect a cruise going up further North in order to touch the subarctic
24 region. Use the term “temperate” instead.

25 (Response) Dr. Eugster suggested that “the subarctic western North Pacific Ocean” should be
26 changed to “the temperate western North Pacific Ocean”. However, this study was carried out
27 over the Western Subarctic Gyre. Moreover, “the subarctic North Pacific” or “the western
28 subarctic Pacific” has been used in many studies conducted over the Western Subarctic Gyre
29 (e.g., Harrison et al., 1999; Wong et al., 2002; Tsuda et al., 2003; Takeda and Tsuda, 2005).
30 In addition, most samples of this study were collected in the northern region of the subarctic
31 boundary (~40°N) (Wong et al., 2002). Therefore, we believe that “the subarctic western
32 North Pacific Ocean” can be used in this study.

1

2 Minor Points:

3 1. Parameters vs. variables. You use the word parameter for variables, which unfortunately is
4 quite widespread in meteorology and other disciplines, although parameter in science is
5 actually a pseudo-constant, not a variable. My suggestion is to call variables “variables” and
6 reserve the word parameters for e.g. model parameters of a statistical distribution etc. (mean
7 and standard deviation are such parameters).

8 (Response) We agree with Dr. Eugster’s opinion. We therefore have revised “parameters” to
9 “variables” in section 2.1 (page 4, line number 22) and Fig. 2.

10

11 2. Section 3.7 (Deposition flux estimates) is a methods section and should be moved to an
12 appropriate position in Section 2 (Methods) .

13 (Response) We have moved section 3.7 (Deposition flux estimates) to section 2.6 in the
14 methods section, as Dr. Eugster suggested. In addition, we have added short explanations for
15 the deposition flux estimates to section 3.7 (page 16, line number 9–12).

16

17 3. Section 3.1, meteorological conditions: in Bruijnzeel et al. (2005) we tried to group
18 published literature according to naming conventions for fog types, and distinguished between
19 “sea fog” (fog forming from atmospheric vapor when warm air rests over cold ocean) and
20 “steam fog” (fog forming from evaporation from a warm ocean into a cold atmosphere). The
21 figure that I made to clarify the usage of words was also reprinted in the introductory paper of
22 the special issue of the 2007 fog conference [Eugster (2008)], downloadable from
23 http://www.dieerde.de/DIE_ERDE_2008_1-2_Eugster.pdf. I know that it is difficult to
24 establish a nomenclatura in fog types, but if you agree with what was published earlier, it
25 would help newcomers to try to use these wordings.

26 (Response) We agree with Dr. Eugster’s opinion. In Eugster (2008), he distinguished between
27 sea fog and steam fog. According to Eugster (2008), the fog collected during the sampling
28 period of this study can be defined as “sea fog” from the results of meteorological variables
29 and backward trajectories shown in Figs. 2 and 5(c), respectively. We therefore have revised
30 “Sea fog typically occurs as a result of warm marine air advection over a region where a cold

1 ocean current affects” to “Sea fog typically occurs as a result of cooling of humid air over a
2 cold ocean surface”. We also have added Eugster (2008) as a reference in the manuscript
3 (page 9, line number 17–18).

4

5 4. p. 19,093, l. 6: it is unclear why you call this impactor a “virtual” impactor. As I understand
6 your description it is a “real” impactor, that is, the particles impact on the filter. It the
7 terminology established to call this a virtual impactor? Please clarify.

8 (Response) The aerosol sampler used in this study is called as “high-volume virtual impactor
9 air sampler”.

10 Theoretical studies of virtual impactors are entirely similar to those of real inertial impactors.
11 In the virtual impaction method, instead of the coarse particles impacting onto a solid
12 collection surface, they impact into a slowly pumped void and are later collected on a filter
13 downstream. Thus, these coarse particles impact through a non-existent void, or virtual
14 surface. To give more information, we have added the explanation on the virtual impactor air
15 sampler to section 2.1 (page 4, line number 4–10). The virtual impactor air sampler utilizes an
16 acceleration nozzle to divide the intake air into the major flow carrying the fine particles ($D <$
17 $2.5 \mu\text{m}$) and the minor flow carrying the coarse particles ($D > 2.5 \mu\text{m}$). The major air flow is
18 diverted laterally after passing through the acceleration nozzle and the inertia of the coarse
19 particles carries them into a collection probe aligned with the acceleration nozzle. Both
20 particle fractions are then collected on the same filter. Details of the virtual impactor are
21 described in Loo and Cork (1988).

22

23 5. p. 19,094, l. 8–9: was the rain sampler opened manually? Most likely so, but it would help
24 to explicitly state that this was not an automatic procedure.

25 (Response) The rain sampler used in this study was opened manually. We therefore have
26 added “manually” to the procedure of rainwater sampling (page 5, line number 1).

27

28 6. p. 19,095, l. 16–22: sounds perfect, but be aware of the fact that the assignment of FM-100
29 signals droplet sizes is somewhat more complex than the standard procedure that we, you and
30 others have used so far. Please have a look at Spiegel et al. (2012) and then decide. My best

1 guess is that in your case the Mie scattering effect is within the uncertainty of your deposition
2 estimates anyway, but only you know exactly how you operated the FM-100.

3 (Response) Until now, the uncertainties of droplet measurements for the fog monitor (FM-
4 100) have not been considered in the most studies (e.g. Burkard et al., 2002; Eugster et al.,
5 2006) where the fog monitor was used. Recently, Spiegel et al. (2012) evaluated the influence
6 of Mie scattering on the droplet size spectra collected with the fog monitor and the droplet
7 losses during sampling with the fog monitor. They also recommended choosing the 40
8 channel thresholds to reduce the error from Mie scattering, and doing loss calculations for the
9 droplet measurements using an ultrasonic anemometer and other instrument (e.g. Particulate
10 Volume Monitor, PVM-100) for a reference. In this study, the 40 channel thresholds were
11 used for the measurement of sea fog droplets as Spiegel et al. (2012) recommended,
12 indicating that it is sufficient for the determination of the total droplet number concentration
13 or the total liquid water content (Spiegel et al., 2012). However, we could not calculate the
14 droplet losses during sampling with the fog monitor because an ultrasonic anemometer and
15 other instrument for a reference were not used in this study. We therefore have added these
16 sentences, which are mentioned above, to section 2.3 (page 6, line number 13–24).

17

18 7. p. 19,110, l. 6–8: to obtain such an estimate you most likely must have made an assumption
19 on the C:N ratio to yield an estimate for N requirements based on C flux data. Please specify
20 what assumptions you have made, which literature values (with references) were available for
21 making such an assumption, or specify what other basis you used for this estimate.

22 (Response) Although the assumption for the estimate of potential primary production was
23 already mentioned in page 18 line number 21–23, we have also added to page 18, line number
24 19–21 that the average elemental C/N requirement for many plankton species and for bulk
25 organic matter in the oceans has been found to be relatively constant at ~6.6 (mol/mol)
26 (Redfield, 1958), as Dr. Eugster suggested. In addition, we have revised “Based on these,” to
27 “Based on these estimates by Elskens et al. (2008) and Wong et al. (2002),” to clarify the
28 context (page 19, line number 1).

29

30 8. Table 1: you honestly mention that you used a value of 0 for cases with negative
31 concentrations or concentrations below detection limit. This however is not the standard

1 approach as I understand; see for example Helsel (1990). Your numbers should not change
2 much, but for the sake of scientific correctness I recommend using the procedure suggested
3 by Helsel.

4 (Response) As Dr. Eugster suggested, we have read the paper. It is written in Helsel (1990)
5 that “The substitution of zero produces estimates of mean and median that are biased low,
6 whereas substituting the reporting limit results in estimates above the true value”. In addition,
7 other studies (e.g., Mace et al., 2003a, 2003b) used the same method we used in this study, to
8 calculate the mean values of chemical components, including the samples where their
9 concentrations were below detection limits, in aerosols and/or rainwater. As given in Table 1,
10 the sample numbers of chemical components not detected (i.e., MSA and nss-ionic species) in
11 aerosols, rainwater, and sea fog water are few (i.e., one or two samples), except for nss-Mg²⁺.
12 Moreover, atmospheric inorganic nitrogen species in aerosols, rainwater, and sea fog water
13 had enough high concentrations to be detected in this study. We therefore believe that the
14 method for calculating the mean concentration would not affect the results of deposition
15 fluxes for atmospheric inorganic nitrogen, and that the mean values would not change much
16 as Dr. Eugster mentioned in the question.

17

18 9. Table 2: caption does not mention that also seawater ratios are given.

19 (Response) We have added the mention of seawater ratios to the caption for Table 2.

20

21 10. Fig. 8: I am surprised how large the droplets are in your fog! But that’s probably the
22 characteristics of this fog and is fine. But I find the labels on the x-axis misleading where you
23 label 20 μm with 2, and 40 μm with 4, a factor 10 off, which should be corrected.

24 (Response) The log scale of x-axis we used in Fig. 8 is a typical form. This form is widely
25 used in other studies. However, Dr. Eugster pointed out that it can be misleading. We
26 therefore have revised the x-axis of Fig. 8 as he suggested.

27

28 11. Fig. 10: nice overview, but since there is a different reference for aerosol concentrations
29 than rainwater/fog water, the circles are misleading in that they imply a much higher aerosol
30 concentration than there actually is. Would it be possible to draw the circles to scale, and

1 since for aerosols they would be utterly tiny, use a zoom and show the two circles that you
2 present now in a graphical display that clearly indicates the reader that these two were
3 magnified (e.g. small box removed to the left and lines connecting it to the same display at
4 correct size).

5 (Response) As Dr. Eugster suggested, we have redrawn the circles for NH_4^+ and NO_3^- in
6 rainwater and sea fog water and the arrows for deposition fluxes to scale in Fig. 10. For the
7 circles of aerosol NH_4^+ and NO_3^- , we have redrawn them to scale; however the circles for
8 aerosol have been considered separately from those for rainwater and sea fog water, because
9 the units (neq m^{-3}) for NH_4^+ and NO_3^- concentrations in aerosols are different from those
10 ($\mu\text{eq l}^{-1}$) in rainwater and sea fog water. Although the scale of the circles for aerosol is
11 different, we believe that Fig. 10 gives the information on atmospheric inorganic nitrogen
12 input via dry, wet, and sea fog deposition to the subarctic western North Pacific Ocean, since
13 the deposition fluxes for atmospheric inorganic nitrogen (the arrows) were drawn to the same
14 scale.

15

16 **Typographical Details**

17 • p. 19,090, l. 11: delete “s” in reveal (it is plural)

18 (Response) We have revised “reveals” to “reveal” (page 1, line number 20).

19 • p. 19,091, l. 7: add “s” to constituents (plural)

20 (Response) We have revised “constituent” to “constituents” (page 2, line number 13).

21 • p. 19,109, l. 23: “co-limitation” is typically written with a hyphen

22 (Response) We have revised “co limitation” to “co-limitation” (page 18, line number 23).

23

24 **References**

25 Burkard, R., Eugster, W., Wrzesinsky, T., and Klemm, O.: Vertical divergence of fogwater
26 fluxes above a spruce forest, *Atmos. Res.*, 64, 133–145, 2002.

27 Eugster, W., Burkard, R., Holwerda, F., Scatena, F. N., and Bruijnzeel, L. A.: Characteristics
28 of fog and fogwater fluxes in a Puerto Rican elfin cloud forest, *Agric. For. Meteorol.*,
29 139, 288–306, 2006.

- 1 Harrison, P. J., Boyd, P. W., Varela, D. E., Takeda, S., Shiomoto, A., and Odate, T.:
2 Comparison of factors controlling phytoplankton productivity in the NE and NW
3 subarctic Pacific gyres, *Prog. Oceanogr.*, 43, 205–234, 1999.
- 4 Loo, B. W. and Cork, C. P.: Development of High Efficiency Virtual Impactors, *Aerosol Sci.*
5 *Technol.*, 9, 167–176, 1988.
- 6 Mace, K. A., Kubilay, N., and Duce, R. A.: Organic nitrogen in rain and aerosol in the eastern
7 Mediterranean atmosphere: An association with atmospheric dust, *J. Geophys. Res.*,
8 108(D10), 4320, doi:10.1029/2002JD002997, 2003a.
- 9 Mace, K. A., Duce, R. A., and Tindale, N. W.: Organic nitrogen in rain and aerosol at Cape
10 Grim, Tasmania, Australia, *J. Geophys. Res.*, 108(D11), 4338,
11 doi:10.1029/2002JD003051, 2003b.
- 12 Spiegel, J. K., Zieger, P., Bukowiecki, N., Hammer, E., Weingartner, E., and Eugster, W.:
13 Evaluating the capabilities and uncertainties of droplet measurements for the fog
14 droplet spectrometer (FM-100), *Atmos. Meas. Tech.*, 5, 2237–2260, 2012.
- 15 Takeda, S. and Tsuda, A.: An in situ iron-enrichment experiment in the western subarctic
16 Pacific (SEEDS): Introduction and summary, *Prog. Oceanogr.*, 64, 95–109, 2005.
- 17 Tsuda, A., Takeda, S., Saito, H., Nishioka, J., Nojiri, Y., Kudo, I., Kiyosawa, H., Shiomoto,
18 A., Imai, K., Ono, T., Shimamoto, A., Tsumune, D., Yoshimura, T., Aono, T., Hinuma,
19 A., Kinugasa, M., Suzuki, K., Sohrin, Y., Noiri, Y., Tani, H., Deguchi, Y., Tsurushima,
20 N., Ogawa, H., Fukami, K., Kuma, K., and Saino, T.: A Mesoscale Iron Enrichment in
21 the Western Subarctic Pacific Induces a Large Centric Diatom Bloom, *Science*, 300,
22 958–961, doi: 10.1126/science.1082000, 2003.
- 23 Wong, C. S., Waser, N. A. D., Nojiri, Y., Whitney, F. A., Page, J. S., and Zeng, J.: Seasonal
24 cycles of nutrients and dissolved inorganic carbon at high and mid latitudes in the
25 North Pacific Ocean during the Skaugran cruises: determination of new production and
26 nutrient uptake ratios, *Deep-Sea Res. Pt. II*, 49, 5317–5338, 2002.
- 27 Zhang, Y., Yu, Q., Ma, W., and Chen, L.: Atmospheric deposition of inorganic nitrogen to the
28 eastern China seas and its implications to marine biogeochemistry, *J. Geophys. Res.*,
29 115, D00K10, doi:10.1029/2009JD012814, 2010.

30

1

2 **Anonymous Referee #2:**

3 General comments

4 The authors of this manuscript have as their goals (1) to describe the characteristics of sea fog,
5 (2) to estimate and apportion the fluxes of N from atmosphere to sea from dry, wet, and fog
6 deposition, (3) to assess the impact of these N fluxes on primary production in the open ocean.
7 They give a well-written report on research conducted at sea in the northwestern Pacific
8 Ocean. Research activities included standard methods for aerosol, rainwater, and sea fog
9 sample collection and analyses. The authors provide a thorough data analyses with reasonable
10 explanations for their observations. The biggest drawback of this research is the limited
11 number of samples and relatively short sampling period, which limit the inferences that can
12 be drawn from the data. This is offset by the opportunity to combine their data with similar
13 data sets from other research cruises for a future meta-analysis.

14 (Response) As Referee #2 suggested, we have tried to combine our results with the data from
15 other studies carried out over the subarctic western North Pacific Ocean to estimate
16 deposition fluxes of atmospheric inorganic nitrogen. However, we could not find it. To our
17 knowledge, our study is the first one to estimate deposition fluxes of atmospheric inorganic
18 nitrogen in the subarctic western North Pacific Ocean. In addition, we have recognized the
19 drawback Referee #2 pointed out; however, we have already mentioned in section 4 that
20 “Further studies, however, are required to understand the biogeochemical cycles of N more
21 clearly and should focus on long-term monitoring of atmospheric reactive N species,
22 including organic N, in the subarctic western North Pacific Ocean” (page 20, line number 12–
23 14). Although sampling period of this study is limited, we believe that this study contributed
24 to the understanding of atmospheric nitrogen cycle in open ocean environment, and that the
25 results for atmospheric inorganic nitrogen deposition from this study are valuable for filling
26 the data gap, especially for the atmospheric inorganic nitrogen input by sea fog deposition to
27 the subarctic western North Pacific Ocean.

28

29 Specific comments

1 On page 19101, line 6, did the authors mean sodium nitrate instead of ammonium nitrate? Per
2 their discussion, nitrate is found in the coarse particle mode and is most likely associated with
3 sodium, calcium, or magnesium rather than ammonium.

4 (Response) In section 3.4, we have discussed the neutralization of acidic substances in
5 rainwater and sea fog water. As Referee #2 pointed out, aerosol NO_3^- was largely associated
6 with coarse mode particles. However, our result suggests that not only aerosol NO_3^- , but also
7 gaseous HNO_3 was scavenged by sea fog water, as we have discussed in section 3.6 (page 15,
8 line number 27–page 16, line number 3). Therefore, NO_3^- derived from gaseous HNO_3 could
9 be neutralized by NH_4^+ in sea fog water and exist as ammonium nitrate (NH_4NO_3).

10

11 Technical comments

12 The location of the research cruise is described as "subarctic Western North Pacific", while
13 references to research by Sasakawa et al (2005, e.g.) refer to "Northwestern North Pacific" as
14 the location of a similar cruise, although the two cruises appear to be in similar quadrants of
15 the Pacific Ocean. Please clarify for the reader if these are similar or different measurement
16 domains.

17 (Response) In the manuscript, we have referred the concentrations of NH_4^+ and NO_3^- in
18 aerosols, rainwater and sea fog water and $\text{NH}_4^+/\text{nss-Ca}^{2+}$ ratio in rainwater reported by
19 Sasakawa and Uematsu (2002). They used 'the northwestern North Pacific' to describe their
20 study area. Although their study area is similar to ours, we have not changed 'the
21 northwestern North Pacific' to 'the subarctic western North Pacific' to respect their
22 expression. However, we have realized that the expression could confuse the reader, as
23 Referee #2 pointed out. Therefore, we have added the ranges of latitude and longitude of their
24 study area (34°N–48°N, 137°E–155°E) to the manuscript (page 11, line number 23; page 13,
25 line number 23; page 14, line number 6; page 14 line number 16).

26