Dear Dr. Nikolaos Mihalopoulos,

Please find below our responses to the reviewers' comments and the marked-up version of manuscript showing the corresponding changes. We would like to thank you for the time and effort you put into the editorial handling of this manuscript.

Best Regards,

Guebuem Kim, on behalf of all authors

Response to Anonymous Referee #1

I understood my questions from your answer. Thank you for your answer! The sources and components of organic nitrogen in aerosols and rain are still unknown, I expect this research will progress in the future. I support this publication as a discussion paper to ACP in a final version.

=> We appreciate Anonymous Referee #1 for his/her constructive comments which improved our original manuscript during the access review period.

Response to Anonymous Referee #2

General comments: This manuscript deals with the atmospheric deposition of nitrogen, particularly the organic fraction, from northwestern Pacific. The data provide new information in key regions where organic N has not previously been measured and help to better evaluate the role of organic N in the global biogeochemical cycle of atmospheric N. The authors acknowledge in their manuscript that" The results presented might be subject to large uncertainties, such as those associated with spatial variations of depositional fluxes across the EJS, the proportions of organic nitrogen in dry deposition, and the bioavailability of organic nitrogen in the atmosphere", however I believe that this study is a step forward to better understand the role of atmospheric organic N in marine environments. Here are some minor comments that may help the authors to improve their manuscript.
=> We would like to thank Anonymous Referee #2 for the careful review and constructive comments.

Specific comments:

Page 5, line 12: Please indicate the number of samples.

=> The number of samples has been added in the revised version of manuscript (Page 4, Line 9).

Page 7, line 4: I wonder how the author estimate the detection limit of DON, since there is no analytical method to measure it directly. Please comment.

=> While in most previous publications detection limit (DL) of DON was not given, Zhang et al. (2001) and Walker et al. (2012) estimated this value using the DLs for each nitrogen species, following the error propagation of standard deviation. In the same manner, using the DLs of NO_3^-, NH_4^+, and TDN (calculated as three times the standard deviation of blanks), we estimated the DL of DON by treating DLs as standard deviations. The DL value for TDN involving in this estimation was 1.1 μmol L^{-1}, which is in line with the typical value for a Shimadzu TOC/TN analyzer using high-temprature catalytic oxidation method (Badr et al., 2003). The value of 2.5 for TDN in the manuscript was calcuated using the blanks obtained during another period when the instrument was less stable. We have clarified the confusions and corrected the mistake in the revised manuscript (Page 5, Lines 21–24).

Page 7, line 5: Please refer what kind of analytes you certified by using the reference materials.

=> This information has been added in the revised version (Page 5, Lines 25–26).

Page 9: The discussion in section 3.1.2 is based on the classification of air mass origin. I suggest to change the title as follows: Potential source regions based on air mass origin.

=> The title for this section has been changed as suggested (Page 7, Line 17).

Page 12, line 1 -11: Maybe it is worth to comment the correlation founded between DON and NH4+.

=> In this study, based on the knowledge of major factors controlling the variations of chemical compositions of our samples (obtained using principal factor analysis), we speculate that DON mainly originated from agricultural activities by using the exclusive method as well as other evidences found in literatures. The correlation between DON and NH_4^+ was less significant (R^2 = 0.39). In addition to the source distribution, the interrelationships among chemical species in wet deposition are also influenced by other factors, such as chemical and physical reactions involving these species (e.g., neutralization of NH_4^+ by acidic anions). Therefore, we did not use correlation analysis for source identification of DON in this work.

Page 37, Fig.4: It would be better, if in this figure you included also the number of samples corresponded to each air mass sector.

=> The number of samples corresponding to each air mass sector has been included in this figure as suggested (Page 35).

Technical corrections:

Page 11, line 17:.... indicates *that* crustal contribution....

=> It has been modified as suggested (Page 9, Line 18).

Page 35, Table 4: Please replace "pus" with "plus"

=> It has been revised (Page 30).

References

Badr, E.-S.A., Achterberg, E.P., Tappin, A.D., Hill, S.J., Braungardt, C.B. (2003) Determination of dissolved organic nitrogen in natural waters using high temperature catalytic oxidation. *Trends in Analytical Chemistry* **22**, 819–827.

Walker, J.T., Dombek, T.L., Green, L.A., Gartman, N., Lehmann, C.M.B. (2012) Stability of organic nitrogen in NADP wet deposition samples. *Atmospheric Environment* **60**, 573–582.

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Sources and fluxes of organic nitrogen in precipitation over the southern East/Japan Sea: Potential impacts on marine productivity

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Abstract

We measured total dissolved reactive nitrogen in precipitation samples collected at Uljin, a Korean coastal site upwind of the southern East/Japan Sea (EJS), selected as a representative study site of atmospheric deposition over the northwestern Pacific margin. NO_3^- was found to be the most abundant nitrogen species, followed by NH_4^+ and dissolved organic nitrogen (DON). Air mass back trajectory analysis revealed that a significant fraction of the inorganic nitrogen (NO_3^- and NH_4^+) originated from mainland Asia, whereas the DON was primarily derived from Korea. Using varimax-rotated factor analysis in combination with major ions as tracers, agricultural activities in Korea were identified as the primary sources of DON in these samples. In addition, a positive correlation was found at Uljin between the size of organic fraction in total reactive nitrogen and nitrogen to carbon atomic ratio in organic matter. This correlation has also been observed at other locations worldwide, implying the utilization potential of atmospheric organic nitrogen might increase with its proportion in total nitrogen. Combining wet deposition measurements in this study with literature values for dry deposition observed at a remote island in the EJS, the total atmospheric depositional flux of reactive nitrogen was estimated to be 115 mmol N m^{-2} yr^{-1} over the southern EJS. Our study sheds new light on the potentially significant contribution to primary productivity of the northwestern Pacific Ocean by atmospheric deposition of nitrogen, especially the organic fraction.

1 Introduction

The availability of reactive nitrogen (or fixed nitrogen), including all nitrogen species except N_2, is often a determinant controlling the primary production in terrestrial and marine ecosystems (Vitousek and Howarth, 1991). However, since the mid-1800s the global nitrogen cycle has been significantly perturbed by excessive production of reactive nitrogen through human activities (at a similar rate to natural biological fixation) (Dentener et al., 2006; Galloway et al., 2008; Fowler et al., 2013), such as the Haber–Bosch process, fossil fuel combustion, and legume cultivation (Grubber and Galloway, 2008). A fraction of the reactive nitrogen is transported from continents to oceans via rivers, groundwater, and the atmosphere, exerting a significant influence on marine ecology and biogeochemistry (e.g., acidification, eutrophication, and oxygen depletion) (Seitzinger et al., 2005; Paytan et al., 2006; Doney et al., 2007; Krishnamurthy et al., 2010). In particular, the supply of external or "new" nitrogen to marine systems through atmospheric deposition can be substantial in some marginal and coastal seas (Beddig et al., 1997; Castro and Driscoll, 2002; de Leeuw et al., 2003; Uno et al., 2007, Zhang et al., 2010), as well as the open ocean (Galloway et al., 2004; Duce et al., 2008).

The East/Japan Sea (EJS) is a semi-closed marginal sea (covering an area of 1.01×10^6 km^2) surrounded by Russia, Korea, and Japan, which has been suggested to be an ideal site to investigate the impacts of atmospheric nutrient deposition on the northwestern Pacific Ocean (Kim and Kim, 2013). It is a highly productive region where nitrogen is severely limited (Talley et al., 2004; Jenkins, 2008; Kim and Kim, 2013). Therefore, the supply of reactive nitrogen is particularly important in determining the primary productivity of this marine ecosystem. In addition to the upwelling of deep waters and N_2 fixation by diazotrophs within the ocean, land-derived reactive nitrogen (which is largely anthropogenic) can also contribute to the nitrogen required for primary production in the EJS. Since no major rivers flow into the EJS from the surrounding coasts, the fluvial inputs of nitrogen can be ignored (Kang et al., 2009; Yoo and Park, 2009). Nevertheless, terrigenous nitrogen transported via the atmosphere may reach the open ocean, contributing to the nitrogen inventory across the EJS. In fact, the mid- and long-range atmospheric transport of dust and pollutants from land to the northwestern Pacific Ocean (including the EJS) has been shown to be remarkable (Jo et al., 2007; Kang et al., 2009; Kang et al., 2011, Kim et al., 2011a; Zhang et al.,2011), particularly since this region is located downwind of East Asia, a densely populated area characterized by

intensive emissions of aerosols and gases by anthropogenic and natural processes(Cooke et al., 1999; Richter et al., 2005; Kim, 2008). Recently, Kim et al. (2011a) suggested the significant atmospheric nitrogen deposition has switched extensive parts of the northwestern Pacific Ocean (including the EJS) from being nitrogen-limited to phosphorous-limited during the last three decades. A follow-up study by Kim et al. (2013), however, attributed this increasing trend in relative abundance of nitrogen over phosphorous to nutrient transport by ocean currents. Nevertheless, Kim and Kim (2013) argued that this oceanic region would remain nitrogen-limited for the next 100 years, based on observational data and model predictions. The debate aroused here signifies the importance of an unambiguous budget of nitrogen in the EJS, especially the fraction transported by the atmosphere, which is poorly constrained due to a lack of data (Zhang et al., 2011).

The atmospheric deposition of nitrogen over the southern EJS was previously evaluated by Kang et al. (2010) using dry deposition observations combined with estimates for wet deposition. However, their approximation on nitrogen deposition was an underestimate, since it did not take into account organic nitrogen, which is a globally significant constituent of total reactive nitrogen in the atmosphere (ca. 30% on average) (Jickells, 2006; Cape et al., 2011; Cornell, 2011). Indeed, the growing weight of evidence from global observational studies suggests that a considerable fraction of deposited nitrogen is in organic form (Cornell, 2011, and references therein), which is able to stimulate the productivity of bacteria and phytoplankton in the receiving marine ecosystems (Seitzinger and Sanders, 1999, Bronk et al., 2007). In particular, atmospheric organic nitrogen deposition was shown to be especially high over Asia (Cornell, 2011; Ito et al., 2014), implying it is imperative to incorporate this fraction into the budget of atmospheric nitrogen input to the EJS. Nevertheless, although quantitatively significant, the sources, chemical compositions, and bioavailability of atmospheric organic nitrogen are largely unknown (Mace et al., 2003a; Duce et al., 2008; Jickells et al., 2013), hindering our understanding of its biogeochemical role in the receiving ecosystems.

In order to provide better constraints on atmospheric nitrogen deposition to the EJS, we analyzed reactive nitrogen species (inorganic and organic) and major ions in precipitation samples collected at a coastal site upwind of this marginal sea. The emphasis of the present work was placed on organic nitrogen, which has been ignored in previous study and is generally poorly characterized in comparison with its inorganic counterpart. With the aid of

air mass back trajectory and factor analysis, the geographical and emission sources of organic nitrogen were explored. Moreover, using the calculated total nitrogen depositional flux, the fraction of new primary production of the EJS which is potentially supported by atmospheric nitrogen deposition was estimated.

2 Materials and methods

2.1 Study site and sample collection

The sampling campaign was conducted at Uljin (37 ℃N, 129.4 ℃E), which is located on the eastern coastline of Korea (Fig. 1). It is a rural area with a small population of around 52,000 inhabiting an area of 989.06 km^2. Precipitation samples (N = 33) were collected on an event basis on the rooftop of a four-storey research facility (ca. 16 m above the ground) at the Korean Institute of Ocean Science and Technology (East Sea branch) from February 2011 to January 2012. A home-made sampler was employed, which is composed of a polypropylene funnel (dia. 250 mm) connected by Tygon FEP-lined tubing to a high-density polyethylene (HDPE) bottle placed in a covered bucket. Prior to use, the apparatus was thoroughly cleaned using dilute hydrochloric acid and rinsed with deionized (Milli-Q) water (18.2 MΩ cm). The sampler was manually deployed at the onset of the precipitation events and retrieved after cessation. The impacts of dry deposition were minimized by restricting the exposure time to dry conditions (i.e., <1 h for daytime events or ≤5 h for overnight events). After collection, the samples were transferred to a laminar flow clean room and filtered through pre-combusted (5 h at 500°C) Whatman 0.7 μm GF/F glass fiber filters. Subsamples for dissolved organic carbon (DOC) analysis were placed in 20 mL pre-muffled glass ampoules (5 h at 500 ℃) and chemically preserved with 6 M pure hydrochloric acid, followed by fire sealing. Aliquots for dissolved nitrogen species and major ions were stored in pre-cleaned HDPE bottles and kept frozen at –20 ℃ until analysis.

2.2 Analytical methods

Total dissolved nitrogen (TDN) and DOC concentrations were measured simultaneously by high temperature catalytic oxidation (HTCO) using a Shimadzu TOC/TN analyzer (Model TOC-$V_{CPH/CPN}$) equipped with an ASI-V auto-sampler. The acidified samples (pH < 2 using HCl) were sparged with carbon dioxide free carrier gas (UHP oxygen) at a flow rate of 150 ml/min for 2 min to remove inorganic carbon. Then the samples were injected into a combustion column packed with Pt coated alumina beads heated to 720 ℃. The carbon

dioxide and nitrogen monoxide evolving from combustion were detected by a non-dispersive infrared detector and a chemiluminescence detector, respectively. Major ions including inorganic nitrogen species were determined by high performance liquid chromatography using a Waters 2695 HPLC system equipped with a Waters 432 conductivity detector. Cation analysis (i.e., Na^+, K^+, Ca^{2+}, Mg^{2+}, and NH_4^+) was carried out using a mobile phase consisting of 3.0 mmol L^{-1} HNO_3 and 0.1 mmol L^{-1} EDTA, and a Waters IC-Pak C M/D column (150 × 3.9 mm, 5 μm). Anions (i.e., Cl^-, NO_3^-, NO_2^-, and SO_4^{2-}) were analyzed using a borate/gluconate eluent containing 12% acetonitrile and a Waters IC-Pak A HR column (75 × 4.6 mm, 6 μm). The column was maintained at 30 ℃ and the detector at 35 ℃. Dissolved organic nitrogen (DON) was quantified as the difference between TDN and dissolved inorganic nitrogen (DIN), which is the sum of NO_3^- and NH_4^+ (NO_2^- in our samples was always below the detection limit and is therefore not included). This approach could yield large uncertainties for DON, especially when DIN dominates the nitrogen pool, as being suggested by previous studies (Cornell et al., 2003; Lesworth et al., 2010). The average relative standard deviations associated with replicate measurements of standards were 4%, 2%, and 3% for TDN, NO_3^-, and NH_4^+, respectively. Based on error propagation using uncertainties of each analyte, the precision of DON was estimated to be less than 18% when DON% is greater than 20%, 18–60% when DON% varies from 5% to 20%, and greater than 100% when DON% is less than 5% (e.g., 18/3/2011, 30/5/2011, and 16/8/2011). The limits of detection calculated as three times the standard deviation of blanks are 0.5 μmol L^{-1} for NH_4^+, 0.2 μmol L^{-1} for NO_3^-, 0.2 μmol L^{-1} for NO_2^-, 1.1 μmol L^{-1} for TDN, and 5.0 μmol L^{-1} for DOC, respectively. The detection limit for DON was estimated to be 1 μmol L^{-1}, (Cornell et al., 1998; Zhang et al., 2001; Walker et al., 2012), by using detection limits of relevant nitrogen speices following error propagation rules for standard deviations. Certified reference materials (from University of Miami for DOC and TDN, and from National Research Council of Canada for DIN) were employed during the sample analysis to confirm the data quality.

3 Results and discussion

3.1 Distributions of dissolved reactive nitrogen in precipitation

3.1.1 Concentrations and speciation

The concentrations of dissolved reactive nitrogen showed considerable variations during the sampling year, ranging from 2 to 428 μmol L^{-1} for NO_3^-, from 1 to 139 μmol L^{-1} for NH_4^+, and from below the detection limit to 145 μmol L^{-1} for DON, respectively (Table 1 and Fig. 2). Rather similar temporal trends were found for all three nitrogen species, with high abundances observed in spring and fall (Fig. 2). Aside from the source strength, the distribution pattern displayed here is likely to be influenced by several factors specific to this location. The precipitation amount might be one of the most important factors, because the variation patterns of these nitrogen species are in general accordance with the precipitation regime of Korea (particularly high in summer), in a manner that the concentrations decrease as the precipitation depth increases. In addition, the wind systems (or air mass origins) of Korea may also contribute significantly to the observed temporal trend (see section 3.1.2.). The northwesterly prevails in most time of the year, transporting large amounts of terrestrially-derived nitrogen from inland Korea and the Asian continent (especially China) to Uljin and the EJS. In contrast, the prevailing wind over the summer blows from the Pacific Ocean, carrying precipitation associated with relatively pristine air masses.

Overall, NO_3^- is the most abundant nitrogen species with an annual average concentration (AVG) of 58 μmol L^{-1} and a volume-weighted average (VWA) (Topol et al., 1985) of 20 μmol L^{-1}, followed by NH_4^+ (AVG = 39 μmol L^{-1}; VWA = 14 μmol L^{-1}) and DON (AVG = 30 μmol L^{-1}; VWA = 13 μmol L^{-1}) (Table 1). The reactive nitrogen concentrations in precipitation observed in this study are generally at the upper end of the range of literature values reported for coastal sites globally over the past decade (Keene et al., 2002; Luo et al., 2002; Mace et al., 2003b; Kieber et al., 2005; Calderon et al., 2007; Violaki et al., 2010; Cape et al., 2011; Zamora et al., 2011). However, much higher concentrations have been found in rainwater over coastal areas of China, which were attributed to significant pollutions associated with the rapid economic development (Chen et al., 2011; Zhang et al., 2012). The relatively high levels of reactive nitrogen in the atmosphere at Uljin probably can be linked to the anthropogenic emissions of nitrogen in East Asia including Korea and subsequent atmospheric transport.

In contrast to observations from regions characterized by intensive agricultural practices (e.g., China and midwestern USA) (Fahey et al., 1999; Zhang et al., 2008), NO_3^- showed higher abundances with respect to NH_4^+, implying that combustion processes contributed more significantly than agricultural activities to reactive inorganic nitrogen in our samples (Galloway et al., 2004; Spokes and Jickells, 2005; Lee et al., 2012). A few exceptions were observed in April, May, October, and November (Fig. 2), when concentrations of NH_4^+ were similar to or higher than those of NO_3^-. These exceptions can be attributed to enhanced agricultural activities (i.e., application of N fertilizer and livestock manure to the farmland) during these time periods in Korea (Lee at al., 2012). Although being a minor component, DON made up a considerable fraction (28% on average) of the TDN in our samples, approximating to that contributed by NH_4^+ (30% on average). In comparison with NO_3^- and NH_4^+, the relative proportion of DON showed remarkable variations from sample to sample, which were found to be relatively lower from April to August (Fig. 2). The seasonality and air mass origins might contribute to the pattern observed here to some extent. However, the major factors that control the proportion of organic nitrogen in atmospheric reactive nitrogen pool remain rather ambiguous (Cornell, 2011) (see section 3.3.1 for further discussion).

3.1.2 Potential source regions based on air mass origins

Atmospheric deposition plays an important role in global biogeochemical cycles, as it allows chemical substances to be transported to locations remote from their source regions over relatively short time scales. Therefore, the reactive nitrogen found in our samples was presumably subject to influences of air masses originating from distant areas. In order to explore the potential source regions and to assess the contributions by long-range atmospheric transport, the provenances of the air masses corresponding to each of the precipitation events were determined based on air mass back trajectory (AMBT) analysis, using the GDAS dataset and the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (version 4) developed at the Air Resources Laboratory of NOAA (Draxler and Hess, 1998). Apart from the nearby continental regions (i.e., the Korean Peninsula), reactive nitrogen could be transported over long distance to Uljin from mainland Asia as well as the surrounding oceans, as observed for other airborne species in Korea (Kim et al., 2005; Kim, 2008). Accordingly, based on calculated AMBTs, air mass origins were sorted into three groups: Korean Peninsula (Type I), Asian Continent (Type II), and Pacific Ocean (Type III) (Fig. 3). A similar classification regime can be found in our previous study (Yan and Kim, 2012), in which

detailed descriptions for each group were presented. It is noteworthy that the reactive nitrogen in each group does not exclusively come from the corresponding source region identified. For example, the precipitation events associated with long-range transported air masses inevitably receive some contributions from local sources.

The distribution patterns of VWA concentrations for the three groups were consistent for NO_3^- and NH_4^+, with the highest concentrations observed in Type II and the lowest in Type III (Fig. 4). As these two nitrogen species are known to be predominantly anthropogenic, this pattern is consistent with the air-mass origins proposed for these groups. Type II air masses also account for the largest fraction (47%) of sampled precipitation events, suggesting a significant contribution to inorganic nitrogen at Uljin from the highly industrialized and urbanized areas of East Asia, especially eastern and northeastern China (Richter et al., 2005; Jeong and Park, 2008; Lee et al., 2014). The lowest VWA DON concentration was also observed for air masses derived from surrounding marine areas (Type III), implying that marine biogenic emission is an insignificant source for DON at Uljin. In contrast to the pattern seen for inorganic nitrogen, the highest abundances of DON were associated with Type I air masses. It is thereby inferred that the emission sources within Korea play a dominant role in supplying organic nitrogen to the atmosphere at our location. Despite the fact that the largest concentrations of atmospheric organic nitrogen worldwide have been observed in China (Cornell, 2011), Type II air masses at Uljin were characterized by relatively lower DON abundances than Type I. This could be partially due to the inclusion of clean marine air masses and/or the loss of the labile portion of organic nitrogen during the long-range transport. In addition, the highest proportion of DON (32%) was also observed in Type I air masses, confirming the importance of organic nitrogen from emissions in Korea.

3.2 Source identification for DON

Whilst the major sources for inorganic reactive nitrogen in the atmosphere are known to be anthropogenic (Galloway et al., 2004; Fowler et al., 2013), the primary origins of the organic fraction remain poorly characterized (Cape et al., 2011; Cornell, 2011). Previous studies have suggested that atmospheric organic nitrogen may stem from a variety of natural and anthropogenic processes, including resuspension of soil dust, marine emissions, biomass burning, agricultural activities, industrial production, and fossil fuel combustion (Jickells et al., 2013 and references therein). In this study, attempts were made to identify the major

sources of DON in our precipitation samples, on the basis of factor analysis in combination with major ions as tracers. The results obtained from varimax-rotated factor analysis suggest that most variations (91%) in the nine variables included can be accounted for by three extracted factors (eigenvalue > 1) (Table 2).

The first factor is characterized by high loadings of Na^+, K^+, Mg^{2+}, Cl^-, and SO_4^{2-}. The enrichment factors (EFs) for these ions (except Na^+) with respect to average seawater composition were calculated using sodium as a reference element (Keene et al., 1986). The EFs fall in between 1 and 10 (Table 3), suggesting these chemical species are primarily derived from marine emissions (Poissant et al., 1994). This interpretation is confirmed by the significant positive correlations between the abundances of these ions and Na^+ (R^2 = 0.86 for K^+, 0.93 for Mg^{2+}, 0.98 for Cl^-, and 0.51 for SO_4^{2-}). Therefore, this factor probably can be attributed to the contributions from marine sources. The largest fraction of variance (50%) (Table 2) in the nine variables explained by this factor is consistent with the marine feature of Uljin. The second factor has high loadings of NO_3^- and Ca^{2+}, which are generally considered to be of different origins. In addition, moderate loadings are shown for NH_4^+ and SO_4^{2-} on this factor. In the atmosphere, NO_3^- is mainly derived from NO_2 emitted by fossil fuel combustion (Dentener et al., 2006), whereas Ca^{2+} comes from both crustal and marine sources (Gabriel et al., 2002). The relatively high EF value (19.17) indicates that crustal contribution is more important for Ca^{2+} in our samples. This factor is therefore likely to represent a mixed source involving inputs from combustion processes as well as soil resuspension. The association of these ions (i.e., Ca^{2+}, NH_4^+, NO_3^-, and SO_4^{2-}) has frequently been observed in factor analysis conducted in previous studies (Hu et al., 2003; Wai et al., 2008; Song and Gao, 2009), and is attributed to the neutralization of acidic anions by alkaline cations in precipitation (Wai et al., 2008). Since the loadings of DON on these two factors are rather low, the contributions from marine emissions, soil dust, and combustion-related processes to DON are expected to be less significant.

The third factor is associated with a moderate loading of NH_4^+ and a high loading of DON. NH_4^+ in precipitation is derived from its gaseous precursor NH_3, which is primarily released during agricultural activities such as animal husbandry and the application of synthetic fertilizer (Galloway et al., 2004). These activities are also known to be the origins of several important atmospheric organic nitrogen species (e.g., urea) (Cornell et al., 1998). According to statistical data from International Fertilizer Industry Association (2013), a considerable

fraction of the nitrogen fertilizer consumed in Korea each year is in the form of urea. In addition, Lee et al. (2012) found NH_4^+ and DON in precipitation collected in a southern Korean city were mainly derived from fertilizer use and livestock excretion in surrounding rural areas using nitrogen isotopic analysis. This suggests that agricultural practices conducted within Korea could be a crucial source of NH_3 and organic nitrogen in the atmosphere, which is in line with the conclusions drawn from AMBT analysis that the Korean Peninsula contributes significantly to NH_4^+ and DON in our samples (Fig. 4). Taken together, it is concluded that the primary fraction of DON in our samples originated from agricultural activities in Korea, albeit contributions from other anthropogenic and biogenic sources cannot be ruled out.

3.3 Biogeochemical impacts of atmospheric nitrogen deposition on the EJS

3.3.1 Seasonal variations in nitrogen fluxes and DON proportions

The wet depositional fluxes (defined as the product of concentration and precipitation depth) of the three reactive nitrogen species showed remarkable seasonal variations, with relatively high values observed in spring and fall for NO_3^- and NH_4^+, and in fall and winter for DON, respectively (Fig. 5). While being a minor component in spring and summer, DON made up the largest fraction of the TDN fluxes in fall and winter. Precipitation amount is discounted as the cause of this temporal trend as it shows no correlation with the variations in nitrogen fluxes. The seasonal variations in nitrogen fluxes are mainly attributed to the strength of emission sources located in Korea as well as other distant regions (see the discussion in Section 3.1.2).

In addition, the relative distributions of DON, TDN, and DOC fluxes were found to be in good agreement over all seasons (Fig. 5), implying there might exist an inherent link among these species. A statistically significant positive correlation (R^2 = 0.64) was observed between the proportion of organic nitrogen (ON) in total reactive nitrogen (TN) and ON to organic carbon (OC) atomic ratio in dissolved organic matter in precipitation (Fig. 6). This study is the first to point out this correlation, but analysis of a larger dataset consisting of the available literature values and data obtained at our site and another location in Korea (Seoul) shows an even stronger correlation (R^2 = 0.72) (Fig. 6). The correlation found in this dataset is significant, since the data incorporate rainwater and aerosol samples collected worldwide from geographically varied locations (e.g., urban vs. rural, terrestrial vs. marine, and

anthropogenic vs. pristine) (Jordan et al., 1995; Eklund and McDowell, 1997; Campbell et al., 2000; Kieber et al., 2005; Miyazaki et al., 2010; Gioda et al., 2011). Therefore, the trend observed herein is likely not simply coincidental, but universal.

Variations in the ON/OC ratio of airborne organic matter could reflect source variations, as being suggested by previous studies (Neff et al., 2002; Cape et al., 2011; Kanakidou et al., 2012). Specifically, the highest ON/OC ratios were observed for aerosols (total concentrations for aerosol sizes between 0.39 and 10.0 μm) over the remote North Pacific Ocean by Miyazaki et al. (2010), who also proposed that aerosols subject to increased biological influences were associated with higher ON/OC ratios. In contrast, the lowest ON/OC ratios were mostly found for dissolved organic matter in precipitation samples collected in Seoul, where atmospheric organic matter is mainly derived from anthropogenic processes, especially fossil-fuel combustion (Yan and Kim, 2012). Atmospheric processing may also affect ON/OC ratios, as being seen in different types of aerosols that undergo various physical and photochemical atmospheric processes (Sun et al., 2011). In addition, it has been suggested that organic nitrogen tends to be less effectively removed from the atmosphere than inorganic nitrogen (Cornell, 2011 and references therein). Therefore, higher proportions of organic nitrogen might be linked to higher contributions from aged aerosols characterized by elevated ON/OC ratios (Sun et al., 2011).

The organic fraction of total reactive nitrogen in the atmosphere is highly variable in time and space, lacking a consistent trend on a global scale (Cornell, 2011; Jickells et al., 2013). In addition, the bioavailability of atmospheric organic nitrogen is poorly constrained (Duce et al., 2008; Kanakidou et al., 2012). Therefore, despite the fact that organic nitrogen is increasingly recognized as a significant factor in the atmospheric deposition of reactive nitrogen, its biogeochemical impact on receiving ecosystems (especially the ocean) remains unclear. The empirical relationship found in the present study between ON/TN and ON/OC (Fig. 6) suggests that the utilization potential (evaluated using ON/OC ratios) of atmospheric deposited organic nitrogen by marine biota is positively correlated with its proportion in total atmospheric reactive nitrogen. Thus, atmospheric deposition of organic nitrogen may play a vital role in supplying bioavailable nitrogen to the surface layer of marine systems, especially when inorganic nitrogen is less abundant, as is the case for the northwestern Pacific Ocean.

3.3.2 Annual depositional fluxes of TDN at Uljin and over the EJS

The annual wet depositional flux is calculated to be 28 mmol N m^{-2} yr^{-1} for NO_3^-, 20 mmol N m^{-2} yr^{-1} for NH_4^+, and 19 mmol N m^{-2} yr^{-1} for DON, respectively (Table 4). Our estimates of inorganic nitrogen fluxes are higher than those obtained for dry deposition by Kang et al. (2010) at Donghae (Fig. 1), a coastal site adjacent to our sampling location. This is mainly due to the higher scavenging efficiency of airborne species by wet deposition comparing with dry deposition. In addition, the increasing anthropogenic emissions in East Asia may also contribute to this discrepancy (Galloway et al., 2004), as the sampling campaign by Kang et al. (2010) was conducted a decade ago. However, the fluxes of NO_3^- for both deposition modes were similar, which can be attributed to the effective dry scavenging of NO_3^- associated with coarse mode aerosols (Nakamura et al., 2005; Matsumoto et al., 2009). In general, atmospheric depositional fluxes of reactive nitrogen are higher in the coastal areas of East Asia (including the western coast of the EJS) than at other coastal locations around the world (Table 4).This is in agreement with the distribution of oceanic regions characterized by the most intensive atmospheric nitrogen deposition on a global basis (Dentener et al., 2006; Krishnamurthy et al., 2007). The considerable contributions by DON to atmospheric TDN fluxes (7–50%) at these coastal/marine sites suggest the significant role of DON in supplying nitrogen to surface waters of the ocean.

Since our sampling site is located on the eastern coast of Korea, potential losses during transport must be considered when using the results of this study as a proxy for atmospheric depositional fluxes of reactive nitrogen over the remote EJS. Oki Island (Japan), an island located downwind of Uljin in the eastern section of the southern EJS (Fig. 1), is mainly influenced by air masses originating from China and Korea, and shows a similar wet depositional flux of inorganic nitrogen to the values found in this study (45 mmol N m^{-2} yr^{-1}, Japanese Acid Deposition Survey) (Kitayama et al., 2012). This implies that losses between the coast of Korea and offshore areas are small for wet depositional fluxes, as being shown for wet depositions of nitrogen over the eastern China Sea (Zhang et al., 2010). Therefore, our estimated atmospheric wet deposition flux can be extrapolated to the offshore region in the southern EJS. By taking the dry depositional flux of inorganic nitrogen observed at Oki Island (35 mmol N m^{-2} yr^{-1} for gases plus particles in 2003–2008) (Endo et al., 2011), the total (wet plus dry) inorganic nitrogen flux was calculated to be 83 mmol N m^{-2} yr^{-1}. Further, if assuming the same DON/TDN ratio in dry deposition as in wet deposition, the total

atmospheric depositional flux of reactive nitrogen (organic plus inorganic) amounts to 115 mmol N m^{-2} yr^{-1} over the southern EJS.

3.3.3 Biogeochemical implications

Considering the prevailing westerly winds and the anthropogenic origins of these atmospheric reactive nitrogen species, their atmospheric deposition represents a source of new rather than recycled oceanic nitrogen to the EJS. Therefore, the atmospheric deposition of this nitrogen could presumably fuel the new primary production of this marine ecosystem. While inorganic nitrogen is readily utilized by marine biota, organic nitrogen is only partially available. The bioavailability of atmospheric organic nitrogen is largely determined by its chemical composition. Specifically, most reduced organic nitrogen species (e.g., amino acids, urea, and amines) can be taken up by marine microorganisms on very short timescales, whereas the organic nitrogen associated with humic-like substances is probably refractory (Bronk et al., 2007). Incubation experiments estimate the bioavailability of atmospheric organic nitrogen to be 20–80% (Peierls and Paerl, 1997; Seitzinger and Sanders, 1999; Wedyan et al., 2007). Although not explicitly measured for the DON in our samples, a significant fraction is expected to be bioavailable, since it is mostly derived from agricultural activities that are known to produce large amounts of atmospheric reduced organic nitrogen, especially urea. Moreover, the severely N-limited conditions of the EJS are likely to promote the utilization of atmospheric organic nitrogen. Assuming a bioavailability of 20–80% for DON and 100% for inorganic nitrogen, atmospheric deposition can supply approximately 89–109 mmol m^{-2} yr^{-1} of reactive nitrogen to support primary production in the southern EJS.

Using a Redfield C/N ratio of 6.625, this nitrogen flux can be converted to 7.1–8.7 g C m^{-2} yr^{-1} fixed by marine phytoplankton. This would account for 12–14% of new primary production in the southern EJS annually, which is taken to be 62 g C m^{-2} yr^{-1} based on the particulate organic carbon export fluxes reported by Hahm and Kim (2001) (using tritium and helium isotopes) and Kim et al. (2011) (using $^{234}Th/^{238}U$ disequilibrium). Our estimate is higher than those obtained by Onitsuka et al. (2009) using a coupled physical-ecosystem model (6–12% for the southern EJS from 1996 to 2003) and by Kang et al. (2010) based on field observations (ca. 10%) (Table 5). These discrepancies can be ascribed to the inclusion of organic nitrogen in our study as well as a potential increase of nitrogen deposition resulting from growing anthropogenic emissions over the past decade. On a global scale, the value

obtained for the southern EJS is generally at the higher level, comparing with other oceanic regions (Table 5). Taking into account that the southern EJS (especially the Ulleung Basin) is highly productive (Hyun et al., 2009), the estimate obtained in this study is rather remarkable. Such high estimates can be explained by the exceptionally high levels of atmospheric anthropogenic nitrogen deposition over the coastal seas downwind of East Asia (Krishnamurthy et al., 2007; Duce et al., 2008). Moreover, the contribution of atmospheric reactive nitrogen to biological productivity in the southern EJS is expected to be more pronounced during summer to fall, when the upward flux of nitrogen from the deep layer is suppressed by water column stratification (Kang et al., 2010; Kim et al., 2011b).

The results presented here might be subject to large uncertainties, such as those associated with spatial variations of depositional fluxes across the EJS, the proportions of organic nitrogen in dry deposition, and the bioavailability of organic nitrogen in the atmosphere. Nevertheless, our estimation evidently suggests atmospheric transport represents a significant source of external nitrogen input to the southern EJS. Previous studies indicate that N_2 fixation is insignificant to the nitrogen budget of this marine system (Yanagi, 2002; Kang et al., 2010). Therefore, in addition to nitrogen upwelled from below the euphotic zone and transported by the Tsushima Warm Current through the Korea/Tsushima Strait (Onitsuka et al., 2007; Yoo and Park, 2009), atmospheric deposition could sustain a considerable fraction of the nitrogen demand by primary productivity in the southern EJS, especially the offshore region. Moreover, the elevated nitrogen deposition may modify the nutrient limitation regime (Duce et al., 2008) of the EJS and thus impact the phytoplankton community structure therein (Krishnamurthy et al., 2007; Galloway et al., 2008). Due to climate warming, El Niño is likely to become more prevalent in future, which could result in suppression of the nutricline and thus restriction of nutrient upwelling from deep waters (Mackey et al., 2010). Conversely, the anthropogenic nitrogen deposition over the northwestern Pacific Ocean is predicted to increase in the near future (Duce et al., 2008). Consequently, atmospheric deposition of nitrogen is expected to play an increasingly important role in the biogeochemistry of the northwestern Pacific Ocean and the EJS over the coming decades. Special attentions should be given to the organic fraction, since little is known with respect to its sources and distributions, rendering the regulation of emission as well as the prediction on its future trend rather difficult.

4 Conclusions

Dissolved reactive nitrogen species were investigated in precipitation samples collected over a one-year period (2011–2012) at Uljin, a coastal site upwind of the southern EJS. Inorganic nitrogen was found to be mostly derived from the Asian Continent (particularly eastern and northeastern China) through long-range atmospheric transport, whereas the primary sources of DON are distributed within Korea. Furthermore, agricultural activities (e.g., animal husbandry and the application of synthetic fertilizer) were identified as the major emission source for DON. A positive correlation was found between the proportion of organic nitrogen in total reactive nitrogen and nitrogen to carbon ratio in organic matter, implying that the biogeochemical impact of organic nitrogen deposition is especially significant (i.e., more bioavailable) when inorganic nitrogen is less abundant. By combining the wet depositional flux recorded at Uljin with the dry depositional flux reported for Oki Island, the total (wet plus dry) atmospheric deposition of reactive inorganic and organic nitrogen to the southern EJS was estimated to be 115 mmol N m^{-2} yr^{-1}. This flux could potentially support 12–14% of the new primary production of the southern EJS on an annual basis, of which up to 3.4% would be attributed to organic nitrogen. The results presented in this paper emphasize the significant impact of atmospheric nitrogen deposition on the biogeochemistry of the EJS and by extension the northwestern Pacific Ocean. The development of this impact in response to climate change and growing anthropogenic emissions should be routinely monitored, especially in the case of organic nitrogen. More extensive information might be required to better understand the biogeochemical role of atmospheric nitrogen deposition, such as microorganisms community structure, the availability of other limiting nutrients (e.g., Fe and P), and the quantity and bioavailability of the insoluble fraction of organic nitrogen.

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Table 1. Statistical summary of concentrations of dissolved chemical species in precipitation analyzed in this study (unit: μmol L^{-1}).

	NO_3^-	NH_4^+	ON	Na^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	SO_4^{2-}	OC
Min.	2	1	BD[a]	8	2	2	7	13	4	9
1st quartile	12	5	7	55	8	7	18	82	23	61
Median	31	24	13	138	15	28	46	206	58	156
3rd quartile	74	34	40	900	25	108	68	1580	117	296
Max.	428	139	145	2930	69	337	259	3780	215	849
Mean	58	39	30	635	21	76	57	894	83	219
VWA[b]	20	14	13	430	13	47	25	578	43	97

[a] Below detection limit.

[b] Volume-weighted average.

Table 2. Varimax-rotated principal factor matrix. [a]

	Factor 1	Factor 2	Factor 3
NO_3^-	-0.12	**0.94**	0.25
NH_4^+	-0.12	0.59	0.46
ON	0.20	0.27	**0.93**
Na^+	**0.98**	-0.05	0.10
K^+	**0.97**	0.12	0.10
Mg^{2+}	**0.99**	0.02	0.02
Ca^{2+}	0.22	**0.95**	0.09
Cl^-	**0.99**	-0.03	0.07
SO_4^{2-}	**0.75**	0.45	0.26
Eigenvalue	4.53	2.44	1.21
% Variance	50.35	27.09	13.47

[a] The software package used for principal factor analysis is SPSS 16.0.

Factor loadings exceeding 0.7 are shown in bold.

Table 3. Enrichment factors of major ions in precipitation at Uljin relative to seawater.

	K^+/Na^+	Mg^{2+}/Na^+	Ca^{2+}/Na^+	Cl^-/Na^+	SO_4^{2-}/Na^+
Ratios in seawater	0.022	0.227	0.044	1.160	0.121
Ratios in precipitation	0.124	0.298	0.844	1.384	0.847
EF[a]	5.64	1.31	19.17	1.19	7.00

[a] The enrichment factor for a given ionic species X is calculated using the following equation:

$EF = [X/Na^+]_{precipitation} / [X/Na^+]_{seawater}$.

Table 4. Annual atmospheric deposition fluxes of reactive nitrogen (mmol N m^{-2} yr^{-1}) at coastal and marine locations estimated based on long-term measurements during the last decade.

Location	Period	Deposition Mode	NO_3^-	NH_4^+	DIN	DON	%DON	Reference
Uljin, Korea	2011-2012	wet	28	20	48	19	28	This study
Donghae, Korea	2002-2003	dry	27	6	33	–	–	Kang et al. (2010)
Taiwan	2006	dry	–	–	39	22	36	Chen et al. (2010)
Shenzhen, China	1986-2006	wet	37	58	95	–	–	Huang et al. (2013)
Singapore	2007-2008	wet	51	26	77	34	31	He et al. (2011)
		dry	19	3	21	21	50	
Crete, Greece	2003-2006	wet	10	7	17	5	23	Violaki et al. (2010)
		dry	25	2	27	17	39	
Baltic Sea	2001-2002	wet plus dry	21	19	40	4	9	Rolff et al. (2008)
Tampa Bay, USA	2005	wet	23	10	33	2	6	Calderon et al. (2007)
Barnegat Bay, USA	1999-2001	wet	29	19	48	–	–	Gao (2002)
		dry	4	2	6	–	–	
Miami, USA	2007-2009	wet	15	12	27	2	7	Zamora et al. (2011)
Puerto Rico	2004-2007	wet	6	4	10	7	41	Gioda et al. (2011)

1 **Table 5.** Fractions of new primary production supported by atmospheric nitrogen input in world oceans on an annual basis.

Ocean Regions	Deposition Mode	Nitrogen Species	Contribution to New Production	Method	Reference
Southern East/Japan Sea	wet plus dry	TDN	12–14%	field observation	This study
Southern East/Japan Sea	wet plus dry	DIN	~10%	field observation	Kang et al. (2010)
East/Japan Sea	wet plus dry	DIN	2–12%	modeling	Onitsuka et al. (2009)
Southern Yellow Sea	wet plus dry	DIN	10.5%	field observation	Lv et al. (2005)
Yellow Sea	wet plus dry	TDN	0.3–6.7%	field observation	Qi et al. (2013)
Southern East China Sea	dry	TDN	8.3%	field observation	Chen et al. (2010)
East China Sea	wet plus dry	DIN	1.1–3.9%	modeling	Zhang et al. (2010)
East China Sea	dry	DIN	0.1–9%	field observation	Nakamura et al. (2005)
South China Sea	wet plus dry	TDN	20%	modeling	Kim et al. (2014a)
Bay of Bengal	dry	TDN	up to 25%	field observation	Srinivas and Sarin (2013)
Arabian Sea	dry	TDN	<1%	field observation	Srinivas and Sarin (2013)
Southeast Mediterranean Sea	wet plus dry	DIN	8–20%	field observation	Herut et al. (1999)
Western Mediterranean Sea	wet	DIN	10–20%	field observation	Loye-Pilot et al. (1993)
Eastern Atlantic	wet plus dry	DIN	4–6%	field observation	Neuer et al. (2004)
Bermuda	wet	TDN	7–15%	field observation	Kim et al. (2014b)
World Oceans	wet plus dry	TDN	1.5–6.9%	modeling	Duce et al. (2008)
World Oceans	wet plus dry	DIN	5.1%	modeling	Krishnamurthy et al. (2010)

2

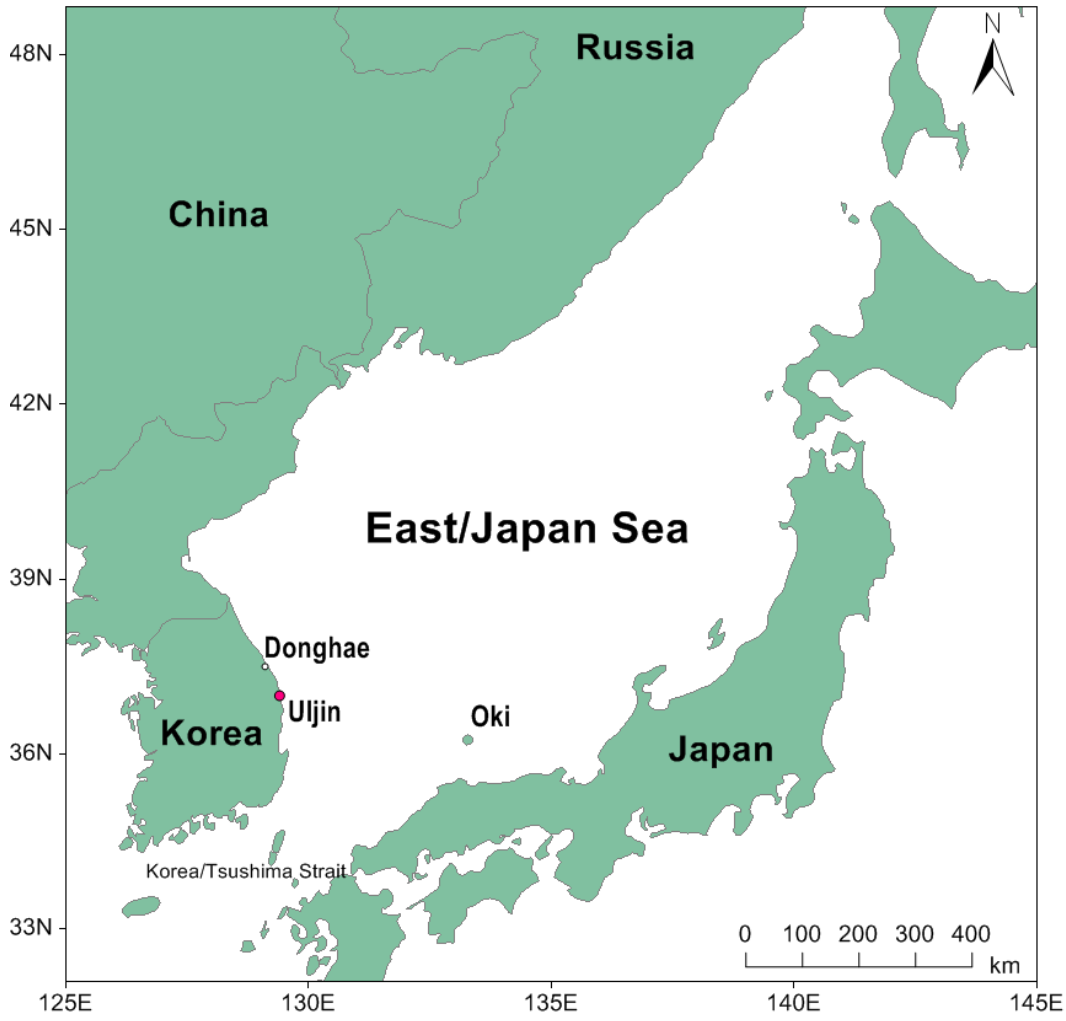

Fig. 1. Map showing the location of Uljin (sampling site, 37 ℕ, 129.4 ℇ), Donghae (37.5 ℕ, 129.1 ℇ), and Oki Island (36.3 ℕ, 133.2 ℇ).

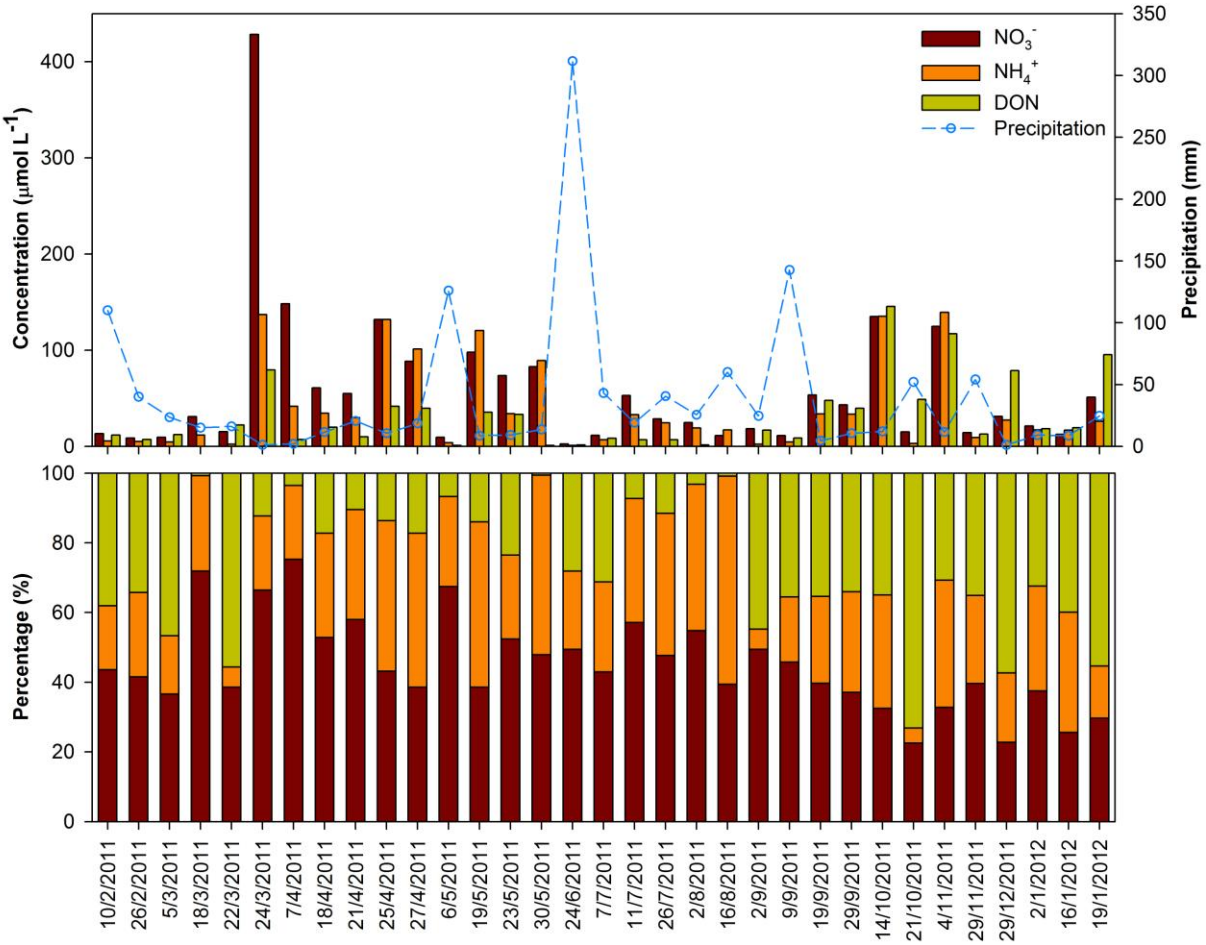

Fig. 2. Concentrations (top) and percentages (bottom) of each reactive nitrogen species in precipitation samples collected at Uljin from 2011 to 2012.

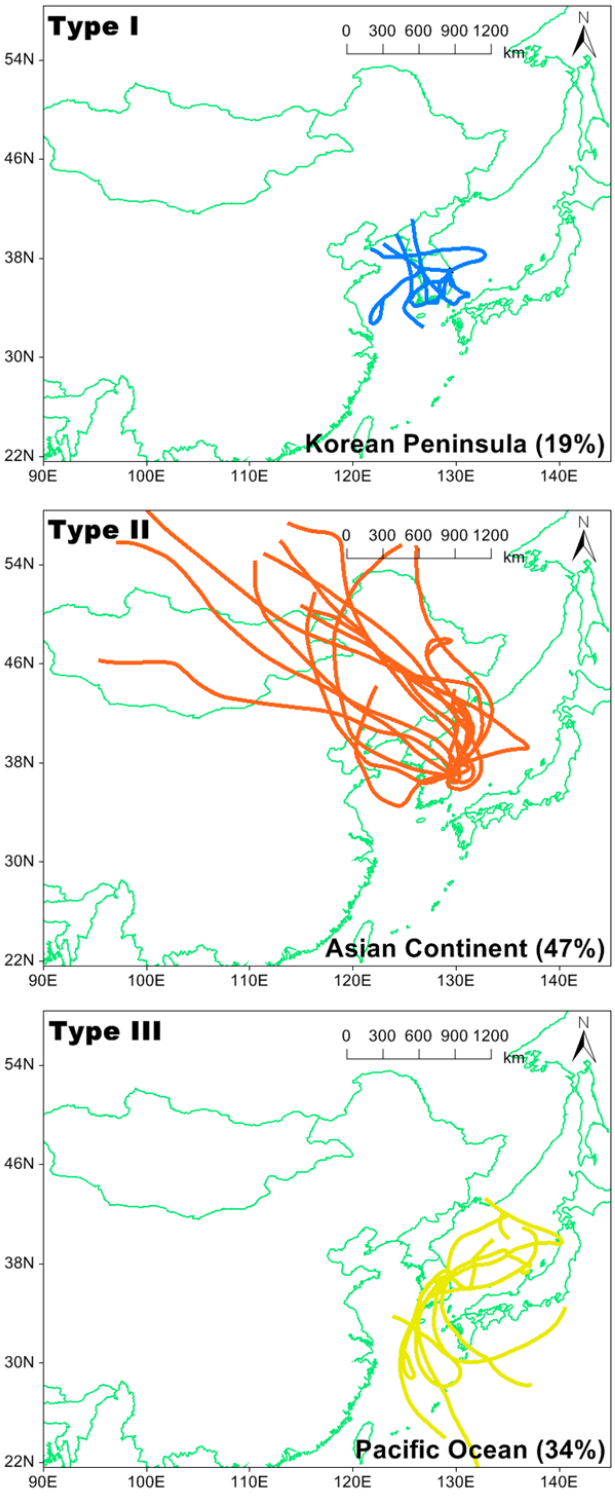

Fig. 3. Three-day air mass backward trajectories at 500 m (above ground level) for precipitation events at Uljin during the sampling period, obtained using the HYSPLIT model. The air mass provenances are sorted into three groups: Korean Peninsula (Type I), Asian Continent (Type II), and Pacific Ocean (Type III). The percentage values in parentheses indicate the occurring frequencies of each type.

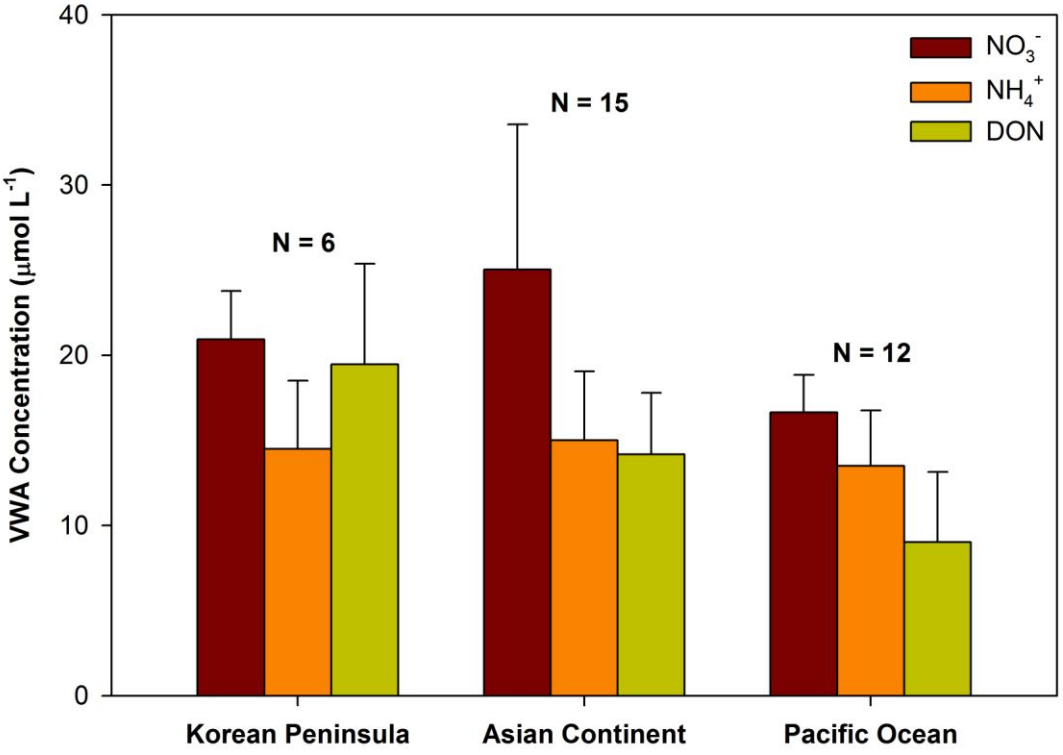

Fig. 4. Volume-weighted average concentrations of reactive nitrogen species for precipitation events associated with the three types of air mass at Uljin. Error bars represent standard deviations.

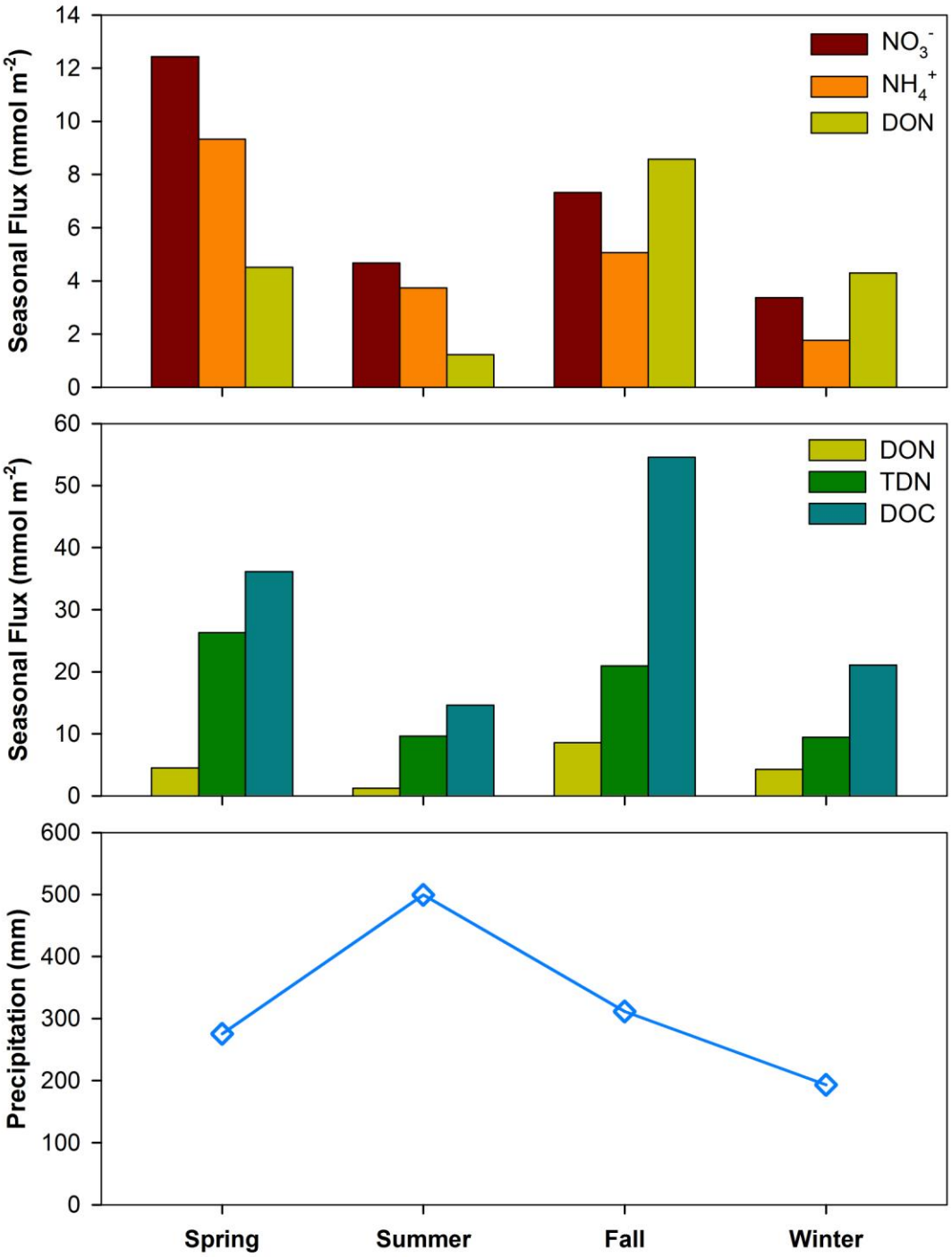

Fig. 5. Seasonal wet depositional fluxes of NO_3^-, NH_4^+, DON, TDN, and DOC at Uljin. The precipitation amounts for each season are also shown.

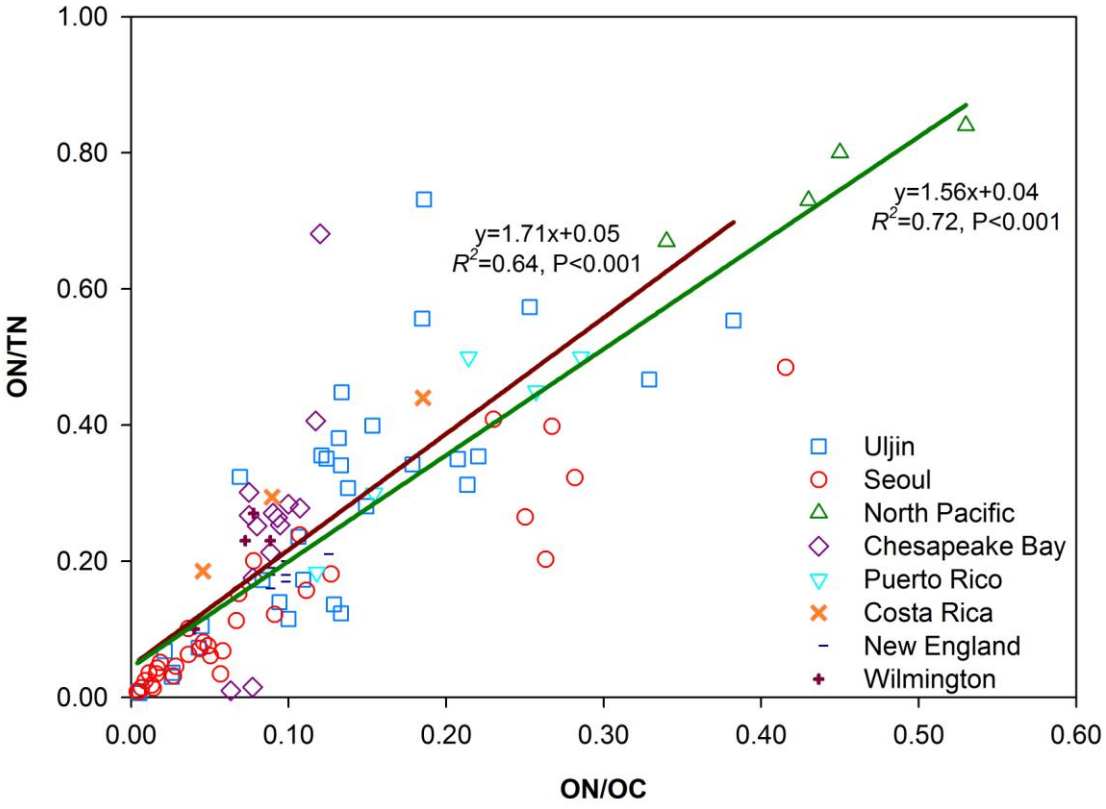

Fig. 6. Plot of ON/TN versus ON/OC in precipitation samples collected at Uljin (N = 33) and Seoul (N = 33) in Korea and other locations worldwide, including the western North Pacific (aerosols, N = 4), Chesapeake Bay (N = 14), Puerto Rico (N = 5), Costa Rica (N = 3), New England (N = 15), and Wilmington (N = 4). The solid lines denote the best-fit correlations for Uljin data (red) and the entire dataset (green).