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Long-term assessment of nitrogen deposition at remote EANET sites in Japan

Satomi Ban [a, b], Kazuhide Matsuda [a, *], Keiichi Sato [c], Tsuyoshi Ohizumi [d]

[a] United Graduate School of Agricultural Science, Tokyo University of Agriculture and Technology, 3-5-8 Saiwai-cho, Fuchu, Tokyo, 183-8509, Japan
[b] Japan Environmental Sanitation Center, 10-6 Yotsuyakami-cho, Kawasaki, 210-0828, Japan
[c] Asia Center for Air Pollution Research, 1182 Sowa Nishi-ku, Niigata-shi, 950-2144, Japan
[d] Niigata Prefectural Institute of Public Health and Environmental Sciences, 314-1 Sowa Nishi-ku, Niigata-shi, 950-2144, Japan

HIGHLIGHTS

- We assessed deposition of oxidized and reduced nitrogen from 2003 to 2012.
- We estimated dry deposition amounts by the inferential method.
- Dry deposition of oxidized nitrogen was mainly influenced by domestic emission sources.
- Reduced nitrogen deposition was mainly influenced by regional emission sources.
- Nitrogen deposition amounts remained high thorough the long period in Japanese remote area.

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ABSTRACT

Atmospheric emissions of reactive nitrogen have increased significantly on a global scale due to increases of the use of artificial fertilizer and the burning of fossil fuels. The Asian region has been identified as a high-risk area for nitrogen deposition effects on ecosystems. This paper describes a measurement-based assessment of nitrogen deposition carried out in cooperation with the Acid Deposition Monitoring Network in East Asia (EANET). The investigation aimed to understand the status and variability of dry, wet and total deposition of oxidized and reduced nitrogen over a 10-year period (2003–2012) at 8 remote sites in Japan (Rishiri, Tappi, Sado-seki, Happo, Oki, Yusuhara, Ogasawara and Hedo). Dry deposition amounts were estimated by the inferential method. All of the sites except Rishiri and Ogasawara had high mean annual total nitrogen deposition amounts of approximately 10 kg N ha^{-1} $year^{-1}$ or more, over the 10-year period. The high contribution of oxidized nitrogen deposition in the central area is mainly caused by domestic emissions, especially for dry deposition processes. An increase in reduced nitrogen deposition originating from regional emissions was found, and is likely to result in a subsequent increase in the total nitrogen deposition in Japan. Since neither a clear increasing nor decreasing trend in total nitrogen deposition was found at any site during the 10-year period, the nitrogen deposition amounts remained high thorough the long period in Japanese remote area. The spatial distribution of nitrogen deposition was found to be significant when uncertainties were accounted for.

1. Introduction

Human activities associated with food and energy consumption have caused a significant increase in reactive nitrogen emissions on a global scale, especially due to the use of artificial fertilizer and the burning of fossil fuels (Erisman et al., 2008; Galloway et al., 2008). The input of reactive nitrogen into the environment seriously influences the natural nitrogen cycle (Galloway et al., 2004). Bleeker et al. (2011) showed that nitrogen deposition was a growing issue for biodiversity in many parts of the world, particularly in Asia, and highlighted the importance of exploring the impacts at a regional or local scale. Vet et al. (2014) assessed the global distribution of deposition of major ions, including reactive nitrogen, and showed a large amount of nitrogen deposition in Asia, as well as in the United States and Europe, using output from global models. The same

* Corresponding author.
E-mail address: kmatsuda@cc.tuat.ac.jp (K. Matsuda).

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authors also carried out regional assessments of depositions utilizing monitoring networks across the world; however, there was lack of observations for dry deposition, except in North America, namely, by the Canadian Air and Precipitation Network (CAPMoN) and the Clean Air Status and Trends Network (CASTNET). It is therefore extremely important to carry out a measurement-based assessment of nitrogen deposition, including dry deposition, on regional or local scales in Asia, a region identified as a high-risk area for nitrogen deposition effects (Bleeker et al., 2011).

To estimate dry deposition, CAPMoN and CASTNET adopt indirect measurements using the so-called inferential method, which estimates dry depositions by multiplying measured concentrations by deposition velocities estimated using modeling techniques. Currently the inferential method is the most suitable technique for assessing long-term dry deposition at the regional scale, since direct measurements require highly sophisticated methods and instrumentation (Wesely and Hicks, 2000). The Acid Deposition Monitoring Network in East Asia (EANET) monitors dry deposition with the inferential method (EANET, 2010a) and estimates are available for sites in Japan. Endo et al. (2011) carried out a first attempt of dry deposition estimation by the inferential method and conducted an assessment of wet and dry deposition in Japan over a 5-year period from April 2003 using the EANET data. However there were large uncertainties related to the dry deposition estimates. In this study, we develop an assessment method that utilizes the data from EANET related to reactive nitrogen deposition, with the long-term aim of exploring the impact of nitrogen deposition on biodiversity in Asia. Using the latest knowledge on dry deposition estimation, we assess the spatial and temporal distribution of the dry, wet and total deposition of oxidized and reduced nitrogen based on the monitoring over a 10-year period, from 2003 to 2012, across remote Japanese areas.

2. Methodology

2.1. Site description

Eight remote Japanese EANET monitoring sites (Rishiri, Tappi, Sado-seki, Happo, Oki, Yusuhara, Ogasawara and Hedo) were used in the assessment (Fig. 1). According to the site criteria of EANET, remote sites are established to assess the background state of acid deposition, and should be located at a sufficient distance from significant stationary sources (such as urban areas, thermal power plants and large factories) and mobile sources (such as major highways, ports and railways) in ordered to minimize these influences (EANET, 2000). Geographic information for the 8 remote sites is shown in Table 1. The Rishiri, Sado-seki, Oki, Hedo and Ogasawara sites are located on islands. Happo and Ysuhara are located at high elevations. The main land use within a 10 km zone of all sites was forest. Although the percentage of forest around Rishiri was lower than for other sites (48%), when the shrub area (49%) was included as part of the forest surface, this increased to 97%.

2.2. Measurements of gas, particle and precipitation

The eight remote sites adopted a four-stage filter pack method for measurements of atmospheric concentrations of HNO_3, NH_3, particulate NO_3^- and NH_4^+ in total suspended particle (TSP) (EANET, 2013a). Sampling of the four-stage filter pack method was carried out biweekly at a constant flow rate of 2.0 L/min at Rishiri, Sado-seki and Ogasawara, and 1.0 L/min at other sites. The four-stage filter pack consists of a Teflon filter (Stage 1), Nylon filter (Stage 2), cellulose filter impregnated with K_2CO_3 (Stage 3) and cellulose filter impregnated with phosphoric acid (Stage 4). The particle

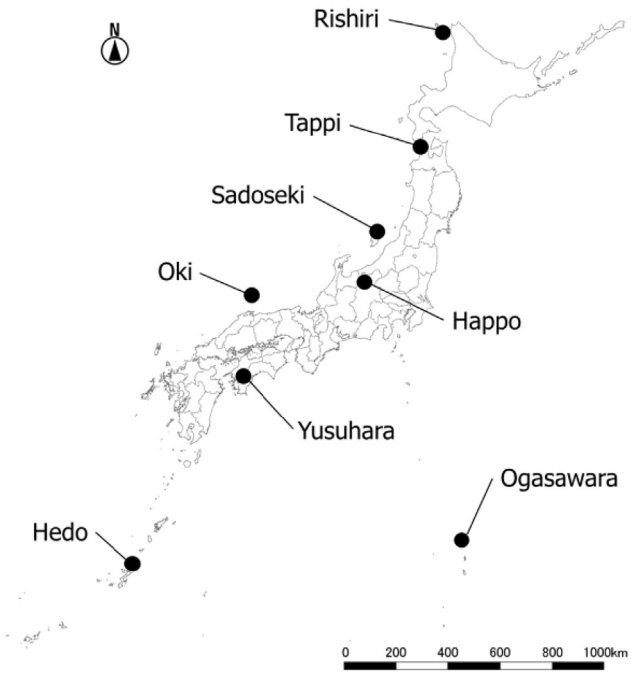

Fig. 1. Location of remote EANET sites used in this study.

components (NO_3^- and NH_4^+), HNO_3 and NH_3 were collected on the Stage 1, 2 and 4 stages, respectively. NH_3 collected on the Nylon filter (Stage 2) was also added to determine the concentration. Inorganic ions were extracted with deionized water from the filter samples, and then analyzed with ion chromatography.

A wet-only sampler was used to collect daily precipitation. Inorganic ions in the precipitation samples were also determined with ion chromatography (EANET, 2010b). All of the sites carried out meteorological observations of temperature, relative humidity, wind direction, wind speed, solar radiation and precipitation amount.

The inter-laboratory comparison activities (discussed in Section 3.3), site audit and data verification were conducted under the quality assurance/quality control program of EANET (EANET, 2000).

2.3. Estimation of deposition amount

2.3.1. Dry deposition

We use the inferential method to estimate dry deposition amounts. Dry depositions of HNO_3, NH_3, particle-NO_3^- and particle-NH_4^+ were taken into account for the total dry deposition. The inferential method estimates the dry deposition based on the following equation:

Table 1
Geographic information of the remote EANET sites used in this study.

site name	Latitude	Longitude	Elevation (m)	%-forest[a]
Rishiri	45°07′11″ N	141°12′33″ E	40	48
Tappi	41°15′06″ N	140°20′59″ E	106	93
Sado-seki	38°14′59″ N	138°24′00″ E	136	90
Happo	36°41′48″ N	137°47′53″ E	1850	75
Oki	36°17′19″ N	133°11′06″ E	90	91
Yusuhara	33°22′45″ N	132°56′05″ E	790	88
Hedo	26°51′58″ N	128°14′55″ E	60	83
Ogasawara	27°05′30″ N	142°12′58″ E	230	69

[a] %-forest means percentage of forest area within a 10 km zone of each site (except sea area).

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