Regional assessment of dry and wet deposition of reactive nitrogen in East Asia

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Abstract
In order to investigate the state of reactive nitrogen deposition in East Asia, we carried out a measurement-based assessment of nitrogen deposition on regional scale in cooperation with the Acid Deposition Monitoring Network in East Asia (EANET). We estimated the dry deposition amounts of HNO_3 and NH_3 in gas phase, and NO_3^- and NH_4^+ in aerosol phase by a modified inferential method using monthly mean inputs of meteorological data. Dry deposition amounts estimated by the modified inferential method well reproduce those estimated by using high time resolution inputs in the case of long-term total dry deposition (e.g. annual deposition). The total (dry and wet) nitrogen depositions at 20 sites in 7 countries in East Asia were in the range of 2.8 - 37 kg N ha^{-1} $year^{-1}$, and high total nitrogen deposition amounts over 10 kg N ha^{-1} $year^{-1}$ were found in wide areas of the region. The highest amount in each site classification (urban, rural, and remote) was found at Chinese sites. The ratios of dry deposition to total deposition were high in the inland areas due to the low precipitation. And the ratios of reduced nitrogen to total nitrogen deposition were relatively high in southern part of East Asia.

Key Words
oxidized nitrogen, reduced nitrogen, deposition velocity, inferential method, monitoring network

Introduction

Asia has been identified as a high-risk area for nitrogen deposition effects on ecosystems (Bleeker et al., 2011). It is therefore extremely important to carry out a measurement-based assessment of nitrogen deposition on regional scale in Asia. Recently, the state of wet deposition of reactive nitrogen such as NO_3^- and NH_4^+ in East Asia have been investigated by using data from the Acid Deposition Monitoring Network in East Asia (EANET). Although gas and aerosol components are monitored at many sites of EANET by means of a filter pack method, the state of dry deposition is still unknown except in Japan. EANET monitors dry deposition with the inferential method (EANET, 2010) and estimates are available for sites in Japan (Endo et al., 2011; Ban et al., 2016). Ban et al. (2016) estimated dry deposition of reactive nitrogen in Japan and showed the spatial distributions and the 10-year trend together with the wet deposition. In this study, we expand the area of the assessment from Japan to East Asian region by means of modified inferential method.

Methods

We estimated dry depositions of HNO_3 and NH_3 in gas phase, NO_3^- and NH_4^+ in aerosol phase, and wet depositions of NO_3^- and NH_4^+ to quantify reactive nitrogen deposition. The inferential method estimates the dry deposition based on the following equation:

$$F = - C V_d$$

where F is the dry deposition flux, C is the atmospheric concentration, and V_d is the deposition velocity. The atmospheric concentrations were monitored by a four-stage filter pack method (EANET, 2013).We applied the resistance model adopted by EANET (EANET, 2010) to estimate V_d. Basically the resistance model requires onsite and hourly meteorological data such as wind speed, temperature, relative humidity, solar radiation and precipitation amount to calculate V_d. The onsite hourly data are currently available only at Japanese EANET sites. The other sites report monthly meteorological conditions observed around the sites (e.g. EANET, 2012). In order to complement hourly meteorological data with monthly mean data at the other sites, we validated the dry deposition amounts estimated by using the monthly mean data as input to the resistance model (modified inferential method). The validation was carried out at 8 sites in Japan (Rishiri, Tappi, Sado-seki, Happo, Oki, Yusuhara, Ogasawara and Hedo) using the data in 2012.
Finally we estimated the dry deposition amounts by the modified inferential method by using the monthly mean inputs at 20 sites in 7 countries (Table 1) and evaluate the total (dry and wet) deposition amounts. In addition, the wet deposition amounts at other 6 sites in China (Table 1) were used for the assessment. Site classification (remote, rural and urban site) based on the site criteria of EANET (EANET, 2000) are also summarised in Table 1.

Results and discussion

Validation of V_d

We compared the dry depositions estimated from high time-resolution inputs (hourly meteorological data) with those estimated from monthly mean inputs (monthly meteorological data) at the 8 sites in Japan. The later monthly inputs are available at almost sites of EANET (e.g. EANET, 2012). We carried out this comparison for two cases of V_d calculations; one for forest surface and another for grass surface. In monthly dry deposition amounts, differences between the former and the latter method were in range of 0.3-36.6% (10.4% on average) for forest surface and 0.001-48.6% (9.3% on average) for grass surface, however both monthly amounts well correlated to each other. It indicates that dry depositions from monthly mean inputs well reproduces dry depositions from high resolution inputs in the case of long-term total dry deposition amounts of HNO_3, NH_3, particle-NO_3^- and particle-NH_4^+ (e.g. annual deposition).

Table 1 Location of EANET sites used in this study.

Country	Site	Class.	Latitude	Longitude
Total deposition (wet and dry)				
China	Hongwen	Urban	24°28'N	118°08'E
Indonesia	Serpong	Rural	6°15'S	106°34'E
Japan	Rishiri	Remote	45°07'N	141°13'E
Japan	Ochiishi	Remote	43°10'N	145°30'E
Japan	Tappi	Remote	41°15'N	140°21'E
Japan	Sado-seki	Remote	38°15'N	138°24'E
Japan	Ijira	Rural	35°34'N	136°42'E
Japan	Oki	Remote	36°18'N	133°11'E
Japan	Banryu	Urban	34°41'N	131°48'E
Japan	Yusuhara	Remote	33°23'N	132°56'E
Japan	Hedo	Remote	26°52'N	128°15'E
Japan	Ogasawara	Remote	27°06'N	142°13'E
Mongolia	Ulaanbaatar	Urban	47°55'N	106°55'E
Mongolia	Terelj	Remote	47°59'N	107°27'E
Russia	Listvyanka	Rural	51°51'N	104°54'E
Russia	Irkutsk	Urban	52°14'N	104°15'E
Thailand	Bangkok	Urban	13°47'N	100°32'E
Thailand	Nakhon Ratchasima	Rural	14°28'N	101°54'E
Vietnam	Hanoi	Urban	21°01'N	105°51'E
Vietnam	Hoa Binh	Rural	20°49'N	105°20'E
Wet depostion				
China	Haifu	Urban	29°37'N	106°30'E
China	Jinyunshan	Rural	29°49'N	106°22'E
China	Shizhan	Urban	34°14'N	108°57'E
China	Xiaoping	Remote	24°51'N	118°02'E
China	Xiang Zhou	Urban	22°16'N	113°34'E
China	Zhuxiandong	Urban	22°12'N	113°31'E

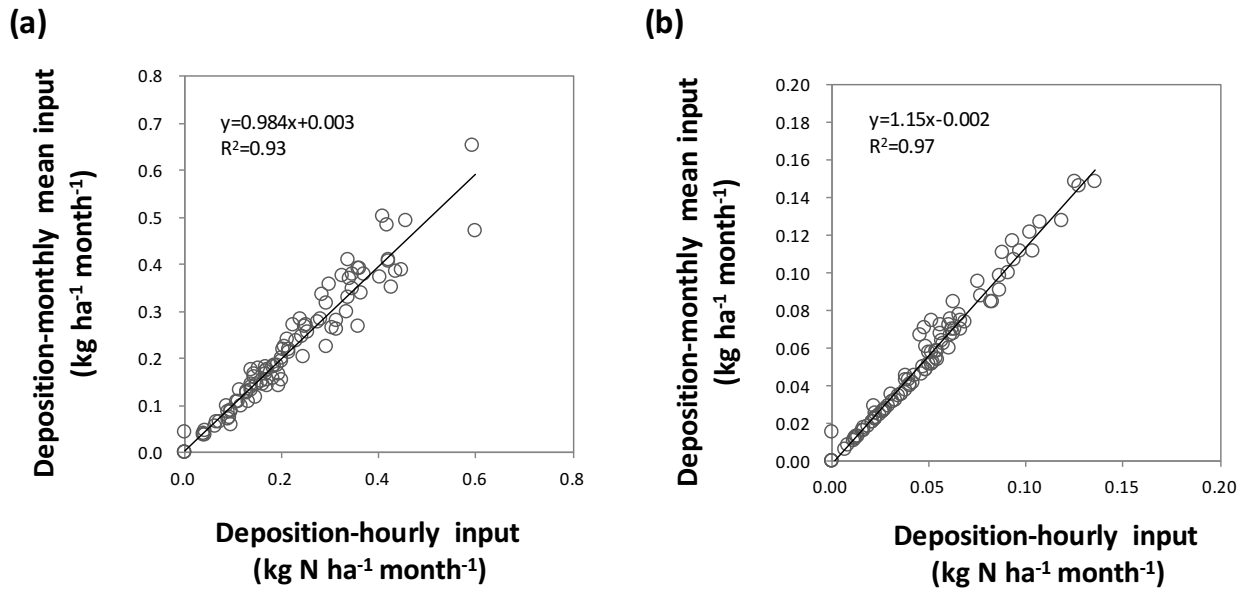

Fig. 1 Comparison between monthly dry deposition amounts estimated from high time-resolution inputs (hourly meteorological data) and those estimated from monthly mean inputs (monthly meteorological data) at 8 sites in Japan. (a) and (b) show dry deposition estimated for forest surface and grass surface, respectively.

Spatial distribution

We applied the modified inferential method using monthly mean inputs to the estimations of annual dry depositions of HNO_3, NH_3, particle-NO_3^- and particle-NH_4^+ at 20 sites in 7 countries (total deposition sites in Table 1) in 2010. A spatial distribution of total (dry and wet) deposition is shown in Fig. 2. It should be noted that the dry depositions were estimated for forest surfaces, although some sites are located in grassland and/or urban areas. At the 20 sites, the total depositions were estimated in the range of 2.8 - 37 kg N ha^{-1} $year^{-1}$ (2.8-14 at 9 remote sites, 7.1-18 at 5 rural sites and 12-37 at 6 urban sites).

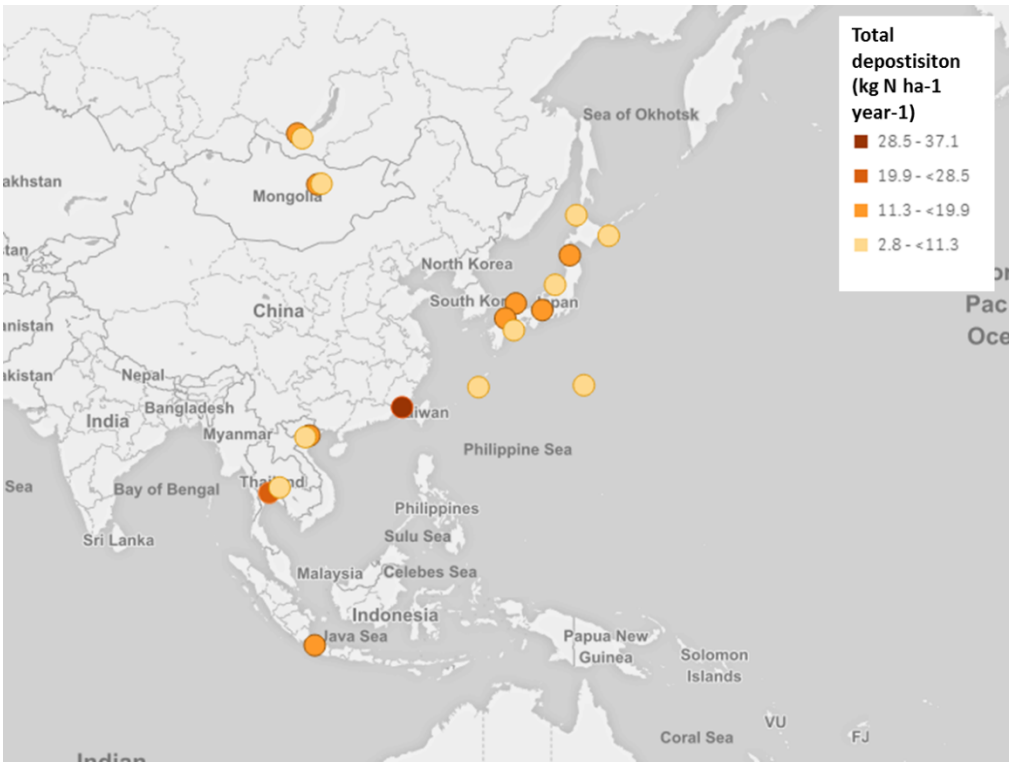

Fig. 2 Spatial distribution of the annual total (dry and wet) deposition of reactive nitrogen at 20 sites of EANET in 2010.

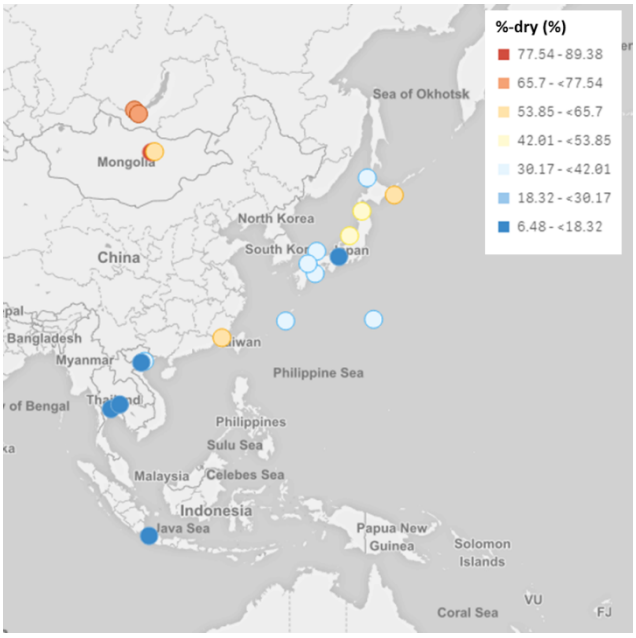

Fig. 3 Spatial distribution of percentage of dry deposition contribution to total nitrogen (%-dry) at 20 sites in EANET in 2010.

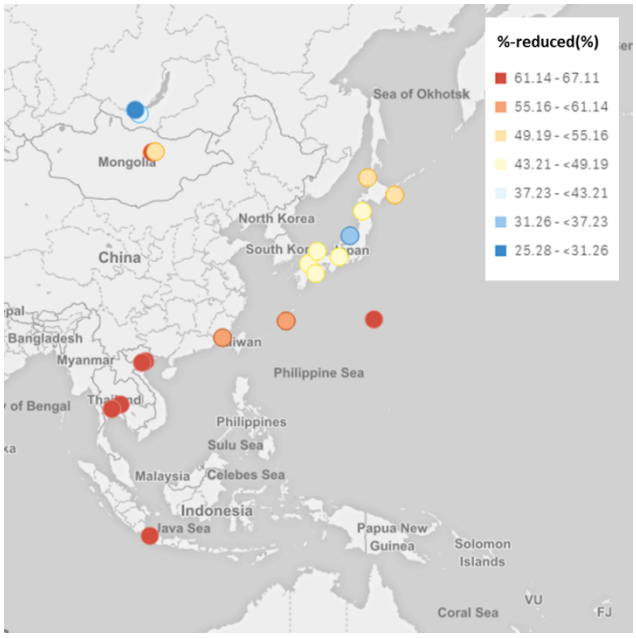

Fig. 4 Spatial distribution of percentage of reduced nitrogen deposition contribution to total nitrogen deposition (%-reduced) at 20 sites in EANET in 2010.

All sites, except 5 remote sites at Terelj, Rishiri, Ochiishi, Yusuhara and Ogasawara, showed high nitrogen deposition amounts over 10 kg N ha^{-1} $year^{-1}$. The highest total deposition amount was found at Hongwen (37 kg N ha^{-1} $year^{-1}$) in China. In the 6 sites for wet deposition in China (Table 1), high wet deposition amounts of reactive nitrogen (NO_3^- and NH_4^+) in the range of 11-30 kg N ha^{-1} $year^{-1}$ were found. The wet deposition amounts in Jinyunshan (27 kg N ha^{-1} $year^{-1}$) and Xiaoping (16 kg N ha^{-1} $year^{-1}$) were the highest among the sites in rural and remote, respectively, even though their amounts did not include dry deposition.
The ratios of dry deposition to total deposition (%-dry) (Fig. 3) were relatively high in the inland area, and low in the coastal area. The high %-dry values at the inland sites in Russia and Mongolia were caused by low wet deposition due to low precipitation. And the ratios of reduced nitrogen to total nitrogen deposition (%-reduced) (Fig. 4) were relatively high in southern part of East Asia. At Jinyunshan (rural) and Xiaoping (remote) located in southern China, the %-reduced values in the wet deposition were also high about 60%. The results agree with global model outputs that indicate the %-reduced values are high in southern part of East Asia (Vet et al., 2014). High %-reduced values in Mongolia are probably caused by the local emission.

Conclusion

In order to investigate the state of reactive nitrogen deposition in East Asia, we estimated the dry deposition amounts of HNO_3, NH_3, particle-NO_3^- and particle-NH_4^+ by the modified inferential method using monthly mean inputs. Dry deposition amounts estimated by the modified inferential method probably well reproduce those estimated by using high resolution inputs in the case of long-term total dry deposition amounts (e.g. annual deposition) at the 8 sites in Japan. The total (dry and wet) nitrogen depositions at 20 sites in 7 countries in East Asia were in the range of 2.8 - 37 kg N ha^{-1} $year^{-1}$, and high total nitrogen deposition amounts over 10 kg N ha^{-1} $year^{-1}$ were found in wide area of the region. The highest amount in each site classification (urban, rural, and remote) was found at Chinese site. The high %-dry values at the inland sites in Russia and Mongolia were caused by low wet deposition due to low precipitation. The %-reduced values were relatively high in southern part of East Asia. The high %-reduced area agreed with global model outputs.

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