Regional nitrate deposition inferred from ground- and space-based measurements

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Abstract
Spatial and temporal nitrate deposition fluxes were assessed using satellite data in the Yangtze River Delta (YRD) from 1996 to 2011. Our study reveals significant spatial variations of nitrate deposition. In general, the fluxes of total (dry plus wet) nitrate deposition was up to 22 kg N ha^{-1} yr^{-1} with large loading rates received in winter. Most high fluxes appeared over urban (38 kg N ha^{-1} yr^{-1}) and cropland (30 kg N ha^{-1} yr^{-1}) areas. During the study period (1996-2011), a significant increasing trend of nitrate deposition was observed with an annual increasing rate of 1.33 kg N ha^{-1} yr^{-1}. The spatial patterns of estimated nitrate deposition also showed that there were much higher fluxes and annual increasing trend in the middle region of YRD, i.e., the metropolitan areas contained Shanghai-Nanjing-Hangzhou cities, than in other areas. Our results also reveal that dry nitrate deposition contributed more than 50% of the total nitrate deposition over all provinces and land covers except coastal sea (14%), which indicates the relative importance of dry deposition to the total nitrate deposition in YRD region. Our study suggests that it is necessary to consider both dry and wet deposition when evaluating the influences of nitrate deposition on environment and ecosystem health.

Key words
Nitrate nitrogen, wet and dry deposition, spectral and temporal trends, satellite data, Yangtze River Delta

Introduction

With the increase of NOx emission caused by anthropogenic activities, a great amount of nitrogen deposition has been observed in China by monitoring observations (Lü 2007) and atmospheric transport models (Fabien 2013). It is essential to estimate the accurate spatio-temporal fluxes of atmospheric N deposition for assessing the effect of N deposition on terrestrial and aquatic ecosystems. Although there have been several N deposition monitoring programs and N deposition simulation experiments in China since the late 1990s (Liu 2006, Mo 2006), there are still large gaps in knowledge of the magnitude, temporal trend and spatial pattern of atmospheric N deposition on different ecosystems across China. Tropospheric NO2 columns retrieved from satellite data have been used to provide top-down estimates of surface NOx emissions via inverse modelling (Han 2009, Streets 2013), to examine specific sources (Foy 2015), to infer NOx lifetimes (Liu 2015), and to estimate surface NO2 concentrations (Lamsal 2008). These studies have provided important experiences on assessment of accuracy and reliability for using satellite data in regional scale. Both the emission and the deposition of nitrate have increased dramatically over Yangtze River Delta during the past two decades because of the expansion of industry, transportation and agriculture. In this study, the spatio-temporal variations of dry, wet and total (dry plus wet) nitrate deposition were investigated based on satellite data over the Yangtze River delta in eastern China (21° - 42° N, 108° - 124° E).

Methods

Nitrate dry deposition estimated

Dry deposition can be estimated when the atmospheric concentrations and the sedimentation velocity of gases are known. The dry deposition flux of nitrate-N is calculated as the product of the sedimentation velocity and the surface NO2 concentration which were retrieved from satellite data (Cheng 2013):

$$F_i(N_d) = V_i(N) \times C_i(N)$$

Here, Fi(Nd) is the dry deposition flux of nitrate-N; Vi(N) is the sedimentation velocity of nitrate-N over the i ecosystem; Ci(N) is the surface NO2 concentration over the i ecosystem.

Nitrate wet deposition estimated

Monthly aqueous NO- 3 concentration was retrieved from satellite data and N deposition was calculated using the following equations (Liu 2006):

$$D_{NO_3^-} = P \times C_{NO_3^-} \times 0.01$$

Here, DNO- 3 is NO- 3 deposition per month (kg N ha-1); P is the monthly mean rainfall (mm); CNO- 3 is the monthly aqueous NO- 3 concentration (mg/L).

Results

Validation of nitrate wet deposition fluxes

The result of the validation indicated that the estimated nitrate dry and wet deposition fluxes are convincing and represent good regression coefficients with the measured deposition fluxes over YRD (Figure 1). The estimated fluxes from this model are slightly higher than the measurement. The main reason is that NO- 3 in precipitation is assumed to be consistent in every precipitation in a month. Additionally the model does not account for aqueous NO- 3 concentration decreases due to continuous precipitation leaching. It is necessary to obtain the daily gridded NO- 3 concentration from satellite data with high precision and a great number of site-specific measurements are required to validate model results and make the estimate of nitrate deposition more reliable.

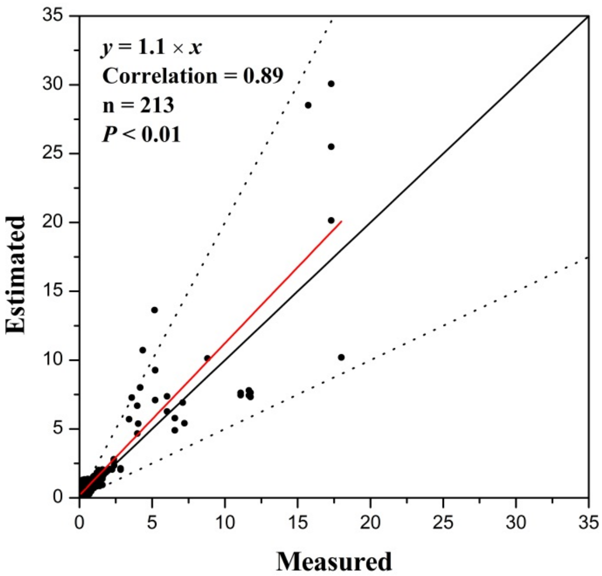

Figure 1 Comparison between measured and estimated nitrate deposition in YRD. The unit is kg N ha^{-1} per month. The dark solid line is the result of linear regression fitting for scatter plots. The dot lines indicate ± factor 1:2 and 2:1. The dash-dot line is the result of 1:1 fitting through 0.

Total annual dry and wet deposition of nitrate

The total (dry plus wet) nitrate deposition of the whole region of YRD ranged from 1.29 to 3.39 kg N ha-1 for monthly values and from 12 to 38 kg N ha^{-1} yr^{-1} for annual values during the study period (1996-2011) as showing in Table 1. When classified by provinces or land use types, total annual mean N deposition fluxes were 54, 28 and 26 kg N ha^{-1} yr^{-1} in Shanghai, Jiangsu and Zhejiang province, or 38, 30, 25, 23, and 12 kg N ha^{-1} yr^{-1} over the urban, crop, forest, inland waters and coastal sea, respectively, which reflects the different impacts of anthropogenic activities.

Table 1 Monthly and annual nitrate deposition fluxes in YRD region, provinces and different land covers in YRD

		Monthly nitrate deposition (kg N ha^{-1})			Yearly nitrate deposition (kg N ha^{-1} yr^{-1})			Ratio (%)	
		Wet	Dry	Total	Wet	Dry	Total	Wet/Total	Dry/Total
Region	YRD	0.92	1.00	1.92	10.66	11.37	22.03	47.89	52.11
Provinces	Jiangsu	0.88	1.59	2.72	10.17	18.11	28.28	35.57	**64.43**
	Shanghai	1.90	2.81	4.72	21.73	32.12	53.86	40.35	**59.65**
	Zhejiang	1.09	1.20	2.38	12.50	13.71	26.21	47.46	52.54
Land covers	Waterbodies	1.03	0.97	2.00	11.82	11.06	22.87	51.29	48.71
	Cropland	0.92	1.73	2.65	10.62	19.66	30.29	34.71	**65.29**
	Urban	1.01	2.29	3.30	11.71	26.01	37.72	30.61	**69.39**
	Coastal sea	0.92	0.15	1.08	10.66	1.76	12.42	**85.73**	14.27

Forest	0.89	1.27	2.16	12.10	10.24	22.34	**41.21**	58.79

Similarity to the monthly nitrate dry deposition, the total nitrate deposition also has a significant seasonal variation (Figure 2a). The biggest fluxes of total nitrate deposition had been observed in cold seasons (November, December and January) with the values ranging from 2.21 to 3.39 kg N ha-1. The lowest value appeared in summer (JJA) with the values ranging from 1.29 to 1.45 kg N ha-1 because of NO2 concentration which performs the highest value in winter and lowest amount in summer (Richter 2005, Cheng 2013). Obviously, the annual total nitrate deposition fluxes had been increasing as the dry and wet deposition with an increasing rate of 1.33 kg N ha^{-1} yr^{-1} from 1996 to 2011 (Figure 2 b).

Overall, the relative importance of dry vs. wet nitrate deposition to the total deposition was different in terms of the whole YRD region, the different provinces and land cover types. In general, dry and wet depositions have the similar contribution to the total nitrate deposition fluxes in the whole YRD region (Table 1). When considering different land cover types in YRD, dry deposition has main contribution to the total deposition fluxes over urban, crop and forest area while wet deposition has main contribution to the total deposition over coastal sea. It can be concluded that dry deposition had a much more important role in the total nitrate deposition in YRD, which is similar to the measured results of Liu et al. (2006) in North China Plain.

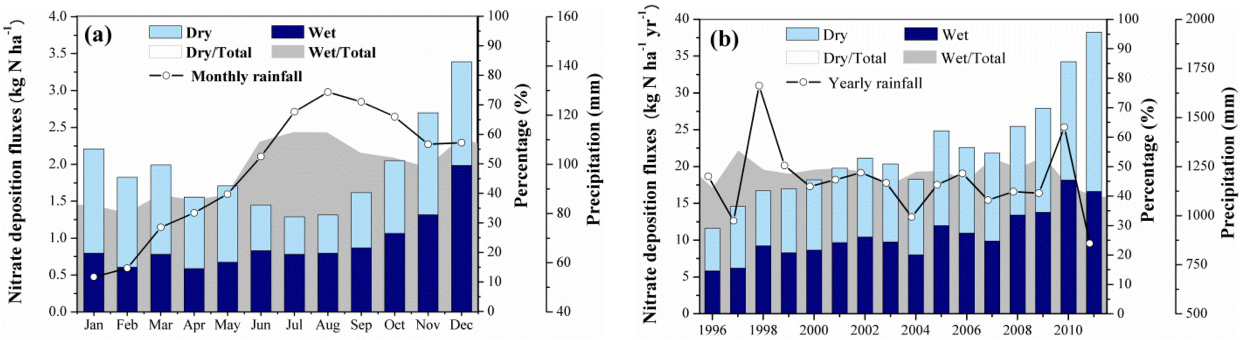

Figure 2 Monthly(a) and annual(b) average nitrate deposition, percentage of wet or dry deposition and the precipitation in YRD from 1996 to 2011

Spatial patterns of dry, wet and total nitrate deposition of Yangtze River Delta

The spatial distributions of dry, wet and total nitrate deposition over YRD were shown in Figure 3. During the study period, significantly high nitrate deposition appeared in the metropolitan area which contains the big cities of Nanjing, Shanghai and Hangzhou (Figure 3a) with the fluxes ranging from 50 to 70 kg N ha^{-1} yr^{-1}. Figure 3 b shows the spatial pattern of annual growth for nitrate deposition over YRD. The same distribution as the annual mean deposition and a large increase in nitrate deposition appeared in the middle of YRD region, especially in the metropolises and those areas along the Yangtze River with a higher annual growth rate of 3 - 5 kg N ha^{-1} yr^{-1} during the period of 1996-2011. Coastal sea, inland waters and the forest in southern part of YRD had lower fluxes of nitrate deposition with an annual average ranging from 5 to 30 kg N ha^{-1} yr^{-1}, the annual growth of the deposition flux was less than 2 kg N ha^{-1} yr^{-1}. Extremely high fluxes and large growth of nitrate deposition in the urban areas may reflect substantial air pollution due to the increased emissions from traffic, industry and domestic heating, and also has consequent effects on the eutrophication of water bodies (Liu 2011) and soil acidification in neighbouring natural and semi-natural ecosystems (Yang 2012).

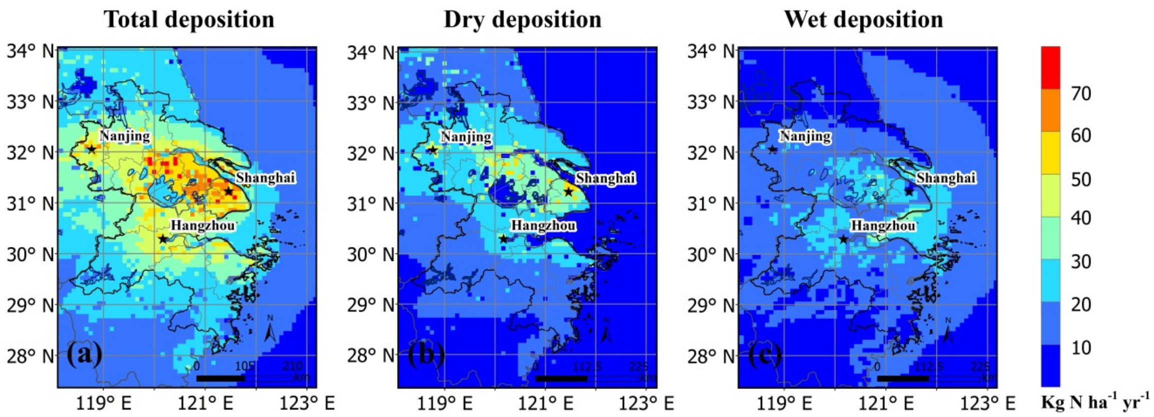

Figure 3 Annual spatial average of wet, dry and total nitrate deposition in YRD from 1996 to 2011

Conclusion

In this paper, we evaluated the estimated dry, wet and total fluxes of nitrate depositions using satellite data over YRD with values determined as 11, 11 and 22 kg ha^{-1} yr^{-1}, respectively. The largest nitrate loading was received in winter and the highest fluxes of nitrate deposition occurred over urban and crop areas, reflecting the impacts of various anthropogenic NOx emissions in different land use/cover types or regions. Dry deposition fluxes of nitrate contributed over 50% of the total nitrate deposition (22 kg ha^{-1} yr^{-1}) across all provinces and different land cover types except coastal sea (14%), indicating the importance of dry deposition to the total nitrate deposition in YRD region. During the study period (1996-2011), significant increasing trends of dry, wet and total nitrate deposition were clearly observed in YRD with annual rates increasing by 0.70, 0.63 and 1.33 kg N ha^{-1} yr^{-1}, respectively. The spatial patterns of estimated nitrate deposition also indicated that the highest value of nitrate deposition appeared in the middle region of YRD, which is a metropolitan area containing Shanghai-Nanjing-Hangzhou cities.

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