Simulation of nitrogen deposition in the North China Plain by the FRAME model

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Abstract. Simulation of atmospheric nitrogen (N) deposition in the North China Plain (NCP) at high resolution, 5 × 5 km², was conducted for the first time by the Fine Resolution Atmospheric Multi-pollutant Exchange (FRAME) model. The total N deposition budget was 1481 Gg in this region, with 77% from reduced N and 23% from oxidized N, and the annual deposition rate (47 kg N ha⁻¹) was much higher than previously reported values for other parts of the world such as the UK (13 kg N ha⁻¹), Poland (7.3 kg N ha⁻¹) and EU27 (8.6 kg N ha⁻¹). The exported N component (1981 Gg) was much higher than the imported N component (584 Gg), suggesting that the NCP is an important net emission source of N pollutants. Contributions of N deposition budgets from the seven provinces in this region were proportional to their area ratios. The calculated spatial distributions of N deposition displayed high rates of reduced N deposition in the south and of oxidized N deposition in the eastern part. The N deposition exceeded an upper limit of 30 kg N ha⁻¹ for natural ecosystems over more than 90% of the region, resulting in terrestrial ecosystem deterioration, impaired air quality and coastal eutrophication not only in the NCP itself but also in surrounding areas including the Bohai Sea and the Yellow Sea.

1 Introduction

Nitrogenous pollutant (NH₃ and NOₓ) emissions have increased sharply in the last three decades in China due to the rapid development of agriculture and industry (GAINS, 2009; EDGAR, 2011; REAS, 2006). The increased reactive N emissions damage air quality (Fowler et al., 1998; Erisman et al., 2004) and are also deposited back to surrounding terrestrial and aquatic ecosystems, contributing to severe environmental problems (Castro et al., 2002; Matson et al., 2002). Atmospheric transport models (ATMs) have been developed and applied to long-range multi-pollutant transport and distribution in Europe and North America for decades, e.g. EMEP, RADM, MATCH, and ACDEP (Jonson et al., 1998; Hertel, et al., 2002; Simpson et al., 2003; Langner et al., 2005). However, application of models to calculate N deposition in China, especially in high N deposition regions, has been rare. Dentener et al. (2006) assessed the global distribution of N deposition by different ATMs and found that China (especially eastern China) has become one of the highest N deposition regions in the world.

The North China Plain (NCP) is an intensively managed agricultural region and economically developed area. The seven major provinces on the NCP, namely Beijing, Tianjing, Hebei, Shandong, Henan, Jiangsu and Anhui, share only 8% of the total area of China but contribute 37% of the total national GDP (China Statistical Yearbook, 2009). They consume 27% of the total N fertilizer and 26% of the total energy nationally (China Statistical Yearbook, 2009), making the NCP one of the great emitters of nitrogenous pollutants (Wang et al., 2005; Zhang et al., 2010; Richter et al., 2005; Clarisse et al., 2009) and one of the high N deposition areas both nationally and globally (Zhang et al., 2008; Shen et al., 2009; He et al., 2010). Although global or continental scale studies of N deposition cover this region (Dentener et al., 2006; Holloway et al., 2002), it is impossible to describe the situation in a region with an area of 313 295 km² at resolutions from 0.5° to 10°. Kim et al. (2003) used a nested grid with a grid size of 8.9 km to model the acid deposition in the Northeast Asia, which covered parts of the NCP. Unfortunately, the simulation was conducted for the year 1996, which is difficult to apply to recent situations. Thus, it is critical to model the long-range transport and regional distribution of N concentration and deposition at higher resolution in this N deposition “hotspot”.

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The FRAME (Fine Resolution Atmospheric Multi-pollutant Exchange) model, a Lagrangian model which was originally developed in the UK, was used to simulate N deposition in the NCP. Simulation of the spatial distribution of N deposition at fine resolution, 5 × 5 km² grid, was undertaken for the year 2008. The import, export and transport of nitrogenous pollutants in the NCP were calculated. The results of the model form the essential inputs for estimation of exceedance of critical loads for N deposition and estimation of the abatement of pollutant gases required to protect natural and semi-natural ecosystems. The objectives of this study were to assess the distribution of N deposition in the NCP at 5 km resolution and to evaluate the environmental consequences of such N deposition to the NCP and surrounding regions.

2 Methods

2.1 Case study region

The domain in the study was about 10° in longitude from 112° to 122° E and 10° in latitude from 31° to 41° N, comprising 11 provinces/municipalities, i.e. Beijing, Tianjin, Hebei, Henan, Shandong, Shanxi, Anhui, Jiangsu, Hubei, Liaoning, Inner Mongolia, as well as parts of the Bohai and Yellow Seas, bordering and within the model domain but may not belong to the NCP. The location, boundary and land-use of the NCP are outlined in Fig. 1, covering parts of Beijing, Tianjin, Hebei, Henan, Shandong, Anhui and Jiangsu provinces (or municipalities). The total area of the NCP is 313 300 km², 39 % of the model domain area.

2.2 Model application

The FRAME model is an atmospheric transport model used for modeling the long-range transport and annual deposition of NH₃ (NH₃ and NH₄⁺), NOₓ and oxidized sulphur (SOₓ). It uses statistically weighted straight-line trajectories and is a multi-layer, Lagrangian model with high horizontal (5 × 5 km²) and vertical (33 layers) resolution. The model is described in detail by Singles et al. (1998); Fournier et al. (2004) and Dore et al. (2007).

Spatial distributions of NH₃, NOₓ and SO₂ emissions at 5 × 5 km² input into the model are shown in Fig. 2. An NCP NH₃ emission inventory and map for the year 2008 were calculated following the methodology adopted by Zhang et al. (2010) for the year 2004. The NOₓ emission inventory was estimated from the census data and the emission factors cited from Kato and Akimoto (1992). The SO₂ emission inventory was cited from the China Statistical Yearbook (2009). Both the NOₓ and SO₂ emission inventories were allocated onto the most suitable land use type by the bottom-up process. Primary census databases used for the estimation of emission inventories of NH₃ and NOₓ were obtained from the China Statistical Yearbook (2009). The total emission of

NH₃-N for the model domain was 3500 Gg, 15 % higher than the result estimated (3100 Gg) by REAS for the year 2003 and consistent with the result (3300 Gg) projected by REAS for the year 2008 (REAS, 2006). Total emissions of NOₓ-N and SO₂-S for the model domain were 2000 Gg and 4700 Gg, respectively, without significant differences among the results estimated by REAS, 2100 Gg NOₓ-N and 5000 Gg SO₂-S for the year 2006 (CGRER, 2007).

Emissions of NH₃, NOₓ and SO₂ outside the NCP domain were taken from a national scale emissions map (cited from REAS database), gridded at 50 km resolution. Boundary conditions for air concentrations of aerosols for the regional (5 km resolution) NCP simulation were calculated with a larger scale simulation at 50 km resolution over the whole of China.

The wind roses employed in the FRAME model used 6-hourly operational radiosonde data from the 6 stations (Fig. 3), spanning a ten-year period (1997–2007) to establish the frequency and harmonic mean wind speed as a function of direction for the NCP. Radiosonde data were obtained from the British Atmospheric Data Centre (2008).

The precipitation and land use data employed in the model were published by the Institute of Geographic Sciences and
2.3 Comparison between modeled and measured results

A comparison with measurements from monitoring sites on the NCP was carried out to evaluate the performance of the model. Because of the lack of monitoring data of gases (NH$_3$) and aerosols (NH$_4^+$, NO$_3^-$, and HNO$_3$), it was difficult to fully quantify the uncertainties from dry deposition. NO$_2$ concentrations were only available at 13 monitoring sites in 2008 (China Statistical Yearbook, 2009). Measured wet deposition data were available at 13 monitoring sites in the NCP in 2008 (reduced N deposition at 13 sites and oxidized N deposition at 12 sites). Volume weighted annual mean concentrations of NH$_4^+$ and NO$_3^-$ from precipitation were selected to validate the model. With the present information, modeled NH$_4^+$, NO$_3^-$ concentrations in precipitation and NO$_2$ concentrations in air were compared against measured results for the year 2008 (Fig. 4), and the model evaluation statistics were listed in Table 3. Linear regressions with non-zero intercepts were conducted and displayed higher intercepts for NH$_4^+$ and NO$_3^-$ than those in the simulation in the UK (Dore et al., 2009), respectively, possibly related to fewer data distributed in the low concentration ranges in the NCP. Although the correlations were statistically significant ($p < 0.05$), they could not be claimed to explain the real situation. Linear regressions with zero intercepts were also conducted. In these comparisons, modeling results of NH$_4^+$ fitted the measured data well with a regression coefficient of 1.05 (Fig. 4a). However, modeling results for NO$_3^-$ were significantly underestimated with a regression coefficient of only 0.63 (Fig. 4b). Underestimation of measured nitrate wet deposition by models has also been reported by other authors (Dore et al., 2007; Chemel et al., 2010). The more complex chemical reactions associated with the formation of nitrate aerosol may mean that wet deposition of this chemical species is more difficult to simulate accurately than wet deposition of ammonium. Some dry deposition of nitric acid onto the surface of the rain collector may have occurred, though assessment of this has not been reported.
in the literature. Also, N deposition derived from shipping emissions of NOx was not included here. Relatively robust regressions were displayed for NO\textsubscript{x} concentrations in both the regressions. Modeled concentrations of NO\textsubscript{2} were higher than measured results with coefficients of 1.10 and 1.23 for the two regressions, respectively (Fig. 4c), which further supports the hypothesis for the underestimation of NO\textsubscript{3}− concentration. Moreover, points located between 2:1 and 1:2 reference lines were analyzed. 92% of the model results for NH\textsubscript{4}+ were accordant within a factor of two times the measurements, 100% of modeled results for NO\textsubscript{3}− were within a factor of two times the measurements, and 80% of modeled results for NO\textsubscript{2} were within a factor of two times the measurements. A model is generally considered fit for purpose if more than 50% of the points fall within the 1:2 and 2:1 lines. Combined with the regression coefficient, this analysis implies that the FRAME model was effective in capturing the spatial variability of N deposition over the NCP.

3 Results and discussion

3.1 Budget of N deposition in the NCP

Budgets of NH\textsubscript{x} and NO\textsubscript{x} from dry and wet deposition over the NCP in the year 2008 were calculated by the FRAME model. Results for import, export, emission, and deposition of materials are listed separately in Table 1. In total, 1484 Gg N were deposited in this region from both dry and wet deposition, with an average N deposition of 47 kg N ha\textsuperscript{-1} yr\textsuperscript{-1}. N deposition was previously calculated in both the UK and Poland by the same model (Dore et al., 2009; Kryza, 2009). It was calculated that the same budget for the UK in the year 2005 by FRAME was 319 Gg (Dore et al., 2009). Compared to the result above, the total deposition of N in the NCP was 4.7 times that in the UK, while the area ratio of the NCP to the UK is only 1.3. Thus, the average N deposition rate at grid cell in the NCP was 3.6 times that in the UK. The NH\textsubscript{3} deposition budget for Poland calculated by the same model was 227 Gg NH\textsubscript{3}− N in 2002 (Kryza, 2009). Modeled NH\textsubscript{3}− N deposition was 1143 Gg in the NCP in our study. Taking the area ratio of the two regions into account, the average deposition rate of NH\textsubscript{3}− N at grid cell in the NCP was about 5 times that in Poland. The EMEP model calculated the total N deposition in the EU27 countries to be 3714 Gg in the year 2008 (EMEP, 2011), equal to an average deposition rate of 8.6 kg N ha\textsuperscript{-1} yr\textsuperscript{-1} in the EU 27 countries and less than 20% of that in the NCP.

Reduced N deposition was higher than oxidized N deposition in the NCP, illustrating higher NH\textsubscript{3} emissions from agricultural sources than NO\textsubscript{x} emissions from industrial or traffic sources (Bouwman et al., 1997; Streets et al., 2000). Average NH\textsubscript{x}−N to NO\textsubscript{x}−N ratio was 3.4 with ratios less than 2.0 found only in the Beijing and Tianjin municipalities, which are more highly developed regions with more NO\textsubscript{x} emissions from traffic and industrial sources. The domination of NH\textsubscript{x}−N was consistent with observed results (Lü and Tian, 2007). However, the lower oxidized N ratio did not correspond to less deposition than in other regions. Average NO\textsubscript{3}− N deposition was 11 kg N ha\textsuperscript{-1} yr\textsuperscript{-1}, comparable to the modeled total N deposition in the UK (Dore et al., 2009). High tropospheric NH\textsubscript{3} and NO\textsubscript{2} concentrations over this region have been observed from space by remote sensing (Richter et al., 2005; Clarisse et al., 2009). Both the high NH\textsubscript{3}−N and NO\textsubscript{3}−N deposition rates may be attributed largely to China’s rapidly developing economy, especially increased fertilizer N application and rapidly increasing energy consumption over recent years (China Statistical Yearbook, 2009).
Table 1. Mass budgets of emission, import, export and deposition of N in the NCP.

<table>
<thead>
<tr>
<th>N species</th>
<th>Import (Gg)</th>
<th>Emission (Gg)</th>
<th>Dry deposition (Gg)</th>
<th>Wet deposition (Gg)</th>
<th>Total deposition (Gg)</th>
<th>Sea deposition (Gg)</th>
<th>Export (Gg)</th>
<th>Export/Import ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>NH₄-N</td>
<td>283</td>
<td>2346</td>
<td>316</td>
<td>827</td>
<td>1143</td>
<td>278</td>
<td>1206</td>
<td>4.27</td>
</tr>
<tr>
<td>NO₂-N</td>
<td>301</td>
<td>899</td>
<td>150</td>
<td>190</td>
<td>340</td>
<td>82</td>
<td>776</td>
<td>2.58</td>
</tr>
<tr>
<td>Total-N</td>
<td>584</td>
<td>3245</td>
<td>466</td>
<td>1018</td>
<td>1484</td>
<td>360</td>
<td>1981</td>
<td>3.40</td>
</tr>
</tbody>
</table>

Fig. 5. Contributions of areas (a) and N deposition budgets (b) in the NCP at province level (only the areas and N deposition budgets of the seven provinces within the NCP were included; areas and N deposition budgets of the seven provinces outside the NCP were omitted).

High N deposition was not restricted to the NCP, but also influenced the surrounding area by the long-distance transportation of N-compound pollutants. The exported N component (1981 Gg) was 3.4 times the imported N component (584 Gg), demonstrating the important pollution source effect of the NCP. Of the total N deposition budget in the NCP, 22%, 19%, 26%, 14%, 13%, 3% and 3% were derived from Hebei, Shandong, Henan, Jiangsu, Anhui, Beijing and Tianjin, respectively. Contributions to the N deposition budgets from the seven provinces were nearly proportional to the area percentages of the individual provinces to the NCP (Fig. 5).

3.2 Spatial distribution of N deposition in the NCP

Emitted N-compounds are deposited onto terrestrial and aquatic surfaces in the form of dry and wet deposition. Chemical reactions take place in the atmosphere in both the aqueous and dry phases, and species of NH₃, NH₄⁺, NO, NO₂, NO₃⁻, PAN and HNO₃ are produced during the emission, transport and deposition processes (S-compounds are also produced but not discussed here). Concentrations of N-compounds, NH₃, NH₄⁺, NOₓ, NO₃⁻ and HNO₃, and deposition rates of reduced and oxidized N from dry and wet deposition are mapped at 5 × 5 km² resolution in Figs. 6 and 7.

The distribution of the modeled NH₃ concentration was correlated to the distribution of NH₃ emission (Figs. 2a and 6a), which can be attributed to the short-distance transport of NH₃ (Asman et al., 1998). In contrast to NH₃, emitted NO₂ has a longer lifetime and deposition occurs far from the sources. Hotspots of the NO₂ emissions extended to larger areas by long-distance transport, resulting in greater regions, even the coastal regions, sharing higher gaseous NO₂ concentrations (Fig. 6b). NH₄⁺ and NO₃⁻ ions are mostly formed through gas to aerosol conversion forming fine (NH₄)₂SO₄, NH₄HSO₄ and NH₄NO₃ particles. These aerosols are removed principally by wet deposition. Relatively higher concentrations of aerosol NH₄⁺ and NO₃⁻ were found in the northern area where lower precipitation and reduced aerosol removal rates occur (Fig. 6c, d). Higher concentrations of HNO₃ were distributed in the areas with lower NH₃ concentration, while lower concentration of HNO₃ occurred in the areas with higher NH₃ concentration (Fig. 6e). This can be explained by the increased production of NH₄NO₃ from the gaseous precursors NH₃ and HNO₃.

The dry deposition rates of reduced and oxidized N were associated with both their concentrations and their deposition velocities. The dry deposition of reduced N exhibited higher rates in areas around rather than within the NCP (Fig. 7a), which could be attributed to land use (Fig. 1c). The NCP is an intensive agricultural region dominated by farmland, whereas it is surrounded by mountains in the west and the
Fig. 6. Modeled concentrations of N-containing pollutants in the NCP in 2008 at 5 × 5 km² resolution (a. NH₃; b. NOₓ; c. NH₄⁺; d. NO₃⁻; e. HNO₃).

Fig. 7. Modeled deposition of N-containing pollutants in the NCP in 2008 at 5 × 5 km² resolution (a. NHₓ-dry; b. NOᵧ-dry; c. NHₓ-wet; d. NOᵧ-wet; e. N-total).
north with forest and grassland. NH$_3$ deposition velocity is connected with bulk canopy resistance varying with vegetation. Bulk canopy resistance increases in regions of farmland and improved grassland, causing dry deposition velocity decreases (Singles et al., 1998; Smith et al., 2000). This explains the higher dry deposition of reduced N at the western and northern boundaries. With less influence from land use, oxidized N deposition was better correlated to the NO$_2$ concentration (Figs. 2b and 7b). High deposition rates of reduced N and oxidized N occurred in the south and the east, respectively (Fig. 7c, d), attributed to both the high aerosol concentrations and the high precipitation. The total N deposition is illustrated in Fig. 7e. Total N deposition in the NCP at 5 × 5 km$^2$ cell grid generally ranged from 20 to 80 kg N ha$^{-1}$ yr$^{-1}$. Hotspots appeared in the southern NCP with N deposition higher than 80 kg N ha$^{-1}$ yr$^{-1}$, consistent with the global spatial distribution of N deposition (Dentener et al., 2006).

### 3.3 Comparison between modeling results and observations

The modeled averages of N wet and dry deposition were 32 and 15 kg N ha$^{-1}$ yr$^{-1}$, respectively. Wet N deposition was consistent with results in the NCP in the 2000s (Zhang et al., 2008). However, dry deposition of N was much lower than results obtained from monitoring results (Shen et al., 2009). Such difference could be attributed by following reasons. Firstly, no measured deposition velocities of gases and aerosols have been available in this region until now. Shen et al. (2009) cited much higher deposition velocities than those applied in the FRAME model (Table 2). Secondly, NH$_3$ is a bi-directional exchange species and only net fluxes were considered in the model, which may underestimate the N dry deposition because the model did not distinguish between the two deposition and emission processes for NH$_3$. Shen et al. (2009) used a simple NH$_3$ compensation point (5 µg N m$^{-3}$) value to estimate dry deposition of reactive N species (assuming dry deposition occurred only when NH$_3$ concentration was higher than 5 µg N m$^{-3}$) in their calculation, which may also lead to some uncertainties due to the absence of a stable NH$_3$ compensation point during the winter wheat and summer maize double cropping system. Therefore there is some need to improve both the monitoring and the simulation of N dry deposition in the NCP.

### 3.4 Potential exceedances

High N concentration was found in the NCP, at a level which is known to cause adverse effects to ecosystems by acidification and eutrophication in the North America and Europe in spite of few studies of critical levels available in this region until now. Cape et al. (2009) reviewed the critical level of NH$_3$, proposing 1 µg m$^{-3}$ for bryophytes and lichens and 3 µg m$^{-3}$ for all other plant species instead of the older critical level of 8 µg m$^{-3}$ for vegetation in Europe. If 3 µg m$^{-3}$ was applied in our study, ammonia concentrations in 99.3% of the areas in the NCP would exceed the critical level (Fig. 8a). Bobbink et al. (2010) summarized empirical critical loads for various terrestrial ecosystems in Europe and the US with a range of 5–30 kg N ha$^{-1}$ yr$^{-1}$ for forests and grasslands. Experiments in temperate deciduous forests and temperate grasslands in China showed similar critical loads of 10–30 kg N ha$^{-1}$ yr$^{-1}$ (Pan et al., 2005; Zhao et al., 2008). Even taking the upper limit, N deposition in 99.7% of the areas of the NCP exceeded the critical load (Fig. 8b).

However, the NCP is an intensive agricultural area dominated by farmland which is nutrient tolerant. Liu et al. (2011) estimated and mapped critical loads in China based on the steady state mass balance (SSMB) method and data from the literature, which resulted in a very high critical load, more than 200 kg N ha$^{-1}$ yr$^{-1}$, for the agricultural area in the NCP. As 92% of the area is arable land in the NCP (including overlaps with forest, grassland and urban areas), this indicates that N deposition in nearly all the areas was under the critical limit, in complete contrast to initial calculations with exceedances estimation at a critical load of 30 kg N ha$^{-1}$ yr$^{-1}$. However, this does not suggest that the current N deposition level in the NCP is completely safe. First of all, the critical loads and exceedances discussed above were based on a theoretical calculation. The cereal and vegetable crops have a high tolerance of eutrophication and the calcareous soil in the NCP has a high tolerance of acidification. The properties of both vegetation and soil determined the high critical load. Actually, agricultural ecosystems in the NCP had already been artificially N saturated with excessive N.

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**Table 2.** Dry deposition velocities of N-compound pollutants cited in Shen et al. (2009) and in the FRAME model (unit: mm s$^{-1}$)

<table>
<thead>
<tr>
<th>Species</th>
<th>Value cited by Shen et al., 2009</th>
<th>Value used in the FRAME model</th>
</tr>
</thead>
<tbody>
<tr>
<td>NH$_3$</td>
<td>7.4</td>
<td>Canopy resistance calculation</td>
</tr>
<tr>
<td>NO$_2$</td>
<td>5.9</td>
<td>Vegetation specific</td>
</tr>
<tr>
<td>HNO$_3$</td>
<td>20</td>
<td></td>
</tr>
<tr>
<td>NH$_4^+$</td>
<td>2.4</td>
<td></td>
</tr>
<tr>
<td>NO$_3^-$</td>
<td>2.4</td>
<td></td>
</tr>
</tbody>
</table>

**Table 3.** Model evaluation statistics for NH$_4^+$, NO$_3^-$ and NO$_2$ concentrations.

<table>
<thead>
<tr>
<th>Species</th>
<th>n</th>
<th>FAC2</th>
<th>MB</th>
<th>MGE</th>
<th>NMB</th>
<th>NMGE</th>
<th>RMSE</th>
<th>r</th>
</tr>
</thead>
<tbody>
<tr>
<td>NH$_4^+$</td>
<td>13</td>
<td>0.92</td>
<td>0.47</td>
<td>1.13</td>
<td>0.14</td>
<td>0.35</td>
<td>1.45</td>
<td>0.35</td>
</tr>
<tr>
<td>NO$_2$</td>
<td>12</td>
<td>1.00</td>
<td>−2.41</td>
<td>2.50</td>
<td>−0.35</td>
<td>0.37</td>
<td>7.00</td>
<td>0.07</td>
</tr>
<tr>
<td>NO$_3^-$</td>
<td>13</td>
<td>0.77</td>
<td>1.00</td>
<td>16.35</td>
<td>0.03</td>
<td>0.52</td>
<td>21.98</td>
<td>0.30</td>
</tr>
</tbody>
</table>
accumulation, nitrate leaching, ammonia volatilization and N₂O emissions (Ju et al., 2009). The simulated critical load of N, 200 kg N ha⁻¹ yr⁻¹, was a safe value consistent with the optimum total N inputs in the agricultural ecosystems in the NCP rather than a real critical load of N deposition in this region. Actual N fertilizer application rate in the NCP is double or triple the optimum value for the agroecosystems (Zhao et al., 1997). Even the lowest N deposition (17 kg N ha⁻¹ yr⁻¹ in the simulation in this study) would still further stimulate environmental pollution related to reactive N. In addition, natural regions surrounding the NCP were exposed to high N deposition for the diffusion of nitrogenous pollutants from intensive emission sources, e.g. Miyun Reservoir, a protected area for the drinking water source of Beijing (Fig. 1b), the total N concentration of the water was around 1.0 mg L⁻¹ in 2000s (China Environmental Statistical Yearbook, 2008). The difference in the inorganic N concentrations between rainwater and seawater indicates that precipitation greatly enriched the N concentration of the seawater in the coastal regions ranged from 0.002 to 5.310 mg L⁻¹, 0.308 mg L⁻¹ on average, with 27.5 % of samples exceeding the critical level (0.3 mg L⁻¹) in 2008 (Coastal Environmental Quality Yearbook, 2008). The difference in the inorganic N concentrations between rainwater and seawater indicates that precipitation greatly enriched the N concentration of the seawater. N deposition derived from shipping emissions of NOₓ was not included here. This has been demonstrated to be highly significant in other regions (Dore et al., 2007). If these emissions were taken into account, the N concentration and deposition would be even higher than the current modeled results. According to data published by the State Oceanic Administration, there were 79 red tides on average in the year 2000s, while there was only one every five year in the 1960s. Kim et al. (2011) reported that significant increasing N abundance driven by N deposition in China kept increasing in the last three decades without any downward trend observed by now (Reis et al., 2009; Liu et al., 2011). And it is much more difficult to depress the concentrations of NH₃ than the concentrations of NOₓ, e.g. the NOₓ emissions were significantly lowered (Wang et al., 2010) but there was no significant decrease in the NH₃ concentrations in Beijing during the 2008 Olympic Games (Shen et al., 2011).

Last but not least, high N deposition affected not only the terrestrial ecosystems but also the offshore regions. In the export budget, 18 % of the N was deposited onto the coastal region (Table 1), corresponding to 22 kg N ha⁻¹ being directly deposited to the surface. Assuming that the total N deposition in the coastal region originates from precipitation, the volume weighted N concentration was as high as 4.4 mg L⁻¹ in our studies. It was reported that the inorganic N concentration of the seawater in the coastal regions ranged from 0.002 to 5.310 mg L⁻¹, 0.308 mg L⁻¹ on average, with 27.5 % of samples exceeding the critical level (0.3 mg L⁻¹) in 2008 (Coastal Environmental Quality Yearbook, 2008). The difference in the inorganic N concentrations between rainwater and seawater indicates that precipitation greatly enriched the N concentration of the seawater. N deposition derived from shipping emissions of NOₓ was not included here. This has been demonstrated to be highly significant in other regions (Dore et al., 2007). If these emissions were taken into account, the N concentration and deposition would be even higher than the current modeled results. According to data published by the State Oceanic Administration, there were 79 red tides on average in the year 2000s, while there was only one every five year in the 1960s. Kim et al. (2011) reported that significant increasing N abundance driven by N deposition in the Northwestern Pacific Ocean, especially in the East China Sea. As the simulated results in this study, high N deposition has become one of the main contributors to coastal eutrophication in China.
4 Conclusions

Simulation of atmospheric N deposition in the NCP, a global hotspot for N deposition, was undertaken using the FRAME model for the first time at a much higher resolution (5 × 5 km²) than previous studies in China. The total N deposition budget was 1481 Gg in this region, with 77 % from reduced N and 23 % from oxidized N. The average grid square deposition of 47 kg N ha⁻¹ yr⁻¹ was 3–5 times higher than values obtained for European countries. The exported N budget (1981 Gg) was much higher than the imported N budget (584 Gg), suggesting that the NCP is an important net emission source of N pollutants. The calculated spatial distributions of N deposition displayed high rates of reduced N deposition in the south and of oxidized N deposition in the east. The N deposition exceeded an upper limit of 30 kg N ha⁻¹ for critical loads for natural ecosystems in more than 90 % of the region and the critical level for ammonia concentrations of 3 µg m⁻³ for all plant species was also exceeded in over 90 % of the region. High concentration of nitrogenous pollutants impaired air quality and threatened human health even without available assessments available by now. At the same time it resulted in coastal eutrophication not only in the NCP itself but also in surrounding areas including Miyun reservoir, the Bohai Sea and the Yellow Sea. Future work to improve spatially disaggregated estimates of N deposition in the NCP will focus on refining techniques to spatially map emissions of NH₃ and NOₓ (and including emissions from international shipping) and expansion of monitoring networks for gas and aerosol concentrations and wet deposition of N compounds. Moreover, we recommend future studies with the application of Eulerian models with detailed dynamic meteorology and more complex atmospheric chemistry to estimate N deposition in the NCP.

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