

Nitrogen deposition as an important nutrient from the environment and its impact on ecosystems in China

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Abstract: As an example of atmospheric nitrogen (N) deposition, the paper summarizes the definition, form and amount of nutrient from the environment (NFE) and the relationship between NFE and anthropogenic reactive N emission. Based on our own study and published articles, we find that N wet and dry deposition has been an important nutrient resource in agricultural and natural ecosystems in China. The total amount of N deposition and other environment-derived N in China was up to 18 Tg N/a, equal to approximately 60% of the national N fertilizer consumption. Nitrogen deposition is expected to contribute substantially to nutrient cycling and net primary productivity in various ecosystems. Therefore, it is crucial to utilize this environment-derived nutrient resource by integrated nutrient resource management in order to realize the sustainable development of both agricultural and non-agricultural ecosystems.

Keywords: atmospheric deposition; nitrogen; nutrient management; ecological impacts

With the exacerbation of global environmental problem caused by anthropogenic reactive nitrogen (N) emissions (Vitousek *et al.*, 1997; Holland *et al.*, 1999), nutrients from the environment (NFE) were paid more and more attention (Galloway *et al.*, 2004; Liu *et al.*, 2004; Liu and Zhang, 2009). However, what is the NFE and how about the relationship between NFE and anthropogenic activity? How to understand the effect of NFE on natural ecosystem, especially agro-ecosystem? A series of these problems need scientific explanation in order to provide scientific support for better utilization of the NFE and the avoidance of its negative effects. As an example of atmospheric N deposition, this paper discussed the definition, form, characteristic of temporal and spatial distribution of the NFE as well as the impacts of NFE on nutrient cycling and management in ecosystems.

1 Introduction of NFE

The NFE is a general designation of all kinds of nutrients in terrestrial and aquatic ecosystems, which comes from atmosphere, hydrosphere and lithosphere

through physical, chemical, and biological processes. It includes nutrients from atmospheric deposition, irrigation water, biological N fixation, seeds and seedlings. The NFE, soil and fertilizer derived nutrients are three major nutrient resources for croplands (Zhang *et al.*, 2003). From the aspect of components, NFE includes nitrogen (N), phosphorus (P), potassium (K), and all other essential elements. However, in the view of quantity and ecological impact, N and sulfur (S) are the two most important components of NFE, both of which mainly come from atmospheric deposition (including dry and wet deposition). As N plays even more important role for plant nutrition and environmental pollution, we will give a detailed discussion on atmospheric N deposition in this paper.

Atmospheric N deposition refers to the process whereby airborne nitrogenous compounds (inorganic N including NH_3 , particulate NH_4^+ , NO_x , HNO_3 , and particulate NO_3^- , and organic N including urea,

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amines, proteins, and nucleic acids) are deposited on the Earth's surface by wet deposition and/or dry deposition (Cornell *et al.*, 2003). Since the industrial revolution, especially the widely application of synthetic ammonia production, atmospheric N deposition has become an important component in the global N cycle with increasing anthropogenic atmospheric reactive N emissions (Vitousek *et al.*, 1997; Holland *et al.*, 1999). It was estimated that the global atmospheric N deposition caused by anthropogenic activities has increased from 31.6 Tg N/a in 1860 to 103 Tg N/a in the early 1990s, and is expected to be a further increase to 195 Tg N/a in 2050 due to the accelerated human demand for food and energy (Galloway *et al.*, 2004). Meanwhile, nitrogenous compounds emitted to atmosphere could form aerosols and particular matters through a serial of physical and chemical reactions, and will be transported hundreds and thousands tons N away via the atmospheric circumfluence, causing regional and global problems on air pollution and N deposition. Therefore, atmospheric N deposition has become a global concern because of the exacerbation of anthropogenic activities and their effects (Matson *et al.*, 2002). As a nutrient and a driving factor of acidification, elevated N deposition will reduce the biodiversity and ecological functions of various ecosystems via acidification and eutrophication (Paerl *et al.*, 1985; Stevens *et al.*, 2004). To date, China, West Europe and North America are the three hotspots of global N deposition (Dentener *et al.*, 2006).

2 Quantification method of NFE

The amount of total NFE can be collected and measured by different methods (Liu and Zhang, 2009). Here we mainly introduce the direct and indirect quantification approaches on N wet and dry deposition. Methods for measuring other NFEs can also be seen from Zhu (1997), Liu and Zhang (2009).

2.1 Direct quantification

2.1.1 Nitrogen wet deposition

Currently, rain gauges and wet-only collectors are the main methods. Samples collected by rain gauges belong to bulk deposition because they comprise part of dry deposition. However, samples collected by wet-only collectors are real wet deposition because they only collected rainfall or snow when precipitation

or snow happens (Liu *et al.*, 2006). Generally, there were no significant difference between bulk deposition and wet deposition. However, both of them have to be differentiated in arid area or regions dust storm occurs regularly.

To prevent losses and the N transformation between N components, samples have to be stored in refrigerators immediately after each collection. The dissolved inorganic N (DIN) (NH_4^+ -N and NO_3^- -N) concentration in the wet deposition can be analyzed by continuous flow analyzer (CFA) or ion chromatogram (IC). The total dissolved N (TDN) in the wet deposition can be analyzed using the alkaline persulfate-oxidation (to nitrate) method followed by ultraviolet spectrophotometry. The dissolved organic N is the difference between TDN and DIN (Zheng *et al.*, 2007).

2.1.2 Nitrogen dry deposition

Compared to wet deposition, atmospheric N dry deposition is more difficult to collect and measure. The main reason is that the occurrence of dry deposition is close related to the physical, chemical and biological traits of the surface of deposited collector, and some atmospheric reactive nitrogenous components (NH_3) can bidirectional flow, both of them could produce large uncertainty of atmospheric N dry deposition fluxes (Schjoerring *et al.*, 2001).

Currently, there are two methods for the determination of N dry deposition. One is inferential method, with which the atmospheric reactive nitrogenous components are collected by passive or active samplers. Atmospheric concentration could be obtained after analyzing these samples and meteorological parameters. N dry deposition fluxes at unit time could be estimated by multiplying the measured concentration of reactive nitrogen with time and deposition velocities obtained from literatures (Schmitta *et al.*, 2005; Shen *et al.*, 2009). The other one is micrometeorological method, such as vertical gradient and eddy correlation, based on the principle of microclimatology. The N dry deposition fluxes could be calculated by detecting gradient variances of nitrogenous components rapidly and combining with micrometeorological conditions (Fowler *et al.*, 2001). Compared between these two methods, the former one is relatively easier to realize because the concentration of

atmospheric nitrogenous components could be obtained only when there are meteorological parameters and appropriate collectors for air and aerosol. And the deposition velocities of these reactive nitrogenous components are determinate in each ecosystem, the N dry deposition fluxes thereby could be calculated according to them. The later one is more difficult to carry out because expensive instruments and wide homogenous underlying surface (the ratio of measured height to radius is 1 : 100) are needed in this method. Even in developed European and American countries, there are few laboratories which could assume this method. Nowadays, the Environment and Nutrient Research Group in China Agricultural University (CAU) has adopted passive samplers, high volume particulate samplers and Denuder Long Term Atmospheric Sampling systems to measure the N dry deposition fluxes in North China Plain (NCP). The preliminary results showed that there is serious atmospheric reactive N_r pollution and high N deposition fluxes in NCP (Shen *et al.*, 2009).

2.1.3 Other environmental nutrients

The nutrient input from irrigative water could also be determined like wet deposition. Analyze nutrient concentrations after collecting irrigation samples, and then multiply with annual irrigation, the product is the amount of nutrient from irrigation. The nutrient amount from seeds and seedlings could also be calculated by multiplying nutrient concentration with dry weight of seeds and seedlings used. The nutrient amount from biological N fixation could be estimated by the ratio of fixed N to total uptake N by leguminous plants. It also could be evaluated with the natural ¹⁵N abundance method or the C₂H₂ (ethyne) deoxidize method (Hardarson *et al.*, 1993; Aveline *et al.*, 1996).

2.2 Indirect quantification

2.2.1 Estimation from zero-N soils

This is an indirect method for quantification of NFE, based on the N uptake in plants aboveground from the zero-N plots. Such estimation includes soil N, resulting in an overestimation of the nutrient amount of NFE. However, it could be used as an indirect measurement of airborne N deposition in long-term experiments (LTEs) (more than ten years) because it has reached a dynamic equilibrium for the C and N con-

tents in the soil of LTEs (Jenkinson *et al.*, 2004). There are more than one hundred LTEs with history longer than ten years in China. We can get a general understanding about the amount and distribution of NFE by summarizing N uptake by crops from the zero-N plots of these LTEs.

2.2.2 Biomonitoring

It is a low cost measurement used in the monitoring of air and water pollution. There are two biomonitoring methods: (1) an Integral Total Nitrogen Input (ITNI) system based on ¹⁵N dilution technique (Russow *et al.*, 2001), which is a sand-plant system; (2) plant indicator based on the passive response to airborne N deposition. And the familiar indicator plants include lichen, moss, herbage, and leaves of trees (Sutton *et al.*, 2004).

The principle for the INTI system is ¹⁵N isotope dilution method which is fitted to crops with short life-cycle. At the start of the experiment the indicator plants was planted in sand and the sand-plant system was labeled with enriched ¹⁵N material. As the plants grew, the tracer was diluted by the input of atmospheric N (with natural ¹⁵N abundance). After harvest, ¹⁵N abundance in the various parts of the plants and in the sand and nutrient solution could be analyzed by mass spectrometry and the amount of N deposited from the atmosphere was calculated from the extent of ¹⁵N dilution (Russow *et al.*, 2001; He *et al.*, 2007).

Nitrogen in lichen and moss plant mainly originates from airborne N wet/dry deposition because both of them grow on rock surface. Therefore the N concentration undoubtedly could reflect the pollution degree of atmospheric reactive N and the deposition rates (Pitcairn *et al.*, 2006). For the same reason, N concentration in the aboveground or foliar N in higher plants (especially for NH₄⁺-N) and even the ratio of N to P concentrations also have some relationship to the concentration of atmospheric reactive N species and their deposition rates. Therefore N in those lower and higher plants can indirectly indicate the amount of airborne N deposition. However, a mathematic relation (i.e. a linear regression equation) has to be established between such bio-monitoring results and atmospheric reactive N concentrations or the direct measurements of N deposition (Sutton *et al.*, 2004; Pitcairn *et al.*, 2006).

3 Amount and regional characteristic of NFE in China

Atmospheric N deposition contributes soil and water acidification, but it is also an important environmental nutrient resource. Traditionally the airborne N input could be neglected because of the high application rates of N and organic fertilizers into the agro-ecosystems, which was disturbed intensively by anthropogenic activities. According to a report by the US national atmospheric deposition (NADP) network in recent two decades, annual N wet deposition was only 3–12 kg N/(hm²·a), accounted for 5%–10% of crop N requirement (NADP, 2007). Studies showed that N wet deposition ranged from 15–20 kg N/(hm²·a) in the 1980s (Zhu *et al.*, 1997). But all of these results did not include the contribution of N dry deposition. Recently, some results showed that the amount and effect of N deposition on agro-ecosystems could be much higher than previous estimation (He *et al.*, 2007, 2010). The results from the Broadbalk LTEs of Lausanne in UK showed that the amount of annual airborne N wet and dry deposition was up to 44 kg N/(hm²·a) according to wheat N uptake from the zero-N plots, while the N amount in rainfall was only 9 kg N/hm² during the same period (Goulding *et al.*, 1998). Using the LTEs of Halle and the ITNI system, Weigel *et al.* (2000) reported 50–60 kg N/(hm²·a) airborne N deposition into agro-ecosystems in Germany. The total N deposition values were largely greater than the amount of local N wet deposition. Our research found that the annual total N deposition in NCP was about 80–90 kg N/hm² (He *et al.*, 2007) and 43–55 kg N/hm² deposited in wheat season (He *et al.*, 2010), while the amount of annual N wet deposition was only one third of the total deposition (Liu *et al.*, 2006; Zhang *et al.*, 2008a). Therefore, it is not enough only take into account the amount of N wet deposition when atmospheric N deposition is considered, and dry deposition has also to be determined. Erisman *et al.* (1998) and Fowler *et al.* (2001) summarized the recent research progresses on atmospheric N dry deposition for both methodology and the understanding of deposition processes.

Nowadays, the research about atmospheric N dry deposition is still scarce in China. Therefore the amount and the characteristics of regional distribution

of NFE in China were evaluated mainly based on our recent work and publications. To simplify the calculation, the total amount of N deposition, including wet and dry deposition, can be calculated through the estimation of wet deposition by assuming that the ratio of wet deposition to dry deposition is 1:1 according to some previous findings (Erisman *et al.*, 1998). Annual precipitation in China was about 600 mm (China Agricultural Yearbook, 2001–2007), and the average N (the sum of NH₄⁺-N, NO₃⁻-N, and dissolved organic N) concentration in rainfall across China was about 1.3 mg N/L. Based on this, the amount of N from wet deposition would amount to 7.50 Tg N/a, and that from wet and dry deposition would be 15 Tg N/a in the Mainland in China (Table 1). Similarly, N wet and dry deposition in China was found to be 7.4 Tg N/a and 11.2 Tg N/a in the 1980s and 1990s (Table 1). Such estimation is consistent with results of anthropogenic NH₃ and NO_x emission inventory in China since 1980 but slightly higher than total N deposition values during 1993 and 2003 reported by Lu and Tian (2007). It was roughly estimated that about one fourth of these N would deposit into agro-ecosystems according to the total amount of annual N deposition about 30 kg N/(hm²·a) in croplands, and the rest three fourths would deposit into forest, grassland, and aquatic ecosystems in China. If the N from biological N fixation (2.0 Tg N/a), irrigation (0.5 Tg N/a), seeds and seedlings (0.5 Tg N/a) was taken into account, the sum of NFE would be up to 18 Tg N/(hm²·a), accounting for 60% of the national annual N fertilizer consumption (China Agriculture Yearbook, 2001–2007). If only considered the amount of NFE input into croplands, it would amount to 7.0 Tg N/a, approximating one fourth of nitrogenous fertilizer applications in the agroecosystems. In this sense, we should pay more attention to the contribution of NFE to plant nutrition in croplands.

Table 1 Anthropogenic emissions of NH₃ and NO_x in China since the 1980s

N emission	1980s	1990s	2000s	Average
	(Tg N/a)			
NH ₃ -N	5.84	9.54	12.0	9.13
NO _x -N	1.41	2.69	4.31	2.63
Total	7.25	12.2	16.3	11.7

Data source: Liu and Zhang (2009).

On the other hand, there was a large regional difference about NFE in China. Generally, the amount of NFE in the inland areas was lower than that in mid-eastern and coastal areas of China, but it was higher than that in the Tibetan Plateau and northwest region of China. For example, our previous study has showed that the amount of inorganic N and organic N from wet deposition was about 27 kg N/(hm²·a) and 5 kg N/(hm²·a) in NCP (Zhang *et al.*, 2008a; Zhang *et al.*, 2008b). The amount of wet deposition was positively related with precipitation, and that occurred from June to September nearly accounted for two third of that happened in the whole year because of the high precipitation during this period. There are three grades about wet deposition of inorganic N (NH₄⁺-N and NO₃⁻-N) in China (Liu and Zhang, 2009): high deposition regions (>25 kg N/(hm²·a)) like Shanghai, Beijing, Henan, Shandong, Sichuan, Chongqing, Jiangsu, Zhejiang, and Jiangxi, etc.; medium deposition regions (15 to 25 kg N/(hm²·a)) like Hebei, Hunan, Hubei, Shannxi, Liaoning, Fujian, and Guangdong, etc.; low deposition regions (< 15 kg N/(hm²·a)) like Yunnan, Guizhou, Xizang, Inner Mongolia, Xinjiang, Gansu, Jilin, Heilongjiang, and so on. If N deposition and the situation of local economy are linked together, we would find that the amount of atmospheric N deposition is higher in those developed areas than those less developed areas (Lu and Tian, 2007). This can be explained that higher emissions of both reduced and oxidized N species are greater in developed areas than undeveloped areas due to faster growth of economy in the developed areas. In addition, the small amount of rainfall in the arid and semi-arid area of western China is also an important reason for the low N wet deposition in those areas. However, the contribution of N dry deposition to total N deposition in these areas is significantly greater than that in south rainy regions. As a part of N dry deposition, the N from sand or dust storm occurred mainly during winter and spring season in northern and northwestern China is a considerable contributor to annual input of NFE. It was estimated that the amount of N input from the dust storm happened on 16–17 April 2005 in Beijing amounted to 6,000 t, corresponding to 4 kg N/hm² of N dry deposition in the whole Beijing area (Liu and Zhang, 2009).

Moreover, sand storm usually happens more than ten times each year in Xinjiang province, in this case, it is undoubtedly an important source of NFE in local agro-ecosystem and natural ecosystem (Wen *et al.*, 2002).

4 Impact of NFE on agro-ecosystems and natural ecosystems

As a main component of NFE, atmospheric N deposition has a typical characteristic of nutrient resource and regional distribution. By this, counter measures of management according to the characteristic of each region have to be taken. In mid-eastern China, including the NCP and coastal developed areas, we have to pay more attention to the contribution of environmental N to croplands and water eutrophication, so as to make full use of this part of NFE and reduce unreasonable input of nitrogenous fertilizer for the efficient utilization of nutrient resources (Zhang *et al.*, 2008). For instance, based on ¹⁵N dilution it was found that the total airborne N input into the whole maize-wheat rotation system was about 80–90 kg/hm², while only 50 kg/hm² could be available for maize and wheat plant in NCP (He *et al.*, 2007), suggested that grain production would not be affected even if 50–80 kg N/hm² N fertilizer application was cut down. However, in the inland and west areas of China, as the contribution of atmospheric N deposition to agro-ecosystems is low, we have to focus on the potential effects of NFE (such as atmospheric N deposition) on forest and grassland ecosystems, including aboveground productivity, biodiversity, acidification, the potential of soil C storage and fixation, and the emission of greenhouse gases. It was found that the annual airborne N deposition would result in the loss of 17% species in grassland ecosystem although it was only 10 kg/hm² higher than the background value (Clark and Tilman, 2008). From this viewpoint, small increase in N deposition may have even greater impacts on plant biodiversity of natural ecosystems. Nevertheless, further studies should be conducted to verify this finding in other grassland and/or forest ecosystems.

With the development of industry and intensive agriculture, characterized by over-fertilization, high

stocking rates of domestic animals, and rapid growth in energy consumption, there is a great increase about anthropogenic atmospheric reactive N emission. Total emissions of NH_3 and NO_x in China increased from 7.2 Tg N/a in the 1980s to 16.3 Tg N/a in the 2000s (Table 1), consistent with the emission inventory by other studies (Streets *et al.*, 2003; Anon, 2007). In this sense, it could be concluded that the atmospheric N deposition has already doubled since 1980 in China although the magnitude and its ecological impacts (e.g. the N deposition effects on productivity, soil potential of C storage, and biodiversity of natural and semi-ecosystems) are not very clear so far. But the patterns of N deposition and its potential effects should be identified through long-term N deposition monitoring networks and cross-site N addition experiments across China.

5 Research recommendations

China's current economy (GDP) is eight times of that in the 1980s and is expected to have further increase in the next decades (Liu and Diamond, 2008). As a result, China's anthropogenic N emissions mainly from agricultural production and fossil fuel combustion are likely to increase substantially in the near future. There are strong research needs to forecast reactive N emission trends in China considering all kinds of trade-offs in N emission increase or decrease due to expanded intensive agriculture (e.g. improved nutrient management and change of land use) and wider application of innovative techniques in traffic and industrial emission reduction.

It is difficult to evaluate the effect of pollution control measures on atmospheric reactive N (and other pollutants) concentrations and their deposition in

whole China without a national atmospheric deposition monitoring network. It is urgent to organize a long-term national deposition network (like the NADP) to monitor N wet and dry deposition across China. Cross-site N addition experiments along with typical forests, grasslands and aquatic ecosystems are greatly needed in China. Such long-term experiments could provide information on the impact of elevated N deposition on both terrestrial and marine ecosystems under the background of global change. Besides, Chinese scientists and governments should take actions for public education to improve their awareness of environmental protection in particular atmospheric N pollution and deposition.

In summary, N deposition as key component of NFE, has become an indicator of anthropogenic reactive N enrichment induced by expanding Chinese economy. Nitrogen deposition can not only cause rapid increase in environmental nutrient input in some intensive agricultural ecosystems (He *et al.*, 2010) but also lead to detrimental effects on many natural and semi-natural ecosystems in those less anthropogenic impacted regions (Stevens *et al.*, 2004; Phoenix *et al.*, 2006; Clark and Tilman, 2008). We believe that heavy N deposition could be used as an important nutrient resource in croplands. On the other hand, the potential risk of N deposition on grasslands, forests and aquatic ecosystems should be controlled within the acceptable level (below critical load) by substantially reducing the reactive N emissions to the environments.

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