A database of atmospheric nitrogen concentration and deposition from the nationwide monitoring network in China

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Atmospheric nitrogen (N) deposition has increased substantially across China since 1980; however, data for N deposition fluxes since the 2000s has been very limited. Understanding and mitigating the impacts of N deposition requires long-term quantification of dry as well as wet deposition of key reactive nitrogen (Nr) species. Here we present a dataset for inorganic N concentrations and deposition for the period 2010–2015 in China, compiled from the nationwide deposition monitoring network. The dataset comprises information from 32 monitoring sites on concentrations and bulk deposition (wet plus part of dry deposition) fluxes of NH4\(^+\)-N and NO3\(^-\)-N, air concentrations and dry deposition fluxes of the major Nr species NH3, NO2, HNO3, and particulate NH4\(^+\) and NO3\(^-\). This unique database is available inter alia to advance understanding of the spatial patterns of inorganic N concentrations and deposition in China and its associated effects, constrain primary Nr (e.g., NH3, NOx) emission inventories, and validate outputs of atmospheric chemistry and transport models.

Background & Summary

The deposition of reactive nitrogen (N) from the atmosphere to the surface is an important component of the human-accelerated global N cycle and a serious form of atmospheric pollution. Reactive nitrogen (Nr) comprises both oxidized (e.g., NO, NO2, HNO3) and reduced (NH3) gases and their particle-phase nitrate (NO3\(^-\)) and ammonium (NH4\(^+\)) equivalents. Excess Nr deposition results in adverse ecological effects, including the loss of biological diversity, nutrient imbalance, soil acidification, and eutrophication of water bodies. Nitrogen deposition of oxidized N has decreased or stabilized in Europe and the U.S. since 1990, mainly due to reductions in emissions of NOx (NO + NO2). In contrast, growing agricultural and industrial activities in China have led to increasing emissions of both NH3 and NOx since the 1980s. Although the trend has slowed recently with the introduction of strict air pollution measures, the atmospheric Nr deposition in China consequent on the large emissions of Nr to the atmosphere is of widespread concern.

Atmospheric deposition occurs via wet and dry deposition pathways. Wet deposition refers to removal of gases and particles from the atmosphere by precipitation events (rain and/or snow), whilst dry deposition is the transport of gases and particles to surface via turbulent exchange and gravitational settling in the absence of precipitation. Compared with wet deposition, data for dry deposition fluxes were much sparser in China and other countries worldwide. This is because wet deposition can be directly quantified from chemical analysis of collected bulk precipitation samples, whereas direct measurement of dry deposition is technically challenging and needs to include a wide range of N-containing compounds in both gaseous and particle phases. The direct measurement of dry deposition fluxes via micrometeorological methods requires complex and expensive instruments and therefore cannot be applied over large domains or for long time series. Instead, the inferential method can be used for estimating dry deposition at large spatial and long-term scales, despite the associated uncertainties. In this approach, dry deposition fluxes are estimated as the product of the atmospheric
concentrations of the species of interest and their deposition velocities, the latter of which are inferred from meteorological variables and land-use type.

National measurements of nitrogen deposition did not exist in China until the instigation in 2004 of the Nationwide Nitrogen Deposition Monitoring Network (NNDMN) operated by China Agricultural University. Initially this network comprised only measurements of bulk (wet) N deposition. In 2010, simultaneous measurements of air concentrations and associated dry deposition fluxes of five major N species (i.e., gaseous NH₃, NO₂, and HNO₃, and particulate NH₄⁺ and NO₃⁻), were added. Examination of the more comprehensive data between 2010 and 2015 has shown both high Nr concentrations and deposition fluxes and a high degree of spatial variability across China, for example greater Nr pollution in the northern region than in the southern region, especially in rural areas. Analysis has also revealed the equal importance of dry and wet deposition at a national scale, which demonstrates the need to include both dry and wet/bulk deposition in evaluating the effects of N deposition on eco-environmental health. In summary, despite achievement of effective controls of SO₂ and NOₓ emissions, the absence of regulation or legislation regarding agricultural NH₃ means that China has still been subject to high levels of Nr deposition in recent years.

The World Meteorological Organization (WMO) Global Atmosphere Watch programme (GAW) recently completed an assessment of global N wet and dry deposition, but only a few Chinese monitoring sites were included for validation of modeled wet deposition, and none for validation of dry deposition. The absence of an open-access database for atmosphericNr composition and deposition in China, especially dry deposition, strongly motivates us to provide global scientists with the dataset of Nr deposition from the NNDMN. We define this dataset as NNDMN 1.0 version, and it comprises three files, the 'data file', a 'profile file' and a 'readme file'. The dataset contains monthly average air concentrations and dry deposition fluxes of NH₃, NO₂ and HNO₃, and particulate NH₄⁺ and NO₃⁻, and volume-weighted mean concentrations and bulk deposition fluxes of NH₄⁺-N and NO₃⁻-N, as well as detailed information on each site including longitude, latitude and land use, and the methods of field sampling and laboratory analysis. This database will enable scientists and policymakers to explore spatiotemporal trends of N deposition in China, validate modelling results and assess the ecological burden of N deposition on sensitive ecosystems (e.g., forest and grassland) in China. At present most sites have about 5 years of measurements and the intention is to add ongoing data collection from the NNDMN to this open-access dataset to enable longer-term trends in bulk and dry N deposition in China to be followed.
**Methods**

**Database structure.** The NNDMN 1.0 database consists of three files (Fig. 1). The 'data file' provides monthly mean data on the concentrations of NH$_4^+$-N and NO$_3^-$-N in precipitation, bulk N deposition fluxes of NH$_4^+$-N and NO$_3^-$-N, monthly integrated precipitation amount, and the concentrations and dry deposition fluxes of gaseous NH$_3$, NO$_2$ and HNO$_3$, and particulate NH$_4^+$ and NO$_3^-$ for 32 monitoring sites. Additional information such as site code, site name, site coordinate (longitude and latitude), land-use type, sampling method, and sampling period associated with each Nr species is included in 'profile file' (Fig. 1). All mean values in the 'data file' are monthly data. The 'readme file' explains the abbreviations used in the 'data file' and 'profile file', and provides the units of all variables included (Fig. 1).

**Data acquisition.** Ambient concentrations of gaseous NH$_3$ and HNO$_3$, and particulate NH$_4^+$-N and NO$_3^-$-N, were measured using an active DELTA (DENuder for Long-Term Atmospheric sampling) system$^{16}$. The system consists of a denuder filter sampling train, a low-volume pump to provide sampling flow rate of 0.2–0.4 L min$^{-1}$, and high sensitivity dry gas meter to record sampled volume. The sampling train comprises of two potassium carbonate plus glycerol (1% (m/v) K$_2$CO$_3$ + 1% (m/v) glycerol in methanol) coated denuders in series to collect acid gases (HNO$_3$, SO$_2$ and HCl), followed by two citric acid (5% (m/v) citric acid in methanol) coated denuders to trap NH$_3$. Two set of coated cellulose filters in a 2-stage filter pack at the end of the sampling train to collect acid gases (HNO$_3$, SO$_2$ and HCl), followed by two citric acid (5% (m/v) citric acid in methanol) coated denuders to trap NH$_3$. Two set of coated cellulose filters in a 2-stage filter pack at the end of the sampling train was used to collect aerosol components, with a first K$_2$CO$_3$/glycerol impregnated filter to capture particle phase anions (NO$_3^-$, SO$_4^{2-}$, Cl$^-$) and cations (Na$^+$, Mg$^{2+}$, Ca$^{2+}$), and a second filter coated with citric acid to collect any volatilized aerosol NH$_4^+$. With a monthly sampling period, the detection limits of the DELTA method for gaseous HNO$_3$ and NH$_3$, and particulate NH$_4^+$ and NO$_3^-$ was determined as 0.03 µg HNO$_3$ m$^{-3}$, 0.01 µg NH$_3$ m$^{-3}$, 0.02 µg NH$_4^+$ m$^{-3}$ and 0.05 µg NO$_3^-$ m$^{-3}$, respectively.

Gaseous NO$_2$ concentration was measured in triplicate with passive Gradko diffusion tubes (Gradko International Limited, UK). Each sampler is made up of acrylic tube (71.0 mm long × 11.0 mm internal diameter) with colored and white thermoplastic rubber caps. Gaseous NO$_2$ was absorbed into a 20% triethanolamine–deionized water solution coated onto two stainless steel wire meshes within the colored cap. As indicated by the manufacturer, the uptake rate of the tube is 68.8 × 10$^{-9}$ m$^{-3}$ h$^{-1}$, the desorption efficiency is 0.98, the detection limit is 1.6 µg NO$_2$ m$^{-3}$ over a 2-week sampling period, and the analytical expanded measurement uncertainty is ±10%. Over the entire period, the standard deviations of three NO$_2$ replicates across all sites were between 0.03 and 20.3 µg NO$_2$ m$^{-3}$ and averaged 1.68 µg NO$_2$ m$^{-3}$ (95% confidence interval 1.58–1.81). The air intakes of the DELTA system and the NO$_2$ tubes were set at 2 m above the ground at most sites. All sampling was performed on a monthly basis, that is, one sample per month for each Nr species.

Precipitation samples (here termed as wet/bulk deposition, which contains wet and some dry deposition) at all sites were collected using a standard precipitation gauge (SDM6, Tianjin Weather Equipment Inc., China) located beside the DELTA system (~2 m). The precipitation gauge consists of a stainless steel funnel and glass bottle (vol. 2000–2500 ml). Precipitation amount was measured using a graduated cylinder (scale range: 0–10 mm; division: 0.1 mm) coupled with the gauge. After each daily (8:00 am–8:00 am next day) event, precipitation samples (including rain and melted snow) were collected and stored in clean polyethylene bottles (50 mL) at 4 °C before analysis. Acid-coated denuders and aerosol filters were extracted with 6 and 10 mL of high-purity water (18.2 MΩ), respectively, and analyzed for NH$_4^+$-N with an AA3 continuous-flow analyzer (CFA) (BranC Luebbe GmbH, Norderstedt, Germany). Carbonate-coated denuders and filters were both cleaned three times with high-purity water after each collection and once every week in order to avoid cross contamination.

In the CAU’s analytical laboratory, the exposed sampling trains of the DELTA systems and NO$_3$- tubes were stored at 4 °C before analysis. Acid-coated denuders and aerosol filters were extracted with 6 and 10 mL of high-purity water (18.2 MΩ), respectively, and analyzed for NH$_4^+$-N with an AA3 continuous-flow analyzer (CFA) (BranC Luebbe GmbH, Norderstedt, Germany). Carbonate-coated denuders and filters were both extracted with 10 mL 0.05% H$_2$O$_2$ solution followed by analysis of NO$_3^-$-N using the same CFA. The detection limits were determined as 0.01 mg N L$^{-1}$ for NH$_4^+$ and NO$_3^-$-N. The meshes from the NO$_2$ diffusion tubes were extracted with a solution containing sulfanilamide, H$_3$PO$_4$, and N-1-naphthylethylene-diamine, and the NO$_2$ content in the extract determined using a colorimetric method by absorption at a wavelength of 542 nm. The detection limit for NO$_2$ was 0.01 mg N L$^{-1}$. Each collected precipitation sample was filtered with a 0.45 µm syringe filter (Tengda Inc., Tianjin, China), and analyzed for NH$_4^+$-N and NO$_3^-$-N using the CFA as mentioned above. Quality assurance and quality control procedures adopted in the analytical laboratory are described in “Technical Validation”. Further details of precipitation measurement, samples handling, and chemical analysis can be found in our previous studies$^{11}$.

Wet/bulk N deposition flux ($D_n$, kg N ha$^{-1}$) was calculated as the product of the precipitation amount ($P$, mm) and the volume-weighted mean concentration of Nr species in precipitation ($C_r$, mg N L$^{-1}$), using Equation (1).

$$D_n = PC_r/100$$  (Equation 1)

The dry deposition flux of gaseous and particulate Nr species was calculated by multiplying measured concentrations with simulated deposition velocities ($V_d$) from the GEOS (Goddard Earth Observing System)-Chem chemical transport model (http://geos-chem.org)$^{17}$. The GEOS-Chem CTM is driven by GEOS (Goddard Earth Observing System) assimilated meteorological data from the NASA Global Modeling and Assimilation Office (GMAO). The GEOS-5 data are available with a temporal resolution of 6 h (3 h for surface variables and mixing depths) and a horizontal resolution of 1/2° latitude × 2/3° longitude. The nested-grid version of GEOS-Chem$^{18}$ was used with the native 1/2° × 2/3° resolution over East Asia (70°E–150°E, 11°S–35°N). The model calculation of dry deposition of Nr species follows a standard big-leaf resistance-in-series model for gases$^{19}$ and aerosols$^{20}$. The aerodynamic resistance to turbulent transfer from the measurement heights (~2 m) to the roughness height
and pNO3

Vd

5%. In addition, triplicate NO2 samplers were deployed and collected at
samples were controlled to be less than
±
during each analysis run. The differences between the determined and "theoretical" results from the standard
every 20 samples) performed using standard (designed specific concentrations of NH4

centrations, and field (travel) blanks were used to check for contamination. Quality assurance was routinely (once
exposed samples. The laboratory blanks were subtracted from the samples to correct all quantified species con-

Table 1. Summary statistics for monthly mean concentrations of Nr species in air (NH3, NO2, HNO3, pNH4+
and pNO3− in µg N m−3) and in precipitation (NH4+ and NO3− in mg N L−1), and their respective dry and bulk
deposition fluxes (kg N ha−1 month−1) during the sampling period at the 32 sites. pNH4+ and pNO3− denote
particulate NH4+ and NO3−; NH4+ and NO3− denote NH4+ and NO3− in precipitation. The number of 1619
included value of 0 from sampling months without precipitation event.

| Speciesa | Concentration | Deposition flux |
|----------|---------------|-----------------|
|          | Min | Max | Median | Mean | SD | N | Min | Max | Median | Mean | SD | N | Min | Max | Median | Mean | SD | N |
| NH3     | 0.16 | 39.6 | 5.8 | 7.0 | 5.4 | 1790 | 0.01 | 4.33 | 0.63 | 0.80 | 0.66 | 1790 |
| NO2     | 0.13 | 29.1 | 5.8 | 6.6 | 3.8 | 1790 | 0.0002 | 2.38 | 0.19 | 0.26 | 0.26 | 1790 |
| HNO3    | 0.02 | 4.9 | 1.0 | 1.2 | 0.7 | 1790 | 0.0004 | 3.88 | 0.29 | 0.43 | 0.46 | 1790 |
| pNH4+   | 0.02 | 57.2 | 4.9 | 6.5 | 6.1 | 1790 | 0.0013 | 4.64 | 0.19 | 0.31 | 0.36 | 1790 |
| pNO3−   | 0.01 | 32.1 | 2.1 | 2.8 | 2.5 | 1790 | 0.0002 | 1.37 | 0.09 | 0.12 | 0.11 | 1790 |
| NH4+    | 0.01 | 26.8 | 1.4 | 2.3 | 2.7 | 1426 | 0 | 10.4 | 0.55 | 0.95 | 1.24 | 1619 |
| NO3−    | 0.02 | 28.9 | 1.5 | 2.4 | 2.9 | 1426 | 0 | 10.7 | 0.57 | 0.86 | 1.03 | 1619 |

is estimated using the GEOS-5 data. The surface uptake resistance is calculated based on the Global Land Cover
Characteristics Data Base Version 2.0 (http://edc2.usgs.gov/glcc/globdoc2_0.php), which defines land types (e.g.,
urban, forest, etc.) at 1 km × 1 km resolution and is then binned to the model resolution as fraction of the grid
cell covered by each land type. The model 1/2° resolution coarsely represents regional land characteristics
around the monitoring sites. Bi-directional NH3 exchange is not considered in the model. The hourly Vj values
were modeled from January 2011 to May 2013, and the period of June 2013-December (when the GEOS-5 meteor-
orological data are unavailable) was filled using mean modeled values for each hour. The monthly Vj at each site
was then averaged based on the hourly dataset for further estimation of dry deposition flux of each Nj species
during the observation.

Data Records

The data are available in a single dataset31, which consists of three Microsoft Excel files: the ‘data file’ (NNDMN
1.0 Data File), the ‘profile file’ (NNDMN 1.0 Profile File), and the ‘readme file’ (NNDMN 1.0 Read Me) which
explains the abbreviations and units (Fig. 1). The NNDMN 1.0 database is the most comprehensive and up-to-date
measurement-based dataset of ground-level concentrations and dry and bulk deposition of key Nr species over
different land-use types (e.g., urban, rural, and coastal, forest, grassland) in China. The data time series runs
from 2010 to the current latest available year of 2015. Specifically, the NNDMN 1.0 database includes records at
32 locations for monthly mean concentrations and bulk deposition of fluxes of NH4+ and NO3− in precipitation,
and monthly mean air concentrations and dry deposition fluxes of NH3, NO2, HNO3, and particulate NH4+ and
NO3−, with summary statistics presented in Table 1. In brief, monthly mean concentrations of NH3, NO2, HNO3,
and particulate NH4+ and NO3−, ranged over 0.16–39.6 (average 7.0), 0.02–4.9 (1.2), 0.02–57.2 (6.5), and 0.01–32.1 µg N m−3 (2.8 µg N m−3), respectively, while monthly volume-weighted mean concentration
of NH4+ and NO3− in precipitation were 0.01–26.8 (2.3), and 0.02–28.9 mg N L−1 (2.4 mg N L−1), respectively.

The averages of matched months during the sampling period at each site were used to calculate annual averages.
Grouped by land-use type, the annual average dry deposition (1.9–16.6 kg N ha−1 yr−1 for NH3, 0.2–16.0 kg N ha−1
yr−1 for HNO3, 0.2–9.5 kg N ha−1 yr−1 for NO2, 0.1–11.8 kg N ha−1 yr−1 for particulate NH4+, and 0.2–4.1 kg N
ha−1 yr−1 for particulate NO3−) and bulk deposition (2.7–18.9 kg N ha−1 yr−1 for NH4+−N, and 1.5–17.4 kg N ha−1
yr−1 for NO3−−N) of inorganic Nr species are ranked by land use as urban > rural > background sites (the latter
comprising the average of forest, grassland and coastal sites). In addition, across all sites the total dry deposition
for the five Nr species (21.5 ± 10.8 kg N ha−1 yr−1 (mean ± standard deviation)) was similar to the total bulk dep-
osition of NH4+−N and NO3−−N (21.5 ± 6.9 kg N ha−1 yr−1), respectively. These results reflect the positive association between anthropic N emissions and N deposition, and also demonstrate that dry deposition is a major pathway in
China that must be included in estimates of the total Nr deposition.

Technical Validation

For all sites in the NNDMN, the data were obtained via internationally widely-used samplers (e.g., rain gauge,
DELTA system and Gradko tubes) and quantification methods (e.g., inferential method). In addition, all field
measurements were subject to uniform standard procedures for sampling and storage methods, undertaken by
trained personnel. All samples were prepared and measured in the Key Laboratory of Plant-Soil Interactions,
Chinese Ministry of Education, China Agricultural University, which operates a full formal quality control sys-

32. Three laboratory blanks and three field (travel) blanks were prepared and analyzed for each batch of field
samples. The laboratory blanks were subtracted from the samples to correct all quantified species con-
centrations, and field (travel) blanks were used to check for contamination. Quality assurance was routinely (once
every 20 samples) performed using standard (designed specific concentrations of NH4+−N and NO3−−N) samples
during each analysis run. The differences between the determined and “theoretical” results from the standard
samples were controlled to be less than ±3%. In addition, triplicate NO3− samples were deployed and collected at
all sites for each sampling, which allowed confirming a good reproducibility of the Gradko method. The NNDMN
1.0 database presents detailed information on the sampling and laboratory analysis methods for the users to eval-
uate for themselves. Where there was occasional missing monthly data, the averages of the previous and following
months were used to gap fill for calculation of the monthly mean (which have been highlighted in red in the ‘data

Table 1. Summary statistics for monthly mean concentrations of Nr species in air (NH3, NO2, HNO3, pNH4+
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deposition fluxes (kg N ha−1 month−1) during the sampling period at the 32 sites. pNH4+ and pNO3− denote
particulate NH4+ and NO3−; NH4+ and NO3− denote NH4+ and NO3− in precipitation. The number of 1619
included value of 0 from sampling months without precipitation event.
file', and were also specified in 'readme file'). Data from this NNDMN 1.0 dataset has been subject to international peer review as part of the process of publication in high-quality international literature.

### Ancillary datasets

Other datasets that users may find helpful in interpreting the data provided in the NNDMN 1.0 database described in this paper include the following:

- The MIX gridded emissions inventory for anthropogenic pollutants (e.g., NH₃ and NOₓ) and greenhouse gases ([http://meicmodel.org/](http://meicmodel.org/))
- The Infrared Atmospheric Sounding Interferometer (IASI) NH₃ data ([https://iasi.aeris-data.fr/NH3/](https://iasi.aeris-data.fr/NH3/))
- The Ozone Monitoring Instrument (OMI) NO₂ data ([http://www.temis.nl/airpollution/NO2.html](http://www.temis.nl/airpollution/NO2.html))
- Precipitation and other meteorological data for China ([http://data.cma.cn/en](http://data.cma.cn/en))
- Land-use cover for China ([http://www.resdc.cn/](http://www.resdc.cn/))
- Air quality data for China ([http://www.cnemc.cn/](http://www.cnemc.cn/)).

### Code Availability

No custom computer code was used to generate the data described in the manuscript.

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### Author Contributions

Wen Xu, Lin Zhang and Xuejun Liu designed the database, compiled the data and wrote the paper.

### Additional Information

**Competing Interests:** The authors declare no competing interests.

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