## Response to Referee #1:

***Interactive comment on* "Quantifying atmospheric nitrogen deposition through a nationwide monitoring network across China" *by* W. Xu et al.**

**Anonymous Referee #1**

Received and published: 14 July 2015

### General comments

The manuscript 'Quantifying atmospheric nitrogen deposition through a nationwide monitoring network across China' presents the results from 5 years of reactive nitrogen atmospheric concentrations and bulk deposition monitoring combined with modeled dry nitrogen deposition across China. This is an important contribution to the field of reactive nitrogen monitoring in a rapidly developing hotspot of air pollution. However, some general aspects may need to be improved so that results can be interpreted correctly.

**Response**: The authors are grateful to the referee for the valuable and insightful suggestions. We believe that addressing the issues raised by the referee will considerably improve the quality of manuscript. Please see our response to each comment below.

### Specific comments

1. The description of the results is based on ranking Chinese regions according to their levels of reactive nitrogen pollution and nitrogen deposition levels. However, the monitoring sites since some regions include more urban sites, with higher pollution levels, than others or a higher proportion of background sites, away from pollution sources. Thus, the mean value obtained in each region may not be informative of the pollution levels of the whole region. Alternatively, the comparisons between regions can be based on the analysis across urban sites, rural and background sites. For instance, the regional ranking based on mean dry deposition levels can change depending on whether all sites are considered (as it is now in the manuscript), only background sites or only rural sites. The same may be true for wet and total deposition. The comparisons across regions would be fairer this way.

**Response:** We agree with the reviewer that the monitoring sites included within each region are not homogenously distributed but they reflected the real situation in China: more research has been done in eastern and mid eastern regions and less work been conducted in western (including Tibetan Plateau) and northwestern regions. To our best knowledge, there is no systematic and comprehensive measurement study focused on comparisons of wet and dry N deposition fluxes between different land use types across China. Thus, according to the reviewer's suggestion we have reanalyzed the data on $N_r$ concentrations as well as N deposition (dry, wet/bulk and total) fluxes based on the three land use types (urban, rural and background sites) within each region, among six regions, and across the country in the revised paper.

2. In the same line with the previous comment, Table 2 presents a comparison between NNDMN results and other monitoring networks. This comparison is biased by the fact that CASTNET monitoring sites, unlike NNDMN, are located in rural and protected areas, with no sites in urban environments. EMEP data considered here is produced from modelled data representing large scale areas within each grid cell that make comparisons with point measurements difficult. EMEP also has a monitoring network of background sites across Europe with data downloadable from the internet that

maybe more useful for comparisons in Table 2. Also, of the 10 EANET sites presented by Endo et al. (2011), 8 were classified as remote stations, one rural and only one urban. The latter two stations showed higher nitrogen deposition fluxes than remote sites. It was recognized in this study that concentrations in Japan were generally lower compared to other EANET sites in East Asia because most locations were categorized as remote sites. Thus, comparisons in Table 2 with CASNET, EMEP and EANET Japan should be based only on rural and/or background sites of the NNDMN.

**Response**: We agree to this comment. We have improved Table 2 as suggested by the reviewer, i.e. removed the urban sites in NNDMN and then made comparisons with other 3 networks. But the revised Table 2 is still not perfect because of the following two reasons: 1) the data on concentrations and deposition fluxes of $N_r$ species for the three land use types (i.e. remote, rural and urban) presented by Endo et al. (2011) cannot be extracted separately; 2) to our knowledge the observation data of EMEP was not available, though the gridded data can be downloaded via the website of EMEP. Since the current study is a Chinese survey, the aim here is to give a general summary of how the range of nitrogen deposition values which we have measured in China compares with other regions of the world and demonstrate that China is a global hot spot for N deposition. During the literature review, we find the most appropriate way of comparison is to use the conclusion from the WMO/GAW assessment work (Vet et al, 2014). So, we have revised the text at the end of Section 4.2 to make a more comprehensive and scientific comparison, instead of comparison from EANET and EMEP.

"On the basis of 2001 ensemble-mean modeling results from 21 global chemical transport models (Vet et al., 2014), three regions of the globe where total deposition is very high: western Europe (with levels from 20.0 to 28.1 kg N $ha^{-1}$ $yr^{-1}$); South Asia (Pakistan, India and Bangladesh) from 20.0 to 30.6 kg N $ha^{-1}$ $yr^{-1}$ and East Asia from 20 to 38.6 kg N $ha^{-1}$ $yr^{-1}$ in eastern China (the global maximum). Extensive areas of high deposition from 10 to 20 kg N $ha^{-1}$ $yr^{-1}$ appear in the eastern U.S. and southeastern Canada as well as most of central Europe. Small areas with total deposition of N from 10 to 20 kg N $ha^{-1}$ $yr^{-1}$ are present, and very large areas of the continents have deposition from 2 to 10 kg N $ha^{-1}$ $yr^{-1}$. In contrast, the present study shows much higher total deposition flux (39.9 kg N $ha^{-1}$ $yr^{-1}$) at a national scale. In China, the consumption rates of chemical fertilizer and fossil fuel have increased 2.0-and 3.2-fold, respectively, between the 1980s and the 2000s (Liu et al., 2013). As a result, the estimated total emission of $NH_3$ reached 9.8 Tg in 2006, contributing approximately 15% and 35% to the global and Asian $NH_3$ emissions (Huang et al., 2012), and $NO_x$ emissions from fossil fuel combustion increased from 1.1 Tg N in 1980 to about 6.0 Tg N in 2010 (Liu et al., 2013). The increasing $NO_x$ and $NH_3$ emissions in China led to higher atmospheric N deposition than those observed in other regions. In addition, emissions of nitrogen compounds in other parts of the world are declining. In the US, for example, $NO_x$ emissions from the power sector and mobile sources were reduced by half from 1990 to 2010 (Xing et al., 2013), which explained the declined N deposition fluxes during period of 1990-2009 observed at 34 paired dry and wet monitoring sites in the eastern US (Sickles II et al., 2015). In Europe, the total $NO_x$ and $NH_3$ emissions decreased by 31% and 29% from 1990 to 2009 (Torseth et al., 2012). N deposition has decreased or stabilized in the United States and Europe since the late 1980s or early 1990s with the implementation of stricter legislation to reduce emissions (Goulding et al., 1998; Holland et al., 2005). However, wet deposition of ammonium, due to no regulation on $NH_3$ emission, has increased over recent decades in the US (Du et al., 2014)."

Table 2 Comparison of dry, wet (wet/bulk), and total deposition fluxes of $N_r$ compounds between NNDMN in China and 3 networks in other countries.

| Network | | Japan EANET network[a] | | | CASTNET[b] | | | EMEP[c] | | | NADMM[d] | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Number of sites or grids | | 10sites | | | 130sites | | | 2447girds (0.5° × 0.5°) | | | 33sites | | |
| Observation period | | Apr. 2003-Mar. 2008 | | | Apr. 2006-Dec. 2013 | | | Jan. 2003-Dec. 2007 | | | Aug. 2006-Sep. 2014 | | |
| N deposition (kg N $ha^{-1}$ $yr^{-1}$) | | Dry | Wet | Total | Dry | Wet | Total | Dry | Wet | Total | Dry | Wet/bulk | Total |
| | Average | 3.9 | 6.6 | 10.6 | 3.1 | 1.3 | 4.4 | 3.9 | 4.8 | 8.7 | 18.7 | 18.2 | 36.9 |
| | Median | 4.1 | 5.9 | 11.2 | 3.0 | 0.7 | 4.1 | 3.7 | 4.7 | 8.5 | 18.7 | 21.3 | 36.5 |
| | Max | 7.0 | 15.8 | 18.2 | 9.7 | 10.3 | 19.6 | 15.8 | 16.9 | 28.0 | 43.1 | 32.4 | 70.9 |
| | Min | 1.0 | 2.1 | 3.0 | 0.03 | 0.1 | 0.3 | 0.1 | 0.6 | 0.7 | 1.1 | 1.5 | 2.9 |

[a]The Japan EANET data are sourced from Endo et al. (2011). Gaseous $NO_2$ was not included in estimates of dry N deposition.

[b] The CASNET data are available online (http://www.epa.gov/castnet/). Gaseous $NH_3$ was not included in estimates of dry N deposition.

[c]The EMEP data are sourced from Endo et al. (2011), in which the dry and wet deposition amounts at each grid covering 27 EMEP countries were estimated by the unified EMEP models (Simpson et al., 2003).

[d] Only including the rural and background sites in NNDMN

References:

Du, E. Z., Vries, W. D., Galloway, J. N., Hu, X. Y., and Fang, J. Y.: Changes in wet nitrogen deposition in the United States between 1985 and 2012, Environ. Res. Lett.,9, 095004, doi:10.1088/1748-9326/9/9/095004, 2014.

Endo, T., Yagoh, H., Sato, K., Matsuda, K., Hayashi, K., Noguchi, I., and Sawada, K.: Regional characteristics of dry deposition of sulfur and nitrogen compounds at EANET sites in Japan from 2003 to 2008, Atmos. Environ., 45, 1259–1267,doi:10.1016/j.atmosenv.2010.12.003, 2010.

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Holland, E. A., Braswell, B. H., Sulzman, J., and Lamarque, J. F.: Nitrogen deposition onto the United States and Western Europe: synthesis of observations and models, Ecol. Appl., 15, 38–57, 2005.
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Sickles, J. E. and Shadwick, D. S.: Air quality and atmospheric deposition in the eastern US:20 years of change, Atmos. Chem. Phys., 15, 173–197, doi: 10.5194/acp-15-173-2015, 2015.
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Torseth, K., Aas, W., Breivik, K., Fjaeraa, A. M., Fiebig, M., Hjellbrekke, A. G., Myhre, C. L., Solberg, S., Yttri, K. E.: Introduction to the European Monitoring and Evaluation Programme (EMEP) and observed atmospheric composition change during 1972–2009, Atmos. Chem. Phys., 12, 5447–5481, doi: 10.5194/acp-12-5447-2012, 2012.
Vet, R., Artz, R. S., Carou, S., Shaw, M., Ro, C-U., Aas, W., Baker, A., and 14 authors: A global assessment of precipitation chemistry and deposition of sulfur, nitrogen, sea salt, base cations, organic acids, acidity and pH, and phosphorus, Atmos. Environ., 93, 3–100, doi:10.1016/j.atmosenv.2013.10.060.
Xing, J., Pleim, J., Mathur, R., Pouliot, G., Hogrefe, C., Gan, C. M., Wei, C.: Historical gaseous and primary aerosol emissions in the United States from 1990 to 2010, Atmos. Chem. Phys., 13, 7531–7549, doi: 10.5194/acp-13-7531-2013, 2013.

3. Another general comment is related with the terminology. Throughout the manuscript it is said that wet deposition was measured with precipitation gauges. However, in the discussion it is acknowledged that dry deposition in precipitation gauges can account for 20 to 40% of the deposition measured in precipitation. Thus, bulk deposition was in fact monitored and the terminology should be clarified in the manuscript.

**Response**: Thank you for pointing this out. We have clarified the difference between bulk and wet-only deposition in the revision. Indeed, the wet deposition fluxes determined with precipitation gauges were commonly regarded as bulk deposition fluxes which contain wet plus unquantifiable dry deposition (including both gases and particles) and therefore it should be higher than wet deposition (Liu et al., 2015). Throughout the revised manuscript we have changed “wet deposition” to “wet/bulk deposition”.

Reference:

Liu, X. J., Xu, W., Pan, Y. P., and Du, E. Z.: Liu et al. suspect that Zhu et al. (2015) may have underestimated dissolved organic nitrogen (N) but overestimated total particulate N in wet deposition in China, Sci. Total Environ., 520, 300–301, doi.org/10.1016/j.scitotenv.2015.03.004, 2015

4. The analysis of uncertainties in section 4.4 does not mention the uncertainties associated with the location and spatial coverage of the network. From Figure 1 it is evident that large areas of the country or islands lack of sampling points may be missing hotspots of nitrogen deposition and/or pristine sites. Some recommendations about this issue could probably be suggested.

**Response**: We agree that some hotspots of nitrogen deposition and/or pristine sites may be missing due to incomplete coverage of the network. We have added some recommendations about this issue in the uncertainty section 4.4 as follows:

“Although the NNDMN is the only long-term national deposition network to monitor both N wet/bulk and dry deposition in China till now, large areas of the country or islands lack of sampling points may be missing hotspots or pristine sites of N deposition. The implementation of an adequate monitoring program is also difficult at present in some regions (e.g., northwest China

and Tibetan Plateau). To address this issue, more new monitoring sites, covering regions with both extremely low and high $N_r$ emissions, should be set up in the NNDMN in future work."

5. P18368, L18: Include some measure of variability in the averaged nitrogen deposition fluxes in China to show that important reactive nitrogen deposition gradients exist in the country.
**Response**: Thank you for this suggestion. We have changed the sentence to "(…) Average dry and wet/bulk N deposition fluxes were 20.6 ± 11.2 (mean ± standard deviation) and 19.3 ± 9.2 kg N $ha^{-1}$ $yr^{-1}$ across China, with reduced N deposition dominating both dry and wet/bulk deposition."

6. P18374, L20 and S5: Which land use map was used to model the deposition velocities across China and how was the land use selected in each sampling point?
**Response**: We now state in the Section 2.5: "The model uses the land map of the Global Land Cover Characteristics Data Base Version 2.0 (http://edc2.usgs.gov/glcc/globdoc2_0.php), which defines the land types (e.g., urban, forest, etc.) at the native 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 may coarsely represent the local land characteristics at the monitoring sites. Future work using a single-point dry deposition model as for CASTNET (Clarke et al., 1997) would further improve the dry deposition flux estimates, but that requires concurrent *in-situ* measurements of meteorological variables which are not available at present."
Added reference:
Clarke, J. F., Edgerton, E. S., and Martin, B. E.: Dry deposition calculations for the Clean Air Status and Trends Network, Atmos. Environ., 31, 3667-3678, 1997.

7. P18376, L2: The comparison presented here is also true for other regions apart from NC, SE and SW?
**Response**: Not the same but the fact is that $NH_3$ concentrations were higher at urban and rural sites than at background sites in almost all regions. We have revised the sentence as follows:
"(…) In NC, SE and SW, the $NH_3$ concentrations at the urban sites (average for the three regions, 9.5 ± 2.1 μg N $m^{-3}$) were about 1/3 higher than at the rural sites (6.2 ± 2.3 μg N $m^{-3}$) and were almost twice of those at the background sites (4.8 ± 1.4 μg N $m^{-3}$), whereas in NE and NW $NH_3$ concentrations at the urban sites were lower (average two regions, 5.5 ± 3.2 μg N $m^{-3}$) than at the rural sites (8.8 ± 0.3 μg N $m^{-3}$) but 4.6-times greater than at the background sites (1.2 ± 0.5 μg N $m^{-3}$)."

8. P18376, L4: What about $NH_3$ levels in urban and background sites?
**Response**: We have added the following sentence here "Comparing land use types by region, annual $NH_3$ concentrations at the rural sites in northern regions (NC, NE and NW) were approximately equal, which on average were 1.8-times greater than the average of southern rural sites. In contrast, annual $NH_3$ concentrations at urban and background sites ranked in the order: SW > NC > NW > SE > TP > NE, and SW > NC > SE > NW > TP > NE, respectively (Fig. 3a)."

9. P18376, L9: The comparison of urban and rural areas for $NO_2$ also holds for other regions of China?
**Response**: Yes, we have revised this sentence to read: "In the six regions, the $NO_2$ concentrations at urban sites were 1.4-4.5 times higher than those at rural sites, and were even 2.0-16.6 times higher than the background sites (except for SW)."

10. P18379, L5: Were there any differences in reduced/oxidized nitrogen ratios depending on the site type (urban, rural or background)?
**Response:** Yes, we noticed some differences in reduced/oxidized N ratios based on site types: urban (1.2 ± 0.4) < rural (1.3 ± 0.5) < background (1.6 ± 0.4). However, such changes were not significantly different ($p$>0.05). We add the following sentence in the revision: "In our network, the $NH_x$ (i.e. wet/bulk $NH_4^+$-N deposition plus dry deposition of $NH_3$ and particulate $NH_4^+$)/$NO_y$ (wet/bulk $NO_3^-$-N deposition plus dry deposition of $NO_2$, $HNO_3$ and particulate $NO_3^-$) ratio at urban sites (from 0.8 to 1.8, averaging 1.2) was not significantly different ($p$>0.05) from rural (from 0.5 to 2.7, averaging 1.3) and background (from 1.0 to 2.5, averaging 1.6) sites."

11. P18379, L13: It is interesting that, despite reactive nitrogen concentrations in rural sites are

consistently lower than in urban sites, total annual mean deposition fluxes are quite similar. Have the authors any hypothesis to explain this result?

**Response:** The reason for this result should be mainly due to lower $V_d$ for $N_r$ species (esp. $V_d$ of $NO_2$ and $HNO_3$) at urban sites than at rural sites. We assume that urban areas act as greater $N_r$ pollution sources (more $N_r$ emission than deposition) compared with rural areas (based on per unit land area). On average, annual $N_r$ concentrations of $NH_3$, $NO_2$, $HNO_3$, and particulate $NH_4^+$ and $NO_3^-$ at rural sites were 14.8, 39.1, 32.1 19.3 and 30.3% lower than those at urban sites. Correspondingly, annual dry deposition fluxes of $NH_3$, $NO_2$, $HNO_3$, and particulate $NH_4^+$ and $NO_3^-$ at rural sites were 17.3, 11.2, 22.7, 11.9 and 28.8% lower than those at urban sites. Therefore, it is quite clear that comparable total dry deposition flux between urban sites (averaged 26.9 kg N $ha^{-1}$ $yr^{-1}$) and rural sites (averaging 23.0 kg N $ha^{-1}$ $yr^{-1}$) probably resulted from similar dry deposition fluxes of $NO_2$ and $HNO_3$ between rural (averaging 3.6 and 5.6 kg N $ha^{-1}$ $yr^{-1}$, respectively) sites and urban sites (averaging 4.0 and 7.2 kg N $ha^{-1}$ $yr^{-1}$, respectively), which can be attributed to somewhat higher deposition velocities of $NO_2$ and $HNO_3$ at rural sites (averaging 0.17 and 1.49 cm $s^{-1}$, respectively), compared with those at urban sites (averaging 0.12 and 1.22 cm $s^{-1}$, respectively).

12. P18379, L14: grassland sites-> background sites

**Response:** Changed as suggested.

13. P18380, L23: I believe the authors refer here to Figure S2 d and e

**Response:** Yes, and we have included a reference here to Figure S2 d and e in the revision.

14. P18381, L10: The discussion here would have benefited from an analysis of differences between regions across land use types. Are all the rural sites in China homogeneously affected by reactive nitrogen pollution?

**Response:** This is a good suggestion. All the rural sites in China were not homogeneously affected by reactive N pollution. We have revised the sentence to be as following: "Rural sites in this study also had relatively high concentrations of all measured $N_r$ species in air, altogether ranking in the order of NC > NE > NW > SE > SW (Fig. 3f). The higher concentrations in northern China are mainly due to the combined effect of high $NH_3$ emissions from N fertilized farmland (Zhang et al., 2008a) and urban air pollution (e.g. $NO_2$, $HNO_3$, $pNH_4^+$ and $pNO_3^-$) transported from population centers to the surrounding rural areas (Luo et al., 2013)."

References:

Luo, X. S., Liu, P., Tang, A. H., Liu, J. Y., Zong, X. Y., Zhang, Q., Kou, C. L., Zhang, L. J., Fowler, D., Fangmeier, A., Christie, P., Zhang, F. S., and Liu, X. J.: An evaluation of atmospheric $N_r$ pollution and deposition in North China after the Beijing Olympics, Atmos. Environ., 74, 209–216, doi:10.1016/j.atmosenv.2013.03.054, 2013.

Zhang, F. S., Wang, J. Q., Zhang, W. F., Cui, Z. L., Ma, W. Q., Chen, X. P., and Jiang, R. F.: Nutrient use efficiency of major cereal crops in China and measures for improvement. Acta Pedologia Sinica, 45, 915–924, 2008a (in Chinese with English abstract).

15. P18383, L4: Does this hypothesis work in the monitoring sites in China? In other words, was the NHx/NOy ratio in urban sites different from rural or background sites in this network?

**Response:** Yes, the $NH_4^+$-N/$NO_3^-$-N ratio in wet/bulk deposition still works for indicating the relative contribution of $N_r$ from agricultural and industrial activities to N deposition at monitoring sites in China. We found that $NH_4^+$-N/$NO_3^-$-N ratios in wet/bulk deposition followed the sequence of urban sites < rural sites < background sites across our monitoring network.

16. P18384, L19: There is no mention in this section of the discussion to differences in modelled deposition velocities for China compared with other estimates, as presented in table S4. This is also applicable in P18387, L8.

**Response:** We now state in the section 4.4 "The dry deposition fluxes were estimated by combining measured concentrations with modeled $V_d$. As summarized in Table S4, our estimates of dry deposition velocities for different $N_r$ species are generally consistent with the estimates in previous studies (e.g., Flechard et al., 2011; Pan et al., 2012). Some uncertainties may still exist in the inputs for dry deposition modeling."

We also have revised the sentence (P18387, L8) to read: "In previous work, dry deposition flux

was inferred from atmospheric $N_r$ concentrations and a literature-based annual mean deposition velocity (Shen et al., 2009), or reported by Luo et al. (2013) who did not consider the different dry deposition velocities of various $N_r$ species among different land use types. Clearly, in this study we have greatly improved the estimation of dry deposition, but further work is still required to increase the reliability and accuracy of N dry deposition values."

References:


Endo, T., Yagoh, H., Sato, K., Matsuda, K., Hayashi, K., Noguchi,I., and Sawada, K.: Regional characteristics of dry deposition of sulfur and nitrogen compounds at EANET sites inJapan from 2003 to 2008, Atmos. Environ., 45, 1259–1267,doi:10.1016/j.atmosenv.2010.12.003, 2010.

Luo, X. S., Liu, P., Tang, A. H., Liu, J. Y., Zong, X. Y., Zhang, Q., Kou, C. L., Zhang, L. J., Fowler, D., Fangmeier, A., Christie, P., Zhang, F. S., and Liu, X. J.: An evaluation of atmospheric $N_r$pollution and deposition in NorthChina after the BeijingOlympics, Atmos. Environ., 74, 209–216, doi:10.1016/j.atmosenv.2013.03.054, 2013.

Shen, J. L., Tang, A. H., Liu, X. J., Fangmeier, A., Goulding, K. T. W., and Zhang, F. S.: High concentrations anddry deposition of reactive nitrogen species at two sites in the North China Plain, Environ. Pollut., 157, 3106–3113,doi:10.1016/j.envpol.2009.05.016, 2009.


17. Figures 2 and 4: Vertical lines could be included to separate regions or even land use categories within regions in order to ease comparisons.

**Response:** Thanks for the suggestion. In the revision, Figure 4 was changed to Figure 5. We have added vertical lines in Figures 2 and 5 to separate regions and land use categories within regions. Please see the revised Figures 2 and 5 as below.

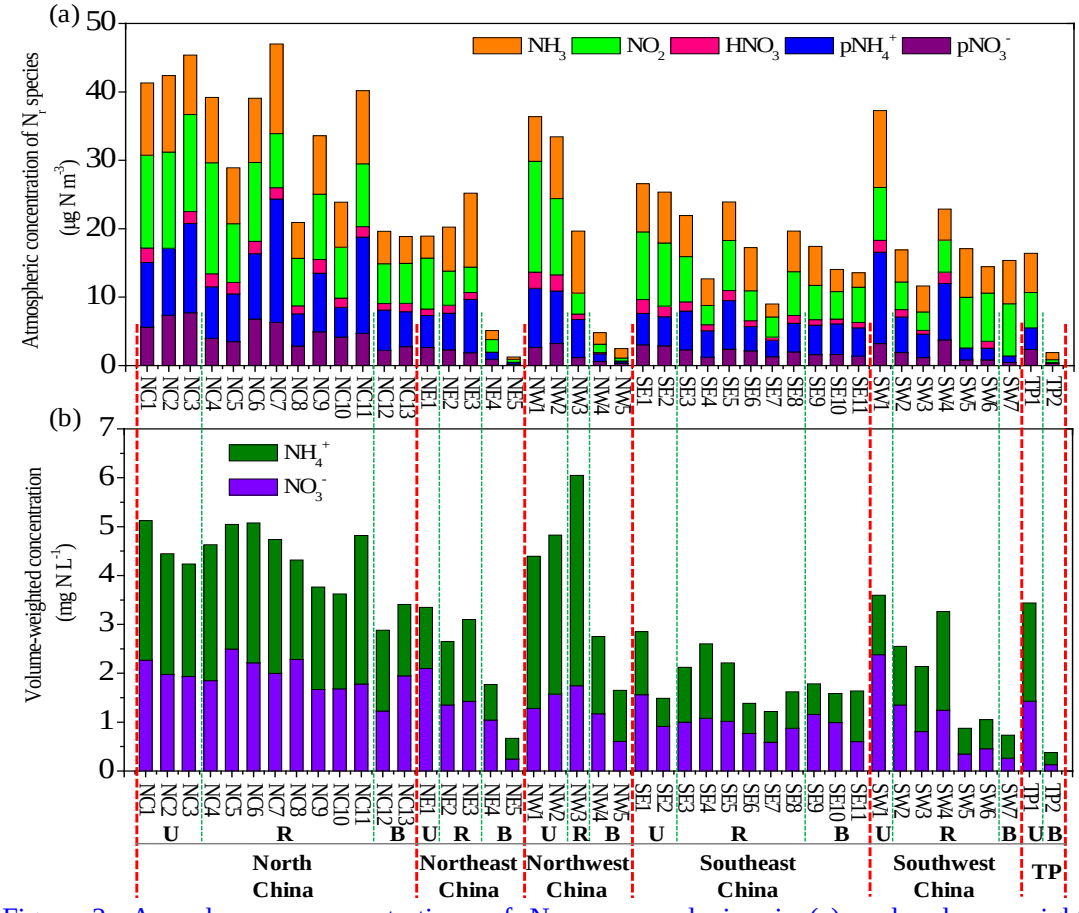


Figure 2. Annual mean concentrations of $N_r$ compounds in air (a) and volume-weighted concentrations of inorganic nitrogen species in precipitation (b) at all monitoring sites. U, R, and B denote urban, rural, and background sites, respectively. TP denotes the Tibetan Plateau.

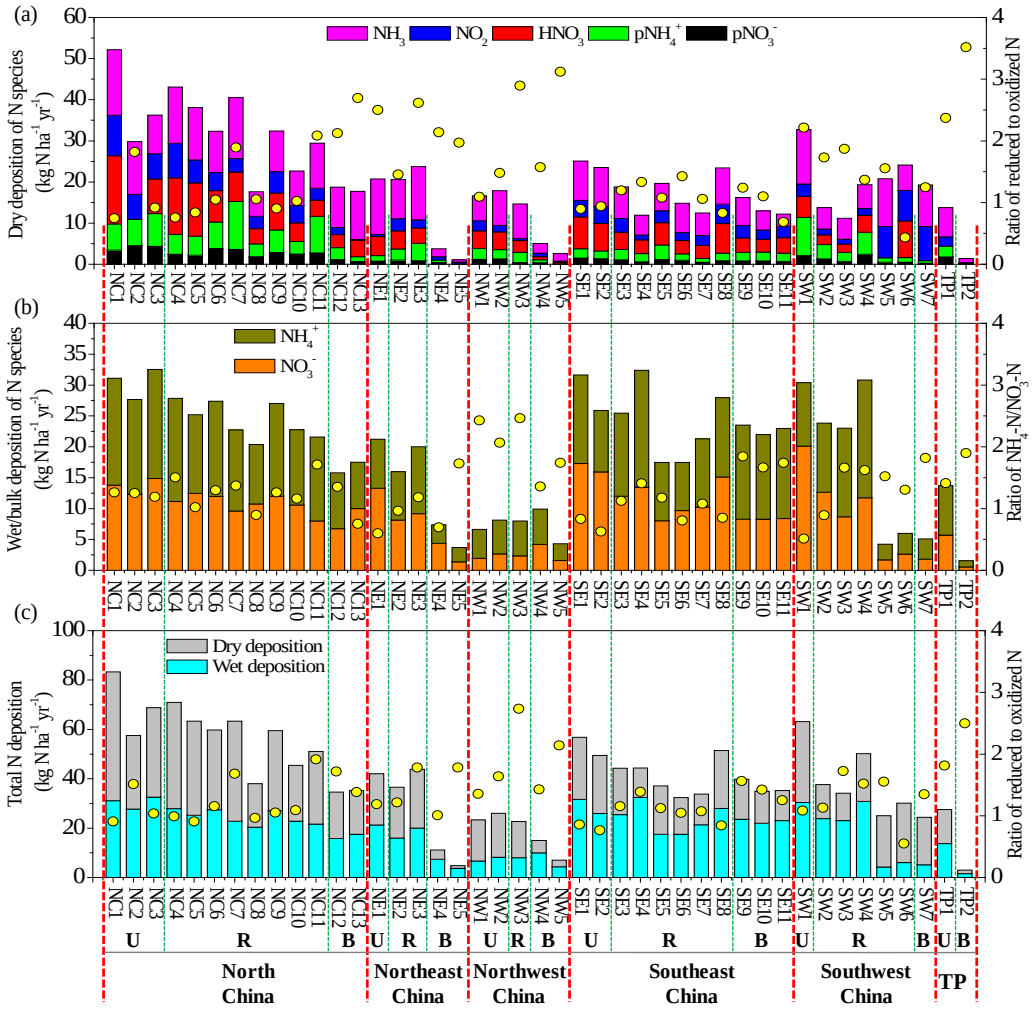


Figure 5. Annual deposition flux of various $N_r$ species at the forty-three selected sites in China: (a) dry deposition flux; (b) wet/bulk deposition flux; (c) total deposition flux. Yellow dots denote ratios of reduced N to oxidized N in dry deposition (a), $NH_4^+$-N to $NO_3^-$-N in wet/bulk deposition (b) and/or reduced N to oxidized N in total deposition (c) at all sampling sites.

18. Supplement S2: Renumber subsections as 2. X.

**Response:** We have renumbered subsections as suggested: S2.1 Sampling sites in north China (NC), S2.2 Sampling sites in northeast China (NE), S2.3 Sampling sites in northwest China (NW), S2.4 Sampling sites in southeast China (SE), S2.5 Sampling sites in southwest China (SW), S2.6 Sampling sites in the Tibetan plateau (TP).

19. Supplement S2.1: Thirty -> Thirteen

**Response:** Changed as suggested.

20. Supplement S2.5: 2 rural sites -> 5 rural sites

**Response:** Changed as suggested.

21. Supplement S5: The tables referenced here should be S3 and S4

**Response:** Yes, we have referenced the tables S3 and S4 in Supplement S5. The text is revised to be: "the monthly $V_d$ at each site was averaged based on the hourly dataset for further estimation of dry deposition flux of each $N_r$ species during the observation, which was statistically summarized according to land use type and is presented in Table S3. Annual mean dry deposition velocities of $N_r$ species for three land use types in this study, averaged from monthly mean values, fit well into range of annual values calculated and used for similar land use types in other studies (Table S4 of Supplement)."