

Anonymous Referee #2

This paper presents a statistical summary and discussion of measurements of components of reactive nitrogen (Nr) in the air and in bulk deposition from the 27 sites of a national network that are located in the eastern part of China. The measurement dataset spans the 5-year period from 2011-2015 inclusive. Measurements are also converted into estimates of wet and dry deposition. The authors analyse various spatiotemporal aspects of the concentrations and deposition dataset including seasonality, trends over the 5-year period, and a comparison between sites in the northern half and the southern half of eastern China. The authors supplement the analysis of measurement data with some GEOS-Chem model runs to explore source contributions to Nr in this region. Discussion includes implication for policymakers concerning the different trends in emissions of Nr versus concentrations and deposition of Nr and of the need to include emissions of NH₃ in emissions reductions planning. The dataset is comprehensive. The presentation of the results is thorough and the text and figures and tables are very clearly presented. There is an extensive discussion. The data are of importance for understanding Nr in eastern China.

Response: Thanks for the recognition of our contribution. Below we provide a point-by-point response to the species comments, together with proposed changes in the revised manuscript (in blue).

Specific comments:

Five years is not a long time period to attempt to discern ‘true’ long-term trends in concentrations of atmospheric species. The authors recognise that their time period is short in respect of this aspect of their analysis but they could phrase relevant parts of their text to be more cautious about conclusions on long-term trends.

Response: The suggestion has been implemented in the revision.

L124: Replace “subsequence” with “subsequent”

Response: Agree and done.

L207: It is not clear what is meant by the phrase “where field sampling was carried out after the year 2010”. Is this intended to mean that at some sites the measurements

did not begin until after 2010?

Response: We are sorry for confusing the referee. It means that at eleven sites the measurements begin after the year 2011. We now state that “...where field sampling was carried out after the year 2011 (i.e., the years between 2012 and 2015) and/or interrupted during the period due to instrument failure (details in Table S1, Supplement)”.

L271-2: There is a contradiction between a sentence that states that IASI data up until 31 December 2015 was used and the following sentence that states that data only up until 30 September 2014 was used.

Response: There was a wrong expression in this sentence. Actually, we used the daily IASI-NH₃ data from 1 January 2011 to 31 December 2015 for the spatial analysis, and from January 2011 to 30 September 2014 for temporal analysis.

We now state that “The daily IASI-NH₃ data (provided by the Atmospheric Spectroscopy Group at Université Libre De Bruxelles, data available at <http://iasi.aeris-data.fr/NH3/>) from 1 January 2011 to 31 December 2015 was used for the spatial analysis in the present study. For the temporal analysis, we used the IASI_NH₃ from 1 January 2011 to 30 September 2014 because an update of the input meteorological data on 30 September 2014 had caused a substantial increase in the retrieved atmospheric NH₃ columns.”

Table 1: (1) State in the caption or footnote what the significance test is testing, i.e. that it is testing for significant difference in mean concentration of a pollutant at a given site type between the northern region and the southern region. (2) The footnote should read LUY not LSY to be consistent with column heading.

Response: We now state in the footnote that “* and ** denote significance at the 0.05 and 0.01 probability levels for difference in annual mean N_r concentrations at a given site type between northern and southern regions, respectively.”

Also, we uniformly used “LUT” as an abbreviation of land use types in the footnote and column heading.

Figure 2: The reader is referred to Table S1 in the supplement for the number of sites for each land use type in each region, but cannot the reader be directed more easily to

Table 1 in the main paper for these numbers?

Response: The reader cannot be directly referred to Table 1. For comparison between the periods 2011-2012 and 2013-2015, the sampling sites for land use types shown in Figure 2 have continuous 5-year (2011-2015) measurements (in total 21 sites for dry measurements, and 16 sites for wet/bulk measurements). For spatial comparisons in Table 1, the annual mean concentrations of N_r species in air and precipitation for land use types were calculated based on measurements at all 27 sites.

Figure 3: (1) I assume the data shown are the means for the 5-year period, in which case it may be helpful to make this explicit in the opening sentence thus: “Seasonal mean concentrations averaged over 2011-2015 of: : :.”. (2) As for Figure 2 (**should be 3?**), can the text “in Table S1 in the supplement” be replaced more directly with “in Table 1”. (3) The last part of the caption should refer to significant differences between “seasons” not “sites”.

Response: In the revised paper, we rephrased the start of caption of Figure 2 to “Seasonal mean concentrations averaged over 2011-2015 of...”.

We replaced “Table S1 in the supplement” by “Table 1”, as seasonal averages were calculated based on measurements at all 27 sites. Also, we changed “sites” to “seasons”.

Figure 4: The same 3 comments as made above in connection with Figure 3.

Response: In the revised paper, we have made corresponding corrections on Figure 4 according to the referee’s comments on Figure 3.

Table 2: Same comments as for Table 1.

Response: In the revision we made corresponding corrections on Table 2 according to the referee’s comments on Table 1.

Figure 5: Can the reader be directed to Table 2, rather than to Table S1 in the supplement, for the number of sites of each type in each region.

Response: The reader cannot be referred to Table 2. For details, please see our response to similar comments on Figure 2.

Figure 7: Same comments as for Figure 3 (but with substitution of reference to Table 2 rather than to Table 1).

Response: In the revised paper, we made corresponding corrections on the caption of Figure 7 according to the referee's comments on Figure 3.

Figure 8: Same comments as for Figure 7.

Response: The reader cannot be directly referred to Table 1. Please see our response to the referee's comment on Figure 2.

L598: Rephrase start of sentence to "Eastern China is a highly industrialized. : :"

Response: Agree and done.

L 761: In comparing ion balance, presumably the (molar) concentration of NH_4^+ was compared against the sum of the molar concentrations of NO_3^- and TWICE the molar concentration of SO_4^{2-} ? The factor 2 is missing from the text and from the axis title of Figure 10f.

Response: Thank you for pointing it out. In the revised paper, we analyzed the correlation of NH_4^+ with the sum of $\text{NO}_3^- + 2\text{SO}_4^{2-}$. Also, Figure 10f was redrawn and the corresponding sentences were changed, now read as: "At urban and rural sites, monthly mean $p\text{NH}_4^+$ concentrations significantly positively correlated with the sum of $p2\text{SO}_4^{2-}$ and $p\text{NO}_3^-$ concentrations (Fig. 10f). However, the slopes of regression equations between them were both smaller than unity (0.35 and 0.46 at urban and rural sites, respectively)...". In addition, we changed "Table S1" to "Table 1" in the caption of Figure 10.