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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Spatial-temporal patterns of inorganic nitrogen air concentrations and deposition in eastern China" *by* Wen Xu et al.

Anonymous Referee #1

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This paper presented spatial and temporal trends of reactive nitrogen species in air, precipitation and deposition in eastern China. Some of the spatial patterns described in the paper are interesting, such as the higher rural concentrations observed in the northern region compared to the southern region. The paper discusses the need for ammonia emissions policies to reduce reactive nitrogen in air and in deposition. The nitrogen datasets from this ground-based measurement network is valuable; however, a longer dataset needs to be collected before it is suitable for analyzing temporal trends. With only five years of data, this could be the main reason why most of the annual trends were not significant. Another concern that I have is a lack of explanation on the causes of the spatial and temporal trends, which requires analyzing the reactive nitrogen data with other datasets. The discussions seems biased towards ammonia

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Discussion paper

emissions reductions as a more effective means of reducing reactive nitrogen than NOx and SO2 emissions reductions, but I don't think there is enough evidence in this study supporting this conclusion.

Specific comments

Line 77: Define Nr since this is the first time that it is mentioned in the paper.

Line 83: Be more careful about linking deposition of N to increased greenhouse gas emissions. The referenced article only suggests that the nitrogen cycle is coupled with the carbon cycle and climate variation; however, the latter could be influenced by many factors.

Lines 110-111: The analysis presented by Xu et al. (2015) is quite similar to this study in terms of the measurement network, nitrogen species, time period, and site categories analyzed. The authors should discuss the previous study and explain how this study is different to avoid presenting a duplicate analysis.

Lines 148-156: This is where it might be appropriate to discuss the previous study, Xu et al. (2015), and emphasize the new work that will be shown in this study.

Line 170: Suggest using "and" instead of "resulting in" because this sentence suggests there is a relationship between economic development and nitrogen emissions. If there is such relationship, please elaborate.

Lines 220-221: You need to be clearer about what type of deposition the open sampler collects. Why is it only "some" dry deposition? Isn't the sampler open to the atmosphere which means it is collecting total deposition?

Line 271: The dates here should be January 2011 to 30 September 2014 because you stated in the next sentence that the data after 30 September 2014 were not used.

Lines 347-349: The concentration ranges are not clear. Is it the range of the mean concentration between sites or between years?

Lines 350-352: What is the reason for the lower concentrations at urban sites in the northern region?

Lines 359-365: I suggest analyzing which nitrogen specie was particularly higher between urban and rural sites and between northern and southern regions because this would provide some insight whether the patterns are related to a specific type of emission source.

Lines 371-374: What is the reason for the higher precipitation concentrations in northern rural sites compared to southern rural sites? Is this related to the higher air concentrations of Nr species in northern rural sites?

Lines 383-401: Presenting only the annual trends in the Nr concentrations is not enough. I think that additional analysis with other variables is necessary to attempt to explain the trends in Nr concentrations (e.g. emissions data). As stated in the introduction, one of the goals of this study is to assess the effectiveness of emissions control measures.

Lines 411-416: Any relationships between precipitation concentration and air concentration trends?

Lines 422-436: What is the reason for the seasonal trends? E.g. changes in emissions, meteorology, and/or air mass patterns? I think these other factors need to be analyzed in order to understand what is influencing the seasonal trends.

Line 478: Instead of presenting bulk deposition, is it possible to estimate wet deposition fluxes by subtracting the dry deposition fluxes from bulk deposition? This allows a comparison between wet and dry deposition.

Lines 462-481: How do these deposition fluxes compare to other parts of the world over this recent time period? I also recommend plotting the spatial distribution of the deposition fluxes on a map because it is difficult to get a sense of the spatial patterns from the text and numbers in this paragraph.

Line 572: If you sum dry and wet/bulk deposition fluxes, the total deposition will be overestimated because the bulk deposition already includes dry deposition.

Figure 8: Could you discuss the results in Fig. 8b? All of the previous trends were urban > rural > background. I find it interesting that the trend for the ratio of reduced to oxidized N is reversed. Also, why is this ratio important?

Section 4.1 and Figure 9: The correlation results show there is good agreement between satellite and ground-based observations. Can you quantify the differences using metrics? E.g., what are the percent differences for each month and annually? The correlation may be good, but the actual concentrations can still be different. Given the good relationship between satellite and surface measurements, are long term satellite data available for conducting temporal trend analysis?

Section 4.2: There is too much speculation on the causes of the seasonal trends. Most of the discussion is based on what previous literature reported. I think you need to analyze other datasets to examine the factors affecting the Nr trends.

Line 725: Could you provide the actual emissions amount from x tonnes in 2010 to y tonnes in 2014? Even though the emissions declined by a certain percentage, the actual emissions amount in 2014 might still be very large. If this is the case, then you will likely not observe a significant decrease in Nr concentrations.

Lines 733-734: How much ammonia is emitted relative to NOx and SO2? I would think NOx and SO2 emissions are higher than those of ammonia. If this is the case, wouldn't NOx and SO2 emissions reductions have larger effects on Nr?

Lines 757-773: I don't think you can really say that ammonia emissions reductions are more important than NOx and SO2 emissions reductions. If ammonia emissions have been increasing, why is the Nr concentration in air and precipitation not increasing (many of the trends were not significant in sect. 3.2)? Also, is it possible that the NOx and SO2 emissions reductions are not large enough? See earlier comment about

the actual emissions amount for NOx and SO2 could be very large despite 9-13% decrease in emissions. Is it appropriate to make this conclusion given that five years of data were analyzed? You also discussed how ammonia neutralizes acidic gases and plays a role in limiting Nr. However, it does not mean that this process is more effective than reducing NOx and SO2 emissions which decrease the formation of acidic gases in the first place.

Lines 775-783: This paragraph needs to mention the NOx and SO2 emissions in the northern region especially given the increased emissions for winter heating? How does they compare with ammonia emissions over an annual basis? A map of the spatial distribution of the ammonia emissions and agriculture activity levels would easily demonstrate that these are higher in the northern region.

Line 801: This should be Fig. S12

Line 803: This should be Sect. S2

Lines 799-811: I think the model simulation and results require further analysis and discussion. The model apportions the contributions of various sources to ammonium and nitrate deposition and suggests agricultural activity is the main contributor. There needs to be more details on the model scenario (e.g. NH3 and NOx emissions estimated from the various sources). Is the larger contribution from agriculture due to larger emissions relative to other sources or is it because area sources have larger impact than point sources in the model? Also, to support the idea that NH3 emissions reductions are important in reducing Nr deposition, you could perform a sensitivity analysis using different scenarios of NH3 emissions reductions for future years.

Line 809: What do you mean by improper fertilizer application? Do you mean too excessive? How much fertilizer is applied annually and is this amount much higher than normal? More background on this issue would be useful.

Line 884: Do you have annual precipitation amounts from weather stations, which can

show whether interannual variability in precipitation amounts affect wet deposition?

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