Interactive comment on “Occurrence of gas phase ammonia in the area of Beijing (China)” by A. Ianniello et al.

Anonymous Referee #1

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General comments

The paper by Ianniello et al. is well written and provides a thorough insight into the measurements taken, the equipment used and the statistical evaluation conducted. The discussion is balanced and well founded in existing literature and the current state-of-the-art of measuring ammonia and interpreting results. There are some gaps and shortcomings, however, which need to be addressed to make the paper more robust and scientifically rounded before publication:

1) The authors conclude, that “NH3 concentrations show regular seasonal variations, having significantly higher summertime concentrations”. While the second half of this sentence is undisputable, the first claim is quite far reaching, taking into account that it is based on two (!) field campaigns within one winter and one summer period only (23
resp. 30 cases). To underpin this conclusion, either more campaign data across different years, or evidence from previous measurements conducted in the same area with supporting data would be required. The references provided and discussed, however, are only used to underpin correlations of different parameters or providing support for individual patterns observed, not for this overall conclusion. It is clear, that the availability of such measurement data for China/Beijing is all but great and it may be difficult to obtain. Yet, to conclude on regular seasonal variations, two periods in one year is hardly justifiable at this point.

2) The conclusion, that no diurnal variation could be observed at all in either the winter or summer period is surprising and contradicts the temperature effect discussed elsewhere in the paper. The influence of temperature on observed NH3 levels is missing in the conclusions, by the way. Unfortunately, only Fig. 3 enables the reader to assess this claim, and only for the winter period. At minimum, one figure for winter and one for summer, over several days (a week?) should be displayed. A closer look then at Fig. 3 then does suggest a peak at noon, likely dependent on temperature increase and human activity (NOx peak is similar) on day 1 and a less pronounced peak on day 2, somewhat earlier. A quick calendar check reveals that Feb 9, 2007 was a Friday (day 1), Feb 10, 2007 a Saturday (day 2), which may explain the lack of a dominant peak on the second day due to less traffic, but a comparison with hourly temperatures would be prudent as well to investigate the reason for this missing diurnal pattern. It can be safely assumed that the diurnal pattern will be even more pronounced in the summer period. In addition, there is likely a weekly cycle due to the influence of e.g. traffic emissions.

3) The influence of different sources to observed levels is highlighted at several points in the paper and features as well in the conclusions (correlation with other trace gas emissions from transport). But while specific events during the campaigns are attributed to the influence of e.g. agricultural sources, depending on wind direction/speed, resp. road transport or other local sources (boundary layer conditions),
even a short discussion of the spatial domain in which the measurements have been taken is missing. A map of the domain within which the measurements have taken place, locating the measurement site and – overlaying the wind roses presented in Figs. 2 and 5 – the location of potential strong sources of NH$_3$ (agricultural fields/farms, major roads/highways, power plants etc.) would be essential to underpin these assumptions now only based on peaks/events observed in the data.

4) Analysing other trace gases to detect correlations between NH$_3$, NOx and CO is a viable and sensible approach to allow for interpretations regarding source-attribution of observed events. The discussion of the results of these comparisons in this paper could be more detailed and thorough. In addition to that, literature quoted for non-transport sources (Sutton et al. 2000 for instance) is valid, but not the primary literature one would expect when looking at NH$_3$ emissions from for instance road transport sources. There are plenty of publications discussing NH$_3$ emission factors from power generation and mobile sources that has not been cited and seems not to have been reviewed (e.g. Heeb et al., 2007, doi:10.1016/j.atmosenv.2007.12.008, or COPERT, http://lat.eng.auth.gr/copert/). As an aside, the comparison with CO emissions may not be straightforward at all, esp. with regard to difference in summer/winter (cold start emissions from vehicles, small combustion sources).

In addition to that, it does not become clear from the description how the authors account for the specific situation of the source composition in China, resp. Beijing. The technologies used in road transport vehicles (e.g. non-catalytic converter equipped, early simple and advanced catalytic converters) and in stationary sources (SCR/SNCR, primary measures etc.) will substantially affect the ratio of different pollutant emissions and hence the conclusions that can be drawn from correlations observed or not observed. A table with an (even coarse) emission inventory for China/Beijing with the relative contributions to each of the major source groups and how this would affect the expected concentrations to be observed would be needed to put the currently unfounded assumptions explaining observed events on a stronger footing. In this context,
Figs 7 and 8 do not offer a substantial contribution to the discussions and could be omitted/combined.

Specific comments

These 4 points raised above lead to the following specific questions that should be thoroughly addressed before publication is considered:

1) Are the 2 periods of measurements sufficient to conclude on the seasonality of NH3 concentrations in Beijing? If not, what other evidence can be used to underpin this claim.

2) What is the underpinning evidence to conclude that no diurnal variation has been observed? Provide figures to either prove this claim, or reassess and discuss this in more detail.

3) Where are major sources (source groups) located in relation to the measurement site and with regard to wind direction/speed and distance from the source? Provide (a) map(s) which indicate source regions to underpin the claims regarding source attributions.

4) What are the relative contributions to the different sources of other trace gases used in comparison to NH3 concentrations observed? Take a look at inventory data to underpin the source attribution and hence improve the interpretation of correlations observed/not observed with e.g. NOx and CO.

Technical corrections

The following technical corrections/points should be addressed:

- p.14211, l.8/9: “is concerning with climate change” – rephrase/reword, not clear what is meant

- p.14211, l.12/13: “causing consequences to remote sensitive ecosystems”, ambiguous, could be phrased better and more concrete
- p.14211, l.24: “is different from developed countries”, in what sense is it different? the way it is phrased suggests that in China, the share of NH3 is higher than in developed countries, while e.g. in Europe in 2005 (based on EMEP data), agriculture contributes more than 90% to total NH3 emissions. Reconsider/rephrase.

- p.14212, l.6: add “emissions” after NH3 and “insolation” should be “insulation”?  

- p.14212, l.8/9: “But the interactions between these factors are so complex that no uniform result is forthcoming” – this sentence is hard to understand, what does it mean? In a scientific paper, one would expect a thorough discussion of parameters that are included in the analysis and those that are not, for obvious reasons; hence, this sentence should be omitted and instead replace with a short paragraph on what is within the scope of the analysis, and what beyond.

- p.14212, l.20: “control systems” better as “emission abatement technology”

- p.14213, l.3: “winds” does it refer to “wind speed/direction?” please clarify

- p.14214, l.9: add “(PKU) after “Peking University” as you use the abbreviation later, without introducing it

- p.14215, l.5: Rephrase the start of this sentence to read “Although the focus of this paper is on ...”

- p.14217, l.8-10: How have day/night hours been grouped on 18:00-6:00/6:00-18:00, or on sunrise/sunset, as indicated in the next sentence? Unclear, please clarify

- p.14217, l.20: “The source of NH3...” which source is referred to? Explain and elaborate a bit more, the following sentence is making quite strong assumptions based on literature.

- p.14219, l.4/5: “solvent use”? The contribution of solvent use to total NH3 emissions should be marginal (less than 2%), in addition, solvent use has not been mentioned in any discussion of NH3 sources before, so how does it enter the picture here? Explain,
or rephrase, please.

- p.14222, l.17/18: the sentence starting “These results confirm ...” does not mention agricultural emissions at all, which is surprising for summer observations of NH3. Explain why agricultural emissions are not among the parameters driving NH3? In addition, the use of “evolution” here is ambiguous, should be replaced by a more concrete reference to e.g. variability, concentration levels etc.

- p.14223, l.2: “didn’t” should be replaced by “did not”

- p.14234: Fig. 4 does not add much evidence/enlightenment to the paper, could be well combined with Figs. 7 and 8 into one set of 3 or 4 correlation plots in one figure.

- p.14236: Fig. 6. is quite cluttered and at its current size makes is hard to read or distinguish the relevant details for the reader. If not possible to increase the size, consider splitting

- p. 14237/8: Figs. 7 and 8 could be omitted/combined with Fig. 4 (see above)

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