

Interactive comment on “Atmospheric ammonia variability and link with PM formation: a case study over the Paris area” by Camille Viatte et al.

Anonymous Referee #1

Received and published: 5 July 2019

This manuscript uses observations from two independent satellites to assess the role of NH₃ in springtime particle pollution episodes in the Paris region by examining the seasonal and interannual variability (IAV) in NH₃ columns over northwestern Europe. The observations are compared to simulations from the CHIMERE chemical transport model. In general, the authors do a good job of reviewing the existing literature to provide context for their results, but it would be useful if they could include a comparison with the study of Schiferl et al. (2016), which examines seasonal cycles and IAV of NH₃ over the US.

In Section 2.2, it is important that the authors report what proportion of the column observations from each satellite were below the limit of detection and how those data were incorporated into the monthly means used throughout the paper. If observations

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below the limit of detection were discarded, then the resulting monthly means will be biased high. It would then be important to filter the model output in a similar way to ensure that the observation-model comparison is more appropriate.

A general concern in Section 3.1 is the confidence with which the authors interpret the causes contributing to the seasonality and IAV of the ammonia columns. In many cases, the explanations provided by the authors seem reasonable, but unless there is conclusive proof, the language should be toned down to indicate that these are possible/likely explanations rather than the only ones:

Lines 282-301, a handful of data are provided to describe farming practices in different regions, but not in a consistent way. What evidence is there that the factors described are the most important in causing the spatial and temporal patterns observed?

Lines 313-314 How do crop type and phenological stage impact ammonia concentrations leading to interannual variability?

Lines 330-333 These seem like plausible explanation for the impact of precipitation amount of ammonia columns, but is there direct evidence that they are the only (most) important factors?

Lines 334-335 The relationship between gas phase ammonia and temperature should be exponential based on the temperature dependence of its volatilization (either vapor pressure or effective solubility). Does the correlation coefficient change if a non-linear fit is tried?

In Section 3.2, the authors compare ‘standardized’ monthly means for the years 2014 and 2015 between the two satellite products and the model. More explanation should be provided about how these standardized means were calculated. Do the emissions used in the model differ between the two years? This would be useful to know to help in interpreting the variability produced by the model.

Lines 371-382 This discussion is a bit confusing because initially the values quoted

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from the correlation plots of are the coefficients of determination, and then the comparison is restricted to select months and the values quoted are the slopes. I would recommend quoting the r^2 values for both, to make it more clear that the coefficients of determination did not increase significantly when the months were restricted. Also, the fact that the slope is close to 1 is not that meaningful since each dataset has already been standardized.

In Section 3.3, which focuses on the role of NH_3 in producing $\text{PM}_{2.5}$ in the Ile de France region, the analysis is overly simplistic. Why have the $\text{PM}_{2.5}$ observations included in the analysis been restricted the measurements between 9 and 11 am? This time interval is particularly challenging to interpret because of the impacts of primary emissions and the role of the rapidly changing boundary layer height. It seems like a poor choice of time window to focus on a phenomenon that is influenced by long-range/regional transport of a precursor species like NH_3 . The role of temperature and relative humidity on the formation of ammonium salts is well-described by thermodynamic relationships. Statements like those on Lines 504-509 and not fully accurate.

Specific comments: Line 46 – ‘biochemical’ should perhaps be ‘biogeochemical’

Line 63 – ‘related to’ should be ‘relative to’

Line 111-114 – It would be helpful to reword the sentence slightly, to clarify that all of the studies being referenced were carried out in Paris.

Figure 1 – The coloring of the map by the emissions is not easy to see. The colors become a very different shade on the map than on the legend. Is it possible to use a map that doesn’t have a green background, or to make the emissions coloring more opaque?

Figure 6 – would be helpful to have the same months identified on the axis for each year

References

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Luke D. Schiferl, Colette L. Heald, Martin van Damme, Lieven Clarisse, Cathy Clerbaux, et al. Interannual variability of ammonia concentrations over the United States: sources and implications. *Atmospheric Chemistry and Physics*, 16 (18), 12305-12328, 2016.

Interactive comment on *Atmos. Chem. Phys. Discuss.*, <https://doi.org/10.5194/acp-2019-138>, 2019.

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