

Review of “Measurement report: Ammonia in Paris derived from ground-based open-path and satellite observations”

Referee: The manuscript discusses relatively long-term NH₃ measurements from the IASI instrument and a miniDOAS instrument located in the center of Paris. Correlation, seasonal and weekly cycles are discussed as well as the potential sources of the NH₃ emissions.

The manuscript is well-written and well structured. The findings do not introduce substantial new discoveries. The finding that traffic is a significant source of NH₃ in the Paris region would be more convincing with a better placement of the miniDOAS or when NH₃ measurements from more stations would have been included. Such measurements seem to be available in the Paris region. Although I find the study a bit lightweight for ACP, I still recommend its publication. However, revisions (detailed below) need to be conducted in the paper before publication. Please note that all questions/comments below should not only be addressed in the author’s reply, but also in the manuscript.

Authors: we would like to thank the referee for his insightful and constructive comments. We have addressed the referees’ suggestions and questions to improve the manuscript. The original review comments are shown in black, our responses are shown in blue.

General comments

p.4 l. 103: One of the main comments is that a clear motivation is needed why you put the instrument at an altitude of 40 m above ground level. You are far less sensitive to the traffic related emissions you want to monitor as well? In the introduction (p.3 l.59) it is stated that this is badly monitored.

Our main goal in this work was not to only study NH₃ traffic related emissions in Paris. Our first motivation is to assess the temporal evolution of NH₃ concentrations over the megacity using the first relatively long-term NH₃ observations (2.5 years, day and night) and disentangle the different sources when studying NH₃ budget at different timescales. This is why we chose to install the mini-DOAS instrument in the Paris city-center (48.8°N, 2.3°E) at the QUALAIR super-site at 40 meters above ground level (<https://qualair.fr/index.php/en/english/>) so that the NH₃ observations footprint is representative of the Paris capital.

We have clarified this by adding this following sentence at the beginning of section 2.1 : “NH₃ concentrations are measured since January 2020 in the Paris city-center (48.8°N, 2.3°E) using the ground-based miniDOAS instrument located at the QUALAIR super-site (40 meters above ground level, <https://qualair.fr/index.php/en/english/>). This dataset constitutes the only current continuous (day and night) NH₃ observations at high temporal frequency representative of the Paris megacity.”

Fig.1 top panel: It is difficult to see the NH₃ distribution in the Paris area on this map. It would be helpful to also provide a zoom for the red, blue and green box (maybe in appendix).

As mention in the manuscript, we demonstrated that that agricultural activities in the surrounding areas control the variability of ammonia in Paris on average over the whole season. This map is computed using the whole IASI dataset (2.5-years) which reveals the main NH₃ agricultural sources

around Paris. We have tried to improve this figure (see Figure R1) but we decided to keep the old one in the revised document.

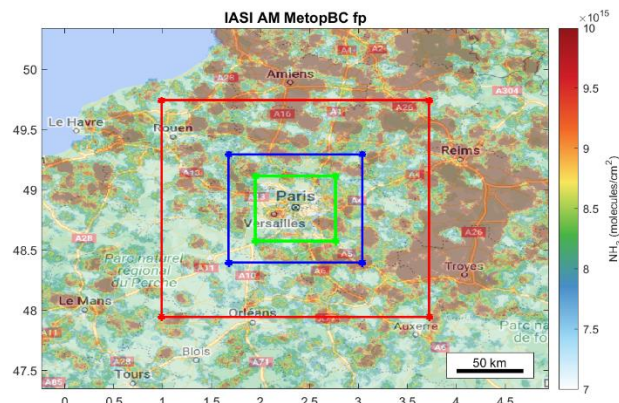


Figure R1: 2.5-years average of IASI NH₃ column distributions (from January 1st 2020 to May 31st 2022).

Fig.1 lower panel: I'm not sure if applying these different spatial criteria make much sense. Also it would be expected that when applying more strict spatial constraints (30 km) that the agreement would be better, due to better co-location, which is not the case. Please explain.

Fig.1 lower panel: Moreover, why don't you make use of the maximum spatial resolution of IASI, i.e. comparing with the closest pixel or smaller radius especially as you mention city-scale studies earlier on.

Fig.1 lower panel: Please explain the spread of data points over the seasons. Why do you have significantly more points in spring than in summer and other seasons?

In this study, we found that the main source of atmospheric NH₃ in Paris is its transport from the surrounding agricultural regions resulting in higher correlations at larger scales between the ~12-km IASI footprints and the miniDOAS observations. Road-traffic emissions are noticeable only when agricultural related emissions are weak.

In addition, we have stated in the manuscript that the correlations at 30 km are not significant (p -value > 0.5) for two seasons (winter and fall) and the number of pairs are reduced by a factor of two compared to comparisons at 100 km.

IASI pixels size varies from 12-km diameter footprint on the ground at nadir to 40 km diameters depending on the satellite observational angle. For cloud free situations, IASI can measure the atmospheric radiation down to the ground so only the data that are not contaminated by clouds in the field-of-view were selected in this analysis. The variability of IASI pixels sizes and the presence of clouds can be seen for one day (3 May 2022) in Figure R2.

The trade-off between analysis box size and number of IASI samples justifies our choice of a 50 km window.

Finally, there are less pairs of comparison in fall and summer because the timeserie extends from January 2020 to May 31st 2022 with 166, 134, and 91 IASI days of observations in 2020, 2021, and 2022, respectively.

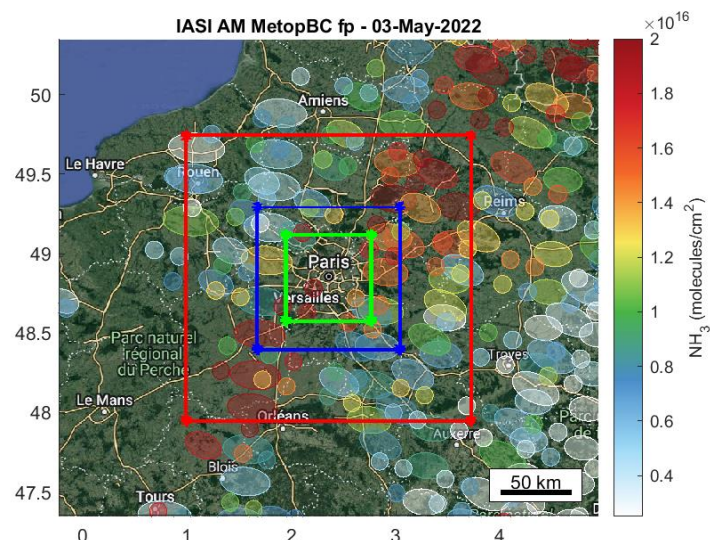


Figure R2: example of IASI morning observations with Metop B and C for one day (3 May 2022) in the domain of study.

We have added the following statements in the revised manuscript to discuss the spatial variability of NH_3 observed with the two datasets:

“The high correlation in spring between the two datasets can be attributed to two factors: 1) NH_3 concentrations are higher and therefore the signal measured by the two instruments are larger leading to a better correlation from the wide range of NH_3 concentrations ($0\text{--}18\ \mu\text{g}\cdot\text{m}^{-3}$ for the miniDOAS and $0\text{--}1\cdot 10^{16}$ molecules. cm^{-2} for IASI, Figure 1) and 2) the high amount of NH_3 emitted in spring in the surrounding regions due to fertilizer applications can be transported to Paris [Viatte et al., 2022; Viatte et al., 2021] resulting in high correlations between the $\sim 12\text{-km}$ IASI footprints and the local miniDOAS observations.”

“In fall and summer, the lower correlations between the ground-based and the satellite NH_3 observations could reveal specific NH_3 sources in the close vicinity of the miniDOAS which might be not representative of the IASI pixels size.”

p.7 l.173: The satellite retrievals are strongly impacted by thermal contrast. It is highly surprising and difficult to believe that there is no impact of temperature on the correlation as it affects the sensitivity of the satellite data. On the other hand near-surface measurements are highly impacted by PBL height, while this is less the case for the column observations. Please explain.

We agree that figure S1 is not convincing and contradicts the statement that the miniDOAS observations are sensitive to the PBL height, while IASI are not, as they measure columns. Thus we have removed this statement and this figure in the revised supplementary information and manuscript documents.

p.7 l.177: If I understand it correctly, in the further analysis you average the IASI retrievals within a box of 50 km e.g. to compare with miniDOAS? In that case you cannot speak about a city-scale study neither about studying the spatiotemporal variability in Paris. Temporal yes, but not spatial!

Indeed, we have average IASI pixels within a 50km box around Paris taking into account the real elliptic size of each pixel as describe in Van Damme et al. (2018). The center of IASI pixels determines if it falls within the averaging box. We have modified “city-scale” to “Greater Paris-scale”, “spatiotemporal” to “temporal”; and added this sentence to the revised manuscript: “we have selected coincident observations made within the same hour and the center of the IASI pixels was used to determine the distance between the miniDOAS and IASI measurements”.

p.7 l.186: Please describe here or provide a reference how the error budget is done.

The uncertainties of the measurements are calculated following the literature recommendation, referenced in Volten et al., [2012]. We have added this reference in the manuscript “Description of the measurement uncertainties can be found in Volten et al. (2012).”

p.8 l.199: Agreed, but vertically you are also at large distance from the potential traffic emission source that you want to assess in this study, when not putting it at the surface. Official monitoring stations usually sample at 3 m altitude. This might also explain why you measure lower concentrations than in other urban areas / other studies.

We agree with the referee that at 40m in altitude, measurements are not affected by local traffic variability, and this could explain why we have lower concentrations that in other urban areas. The main goal of this study is to assess the NH₃ budget over the Greater Paris area, which is why we installed the instrument at the urban canopy so that the measurements footprint is representative of the Parisian urban area.

We have added a sentence in the revised manuscript to clarify: “The miniDOAS is located at an altitude of 40m so that its observation footprint is representative of the Greater Paris. This may partly explain the lower NH₃ concentrations observed in Paris compared to other urban areas.”

p.15 l. 376: If the data from this traffic station (and maybe others) is available, why isn’t it used in this study, even if it isn’t a miniDOAS? It would make your results far more convincing, e.g. to check if the results are consistent for different stations and to assess the effect of having the miniDOAS at 40 m.

To our knowledge, there are no available NH₃ measurements in Paris close to traffic, and there are no other relatively long-term and high temporal resolution NH₃ observations in Paris at all. The reference we cited in this study (e. g., [Chatain et al., 2022]) is for another city of France (Reims). We clarified this in the revised manuscript. We would, indeed, be delighted to compare our NH₃ budget at 40m to the one close to the ground but unfortunately it is impossible for now.

Chatain, M., Chretien, E., Crunaire, S. and Jantzen, E.: Road Traffic and Its Influence on Urban Ammonia Concentrations (France), *Atmosphere (Basel)*, 13(7), doi:10.3390/atmos13071032, 2022.

Van Damme, M.; Clarisse, L.; Whitburn, S.; Hadji-Lazaro, J.; Hurtmans, D.; Clerbaux, C.; Coheur, P.-F. Industrial and agricultural ammonia point sources exposed. *Nature* 2018, 564, 99-103, doi:10.1038/s41586-018-0747-1.

Volten, H., Bergwerff, J. B., Haaima, M., Lolkema, D. E., Berkhout, A. J. C., van der Hoff, G. R., Potma, C. J. M., Wichink Kruit, R. J., van Pul, W. A. J. and Swart, D. P. J.: Two instruments based on differential optical absorption spectroscopy (DOAS) to measure accurate ammonia concentrations in the atmosphere, *Atmos. Meas. Tech.*, 5(2), 413–427, doi:10.5194/amt-5-413-2012, 2012.

Minor comments

The minor revisions have all been considered in the revised manuscript.

p.4 l.88: Would be good to include as well <https://amt.copernicus.org/articles/9/2721/2016/amt-9-2721-2016.pdf>

p.8 l.202: I assume you provide first averages and after min max ranges? Please specify the numbers that are provided here.

We have specified that the first number is the average and the second one is the standard deviation.

p.8 l.212: Please elaborate shortly on these high pollution events.

We have modified the sentence: "IASI NH₃ total columns around Paris exhibit a higher NH₃ annual concentration and standard deviation in 2020 compared to the other years because of the multiple high pollution events occurring in spring during the 2020-COVID lockdown as described in Viatte et al. (2021)."

p.10 l.250: It is not really clear why this is then only the case in 2021, and for example not seen in 2020?

Interannual variability of NH₃ concentrations over Paris depends, in the first order, on agricultural activities and meteorological conditions. In September 2020, atmospheric temperatures were lower than in 2021 (see appendix S2) which could explain why the miniDOAS and IASI haven't monitor NH₃ enhancements. We have added this explanation to the revised manuscript: "In 2021, a second NH₃ enhancement is measured in September by the miniDOAS ($2.73 \pm 1.14 \mu\text{g}\cdot\text{m}^{-3}$) and IASI ($7.93 \times 10^{15} \pm 4.64 \times 10^{15} \text{ molecules}\cdot\text{cm}^{-2}$) which is not observed in 2020 possibly because atmospheric temperatures were lower than in 2021 (Figure S1)"

p.12 l.296: Please elaborate on why this is observed in agricultural areas. Is it related to reduced agricultural activity over the weekend?

Indeed, we have clarified this in the revised manuscript: "The weekly cycle of IASI measurements in Paris is almost analogous to the one observed over European agricultural areas with low concentrations observed on Mondays, as a result of reduced NH₃ emissions over the weekend, and an accumulation of ammonia during the week [Van Damme et al., 2022]."

Technical corrections

All technical corrections have been applied to the revised document.

p.1 l.20: in other urban areas

p.3 l.62: replace 'monitored' by 'detected'?

We replaced "monitored" by "observed".

p.7 l.156: do you mean quantitatively?

We have deleted this word in the revised manuscript.

p.11 l.278: 'raw' should be 'row'

p.15 l.380: I suggest replacing 'Atmospheric' by 'Temporal'