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

Referee: This paper analyzes NH_3 measurements from the IASI instrument on the Metop-B and Metop-C platforms and from a miniDOAS instrument located in the center of Paris. The two datasets are shown to be correlated overall and also on seasonal and weekly time scales. These analyses suggest that traffic is the very significant source of NH3 in Paris, except in the spring. A source contribution approach (PSCF) uses the miniDOAS data and ERA-5 meteorological fields to demonstrate that transport from agricultural regions to the northeast, east and southeast dominates in the spring. The contribution from traffic is further confirmed by comparisons with weekly and daily traffic and NO_2 data.

This is a clearly organized and well written paper that adds to the growing body of work on NH_3 sources and variability in large urban areas. It requires only minor revisions to be acceptable for publication.

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.

Technical comments

1. The authors need to make clear in the section 2.1 that there is only one miniDOAS instrument and provide its location. This becomes apparent later in the paper, but it would make the earlier sections clearer if stated up front.

We have clarified this by adding these following sentences at the beginning of section 2.1 : “NH_3 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) NH_3 observations at high temporal frequency representative of the Paris megacity.”

2. On line 62 comment on why there are pollution episodes every spring in Paris.

Several publications have shown that particulate matter pollution episodes in springtime are found to be almost annually frequent in Paris and often associated with emissions from agricultural activities in the areas surrounding the agglomerations. We have listed other significant papers and added this statement to the revised manuscript: “Monitoring NH_3 is therefore essential, especially in urban areas such as in Paris, where particulate pollution episodes are monitored almost every spring [Viatte et al., 2022] and often associated with emissions from agricultural activities in the surrounding areas [Viatte et al., 2021; Kutzner et al., 2021; Viatte et al., 2020; Petetin et al., 2016; Petit et al., 2015].”

3. Is the center of the IASI pixel used to determine if it falls within the averaging box? If yes, please state this.

Indeed, the center of IASI pixels determines if it falls within the averaging box. We have clarified this in section 2.2 of 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”.

4. While the authors do discuss the trade-off between analysis box size and number of IASI samples, to justify their choice of a 50 km window, given the high spatial variability of NH_3 it is certain that the air masses sampled by IASI and the miniDOAS are very different. Could the high correlations in spring between the two datasets be attributed to the fact that the main sources of NH3 are outside Paris? Do the authors think that the lower correlations in spring and fall could be in part due to urban sources playing a larger role, which may drive greater variability within the 50 km box? Any further comments on the spatial variability issue?

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 (0-18 $\mu g.m^{-3}$ for the miniDOAS and 0-1.10^{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 ~12-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."

5. On line 173 the authors state that correlations between the IASI and miniDOAS observations are independent of atmospheric temperature and PBL height, and point to Figure S1. This figure is not very convincing. And it contradicts the statement in section 3.3.2 (and in the conclusions) that the miniDOAS observations are sensitive to the PBL height, as they measure a local concentration, while the FTIR (and IASI) are not, as they measure columns. Thus it is difficult to believe that the correlation does not depend on PBL height. I suggest either removing the statement, or clarifying it by calculating and showing the correlations within temperature and PBL bins.

We agree that figure S1 is not convincing, thus we have removed this statement and this figure in the revised supplementary information and manuscript documents.

6. The PSCF is shown for all data points and for spring. I suggest showing this function for summer and fall also. If it shows that the NH_3 sources in these seasons are within the urban area, it would provide an additional argument to the importance of urban sources.

The goal of the PSCF analysis is to attribute the main geographical sources of NH_3 emissions that affect the urban air quality of Paris. As mentioned previously, it is known that agricultural practices are enhanced in spring over our region of study. This is why we have performed PSCF analysis in spring for three different years (2020, 2021, and 2022) to study the interannual variability of the major NH_3 source regions that affect the NH_3 budget in Paris.

As the referee's suggested, we have also performed the PSCF analysis per seasons, as shown in Figure R1 of this document. We clearly see that the PSCF shows an extended area of the regional contribution in spring compared to the other seasons. In addition, NH_3 concentrations and variability in Paris in spring are at least two times higher than for the other seasons, proving the importance of the surrounding agriculture practices in the high NH_3 budget in Paris. However, even if we see that PSCF are significantly reduced around Paris during fall, winter, and summer compared to spring, we cannot conclude from this figure that the urban area is the unique contribution of NH_3 budget in Paris. One reason might be that we have used 6-hours back-trajectories to compute the PSCF analysis for the whole dataset. This value was chosen based on the large range of NH_3 lifetimes found in the literature, which are from 2 hours to few days [Hertel, et al., 2012; Behera et al., 2013; Haugustaine et al., 2014; Whitburn et al., 2016; Van Damme et al., 2018; Dammers et al., 2019; Evangeliou et al., 2021; Luo et al., 2022]. Since NH_3 lifetime depends on atmospheric chemical loss and wet deposition (which are linked to atmospheric temperature and humidity), it will thus be different depending on the season. Choosing a constant value of NH_3 lifetime throughout the year will bias the length of the back-trajectories depending on the seasons.

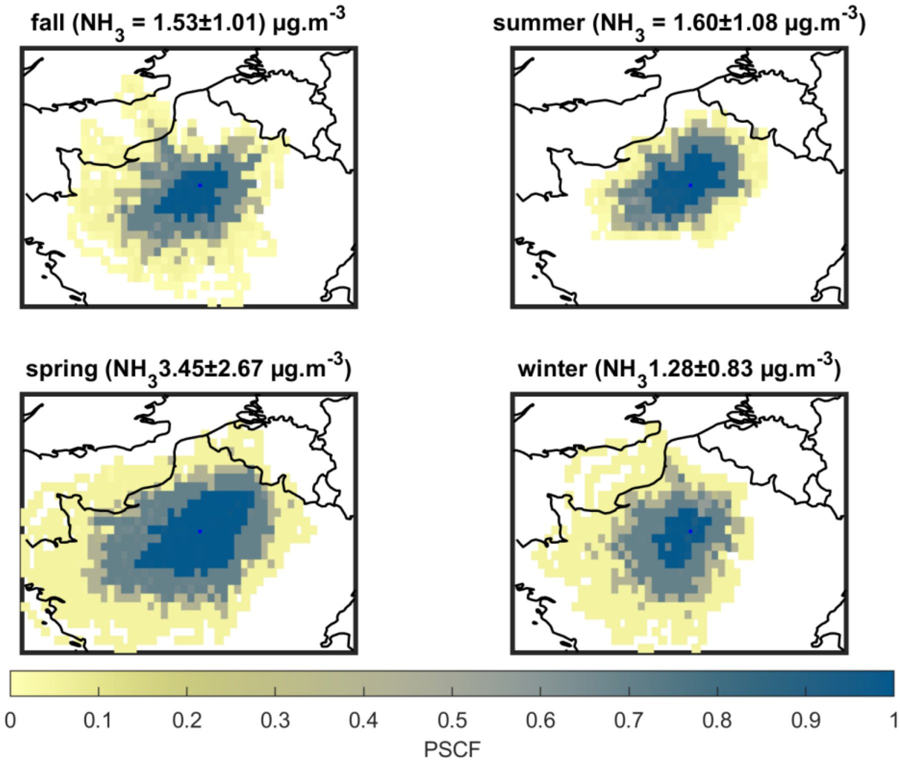

Figure R1: Potential Source Contribution Function (PSCF) per season. The blue dot indicates the location of Paris.

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Behera, S.N.; Sharma, M.; Aneja, V.P.; Balasubramanian, R. Ammonia in the atmosphere: a review on emission sources, atmospheric chemistry and deposition on terrestrial bodies. Environ. Sci. Pollut. Res. 2013, 20, 8092-8131, doi:10.1007/s11356-013- 2051-9.

Hauglustaine, D.A.; Balkanski, Y.; Schulz, M. A global model simulation of present and future nitrate aerosols and their direct radiative forcing of climate. Atmos. Chem. Phys. 2014, 14, 11031-11063, doi:10.5194/acp-14-11031-2014.

Whitburn, S.; Van Damme, M.; Clarisse, L.; Turquety, S.; Clerbaux, C.; Coheur, P.F. Doubling of annual ammonia emissions from the peat fires in Indonesia during the 2015 El Niño. Geophys. Res. Lett. 2016, 43, 11,007-011,014, doi:10.1002/2016gl070620.

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.

Dammers, E.; McLinden, C.A.; Griffin, D.; Shephard, M.W.; Van der Graaf, S.; Lutsch, E.; Schaap, M.; Gainairu-Matz, Y.; Fioletov, V.; Van Damme, M., et al. NH3 emissions from large point sources derived from CrIS and IASI satellite observations. Atmos. Chem. Phys. 2019, 19, 12261-12293, doi:10.5194/acp-19-12261-2019.

Evangeliou, N.; Balkanski, Y.; Eckhardt, S.; Cozic, A.; Van Damme, M.; Coheur, P.-F.; Clarisse, L.; Shephard, M.W.; Cady-Pereira, K.E.; Hauglustaine, D. 10-year satellite-constrained fluxes of ammonia improve performance of chemistry transport models. Atmos. Chem. Phys. 2021, 21, 4431-4451, doi:10.5194/acp-21-4431-2021.

Luo, Z.; Zhang, Y.; Chen, W.; Van Damme, M.; Coheur, P.-F.; Clarisse, L. Estimating global ammonia (NH3) emissions based on IASI observations from 2008 to 2018. Atmos. Chem. Phys. 2022, 22, 10375-10388, doi:10.5194/acp-22-10375-2022.

Minor revisions

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

Line 1: Title is odd: I would remove the "measurement report" phrase.

We agree but it is mandatory to publish in the ACP measurement report journal: "The title must clearly reflect the manuscript type and start with "Measurement report:" (https://www.atmospheric-chemistry-and-physics.net/about/manuscript_types.html)"

Line 36: …which plays a role in …

Line 62: … are observed almost every spring …

Line 69: … their variability …

Line 99: … day and night, and does not suffer from sampling artifacts, since it does not use a filter or inlet, unlike other commonly used instruments (also provide examples of instruments, not just references)

Line 101: Using ammonia measurements obtained from the miniDOAS at the QUALAIR supersite (40 meters above ground level, https://qualair.fr/index.php/en/english/) in the Paris citycenter, Viatte et al. (2021) demonstrated the contribution of NH3 to particulate pollution events that occurred ….

Line 117: … 2020], which is built from observations recalibrated into a global assimilation model at a 30 km resolution.

Line 132: Using the hourly …

Line 145: we used the oversampling method described by van Damme et al. (2018), which takes into account the real elliptical sizes of each IASI pixel… Are the values in this map ever used in the analysis? Please clarify.

To clarify, we have added this sentence to the revised manuscript: "All IASI maps shown in this study were computed using this methodology".

Line 212: … because of the multiple high pollution events …

Line 230: city center. Note that …

Line 258: …, in the spring of 2020, 2021 …

Line 278: … top row …

Line 279: Make blue dot larger

Line 317: suggestion for Figure 5: always use the darker color for miniDOAS

Line 380: …assessed using joint observations …

Line 390: …due to fertilizer spreading …

Line 393: The PSCF analyses indicate that the agricultural regions to the east and northeast within 100 to 200 km from Paris city-center have the greatest impact on the NH3 budget in Paris.

Figure S3: cite source for these data.