Review 2 of “NH3 Spatio-temporal variability over Paris, Mexico and Toronto and its link to PM2.5 during pollution events”

Referee :

Summary

This paper analyses 10 years of IASI NH3 data over three large domains, each of which encompasses a major metropolitan area (Paris, Toronto and Mexico City). The ten year average and seasonal means of the NH3 total columns from the IASI instruments deployed on Metop-A and Metop-B are presented and the relationships between NH3 amounts and temperature and precipitation are evaluated; the authors find a strong correlation between temperature and NH3 amounts in the Paris and Toronto domains, but only a weak one in the Mexico City domain. This analysis is extended to sub-domain scales, focusing on a number of previously identified source regions, and expanded to include the effect of relative humidity. Many of the smaller domains show an interesting local maxima in the NH3 vs temperature plot that coincides with fertilization activities. The impact of wind direction on air quality is also examined, using back-trajectories from HYSPLIT and a cluster analysis, along with in situ PM2.5 data from local networks. The PM 2.5 data is also used in conjunction with the NH3 columns to identify and count pollution events; wind roses are then constructed to determine the wind patterns on days with high pollution; interestingly PM2.5 and NH 3 are not always high on the same days. Finally IASI NH3 and the local PM2.5 data are compared against GEOS-Chem output for one month. This last analysis is interesting but too limited to provide really useful information on GEOS-Chem performance. The paper is well laid out and clearly written. The plots are of high quality and in general easily understood, though a few require more detailed captions, described below. It requires only some minor edits and clarifications to be acceptable for publication. Overall, a good illustration of how to apply a number of different techniques and data sources to the problem of understanding the drivers of pollution events.

Authors: We would like to thank the referee for this positive review and all the relevant comments that we have addressed in the following document.

Technical comments

Section 3.1: How are the IASI averaged and what is the grid resolution? How are the boxes doe each source region defined?

We have gridded the IASI data at 0.25° x 0.25° degrees. This information has been included in the revised manuscript. The sources regions are defined by looking at local enhancement in the 10-year average of the IASI total column.

Figure 3: Please comment on the high NH_3 values over the Arctic.

The sensitivity of IASI measurements is intimately related to the thermal contrast between the surface and the first layers of the atmosphere [Clerbaux et al., 2009]. When the detection is possible (with good thermal contrast), the peak sensitivity for NH_3 is in the boundary layer [Clarisse et al., 2010]. The high value over Canada and the Arctic in winter can be associated with high uncertainties in the NH_3

retrievals due to low thermal contrast and high emissivity from snow. We have added in the revised manuscript this sentence to clarify: «The high value over Canada and the Arctic in winter can be associated with high uncertainties in the NH_3 retrievals due to low thermal contrast».

Lines 266-274: Is each back-trajectory associated with the 50 km NH3 mean for that day? Could the authors please briefly describe the clustering approach? Are the NH3 means clustered according to the corresponding back-trajectories?

We have explained the method following the different steps (Figure R1) and added these descriptions in the revised supplementary information Figure S7.

1) For each day, we have run HYSPLIT back-trajectories ending in the cities at the overpass time of the IASI satellite (blue lines in Figure R1).

2) For each day, we have calculated the amount of NH_3 derived from IASI observations within a circle of 50km radius around the cities (orange cylinder in Figure R1).

3) We have run the cluster analysis to merge trajectories that are near each other (green lines in Figure R1). The cluster analysis computes the spatial variance and minimize differences between trajectories within a cluster while differences between clusters are maximized [Abdalmogith et al., 2005; https://www.ready.noaa.gov/documents/Tutorial/html/traj_cluseqn.html]. NH_3 mean concentrations measured inside the cities by IASI have been allocated to the different mean cluster trajectories according to the corresponding back-trajectories.

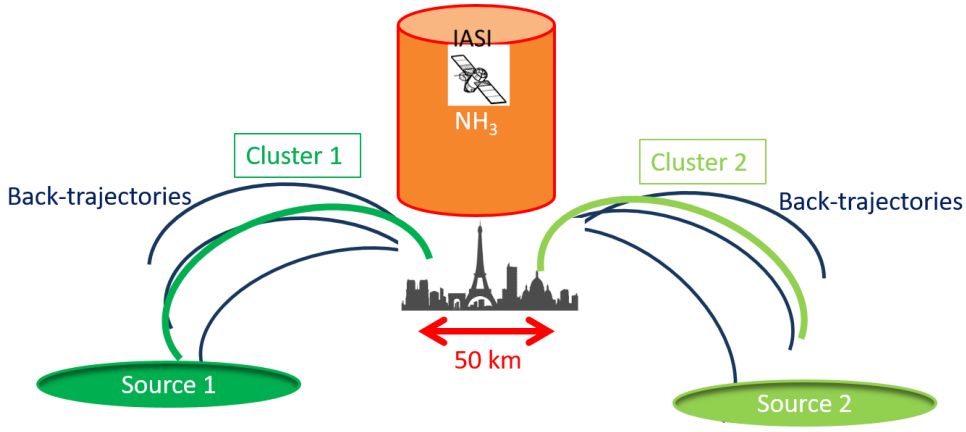

Figure R1: method to analyze the impact of long-range transport on NH_3 concentrations measured over the cities

Line 289: Which sources have increased near Toronto?

According Yao and Zhang (2019), NH_3 concentrations around Toronto (near Granby – box A in Figure 2 and Elmira-Kitchener-Guelp – box B in Figure 2) have increased in the last decade. We have modified the sentence in the revised manuscript to clarify this point: "9 to 17% of NH_3 concentrations are coming from the west and the east of Toronto (cluster 3, 4, and 5) where atmospheric NH_3 have increased in the last decade (Boxes A and B in Figure 2, [Yao and Zhang, 2019])"

Line 303: What are the criteria used for defining a pollution event?

We have used the same method than the one conducted in Yamanouchi et al. (2021). Pollution events are defined when residuals of the Fourier fit are above 2 standard deviations. We have inserted "2 standard deviations" in the sentence of the revised manuscript.

Line 313: It would be useful to add the spring plot here

We have added the plot showing the seasonal occurrence of the pollution events in the revised manuscript.

Line 319: What does the radial distance in the wind roses in Figure 7 indicate?

The radial distance in the wind roses indicates the frequency of the wind direction occurrence. We have added this information in the revised manuscript.

Line 350: Please state the coincidence criteria.

The criteria were stated in the following sentence. To clarify, we have arranged this sentence: "Spatial and temporal coincidence criteria have been applied to GEOS-Chem outputs to compare with IASI morning observations, such as: model outputs between 8.30 and 11.30 AM coincident with IASI overpasses have been selected, and only collocated model outputs (at 0.5° × 0.625° horizontal resolution) have been selected coincident with IASI observations."

Lines 360-364: I don't agree that the GEOS-Chem and IASI are in good agreement; the authors should state that the two datasets capture some of the same pollution events.

We have modified the text accordingly.

Figure S4: Are the bins for the RH plot also specified by temperature? This does not seem right.

In Figure S4, lower panel, the NH_3 values are averaged per bins of relative humidity with a 1% RH between each consecutive bin. We modified the description of Figure S4 in order to make this clearer.

Minor edits

Authors: All minor edits have been addressed in the revised manuscript.

Lines 32-33: ... in Paris and Mexico City;

Line 65: However, NH3 concentrations are increasing in many countries: France, Canada and

Mexico reported increases of .....

Line 67: ... are composed of ...

Line 72: ...nitrate formed from ...

Line 127: ... are located within a 50-km

Line 134: ... the same month

Line 150: Note that

Line 153: ... daily 24-hour back-trajectories ...

Line 156: Finally, all back-trajectories are combined .... (not sure here what the authors mean to say)

We have modified this sentence as: "Finally, every back-trajectory that are near to each other are merged in clusters and associated with the corresponding local-scale IASI NH_3 concentrations".

Line 182: ... practices (which are dominant over Europe and North America)

Lines205: ... in this region closer to the Equator

Line 220: ...(with small contributions from industries)

Line 255: ...nitrogen fertilizers to

Line 267: concentrations. In order to analyze ...

Line 280: ... associated with the highest ...

Line 281: ...on average, are originating ...

Line 322: ... concentrations are observed

Line 352: ...numbers of coincident observations

Line 403: ...sources are

Line 440: ... surrounding regions:

Line 441: ...These lead

Line 444: ... and Mexico pollution is transported along the northeast-southwest line,

Line 458: ... launched in

References:

Abdalmogith, S. S. and Harrison, R. M.: The use of trajectory cluster analysis to examine the long-range transport of secondary inorganic aerosol in the UK, Atmos. Environ., 39(35), 6686–6695, doi:https://doi.org/10.1016/j.atmosenv.2005.07.059, 2005.

Clarisse, L., Shephard, M. W., Dentener, F., Hurtmans, D., Cady-Pereira, K., Karagulian, F., Van Damme, M., Clerbaux, C. and Coheur, P.-F.: Satellite monitoring of ammonia: A case study of the San Joaquin Valley, J. Geophys. Res. Atmos., 115(D13), doi:https://doi.org/10.1029/2009JD013291, 2010.

Clerbaux, C., Boynard, A., Clarisse, L., George, M., Hadji-Lazaro, J., Herbin, H., Hurtmans, D., Pommier, M., Razavi, A., Turquety, S., Wespes, C. and Coheur, P.-F.: Monitoring of atmospheric composition using the thermal infrared IASI/MetOp sounder, Atmos. Chem. Phys., 9(16), 6041–6054, doi:10.5194/acp-9-6041-2009, 2009.

Yamanouchi, S., Viatte, C., Strong, K., Lutsch, E., Jones, D. B. A., Clerbaux, C., Van Damme, M., Clarisse, L. and Coheur, P.-F.: Multiscale observations of NH3 around Toronto, Canada, Atmos. Meas. Tech., 14(2), doi:10.5194/amt-14-905-2021, 2021.