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Atmospheric Chemistry and Physics Discussions

Interactive comment on "10–year satellite–constrained fluxes of ammonia improve performance of chemistry transport models" *by* Nikolaos Evangeliou et al.

Anonymous Referee #1

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This manuscript provides a description of an inverse method based on the NH3 lifetime to estimate NH3 global emissions from the satellite IASI observations over the 10 yr-period 2008-2017. As NH3 is a key species for understanding the PM levels, the quantification of its global emissions is important and would be useful to a wide community. The authors cover an important topic, appropriate for ACP. Nevertheless, I have some major comments listed below that should be considered by the authors before publication.

Major comments:

1/ The fact that NH3 columns in the atmosphere depend not only on NH3 emissions,

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but is also linked to the abundance of nitric and sulfuric acids (and consequently to NOx and SO2 emissions) is not fully described. To tackle the large variability of the ammonia lifetime, the authors calculated the NH3 lifetime with a CTM and the spatial variability of ammonia is taken into account. I have more doubt about the temporal variability of ammonia and its main drivers in the atmosphere. If I well understand, the variable lifetime chosen for this study is a gridded average over the 10-yr period. If it is correct, the temporal trend in nitric and sulfuric acids is not fully taken into account, while it could have an importance for the deduced NH3 emissions over a 10-yr period. This choice should be explained in the text. Would it possible to calculate yearly lifetimes as a sensitivity test to assess the robustness of your study?

2/ A comprehensive overview about the existing literature is missing. For example, result for SO2 changes in Figure S2 is not in agreement with Krotkov et al., 2016, ACP, showing strong decrease of SO2 between 2005 and 2015 at least over Eastern US and over Eastern Europe. Also, different publications have shown NH3 peak in spring over northwestern European countries, not seen here. At least, discrepancies with previous studies should be discussed. These features could be explained by the choice of the authors to analyze their results for Europe or for the US as a whole. An analysis done for the hot-spot regions, of interest, where the emissions are high in Figure 4 may help the analysis.

3/ The impact of the abundance of sulfuric acid on NH3 columns is detailed, but not the impact of the abundance of nitric acid. Is this impact considered negligible compared to those of sulfuric acids? This should be discussed. The same Figure S2 for NO2 columns and nitrate concentrations may help analyzing the results.

Specific comments:

line 87: a comma is missing before "the Tropospheric Emission Spectrometer"

line 90-95: a verb is missing in this sentence

line 96-97: Note that Kuenen and Dore, [2019] estimated the uncertainties linked to the agricultural sector at about 100-300% at the European and annual scale. https://www.eea.europa.eu/publications/emep-eea-guidebook-2019/part-a-general-guidance-chapters/5-uncertainties/view

line 98-102 : What is the differences between the different IASI products? The terms NE, VD0.5 and VDgrlf are not intuitive and are not explained at this stage.

line 105: please add references of studies using this state-of-the-art inventory.

Line 124: could a difference of 2%±24% just due to the use of particular vertical profiles be interpreted as “small uncertainties”?

Line 126-151: the description for CrIS gives more information than for IASI. The analysis of the results may be facilitating with the same information for both the instruments. I encourage you to give more information for IASI (total column uncertainties, peak sensitivity, detection limit, etc)

Line 152, Section 2.2: could you please provide a map of the interpolated IASI observations? As you performed simulations, it would be great to see the comparison between IASI and the CTM.

Line 155: What is the CTM? As the variable lifetime in section 2.3 is based on this CTM, it should be described before. I would have described LMDZ-OR-INCA before section 2.3.

Line 160: I would refer to IASI ammonia total columns.

Line 188: Please precise the regions where nitric and sulfuric acids are abundant in the text or at least, refer to Figure 2c and to Figure 2d.

Line 211: Is the variable lifetime from a CTM for the quantification of VDgrlf emissions similar to the one for the quantification of NE emissions? This is not clear.

Line 227-239: Has the NH3 deposition of LMDz-OR-INCA been already evaluated? Is

the bi-directional exchange with surfaces taken into account? This is not discussed. If not, how does it impact your NH3 emissions?

Line 253: you do not focus on hotspot regions but on continents as a whole.

Line 256-271: the different lifetimes of the literature and your results could be highlighted in a Table. Line 276: As Ammonia lifetime depends on the presence of ammonia's reactants (sulfuric and nitric acid), it also depends on NOx and SO2 emissions, not only NH3 emissions. I would have written "(sulfuric and nitric acids, through SO2 and NOx emissions)".

Figure 1: space is missing between the legend and Figure 1c and 1d

Line 287: "which is in the range of the previously reported values". Your results are far from the results from Dammers et al [2019] for example. How do you explain such differences? Could the simulated NH3 lifetime by CTM be over-estimated?

Line 296: Please note in the legend of Figure 1b that the average ammonia emissions are calculated from the 10-year IASI observations and precise with which lifetime. I first thought it was the average ammonia emissions from ECLIPSEv5-GFED4-GEIA. Please also verify the legend of Figure S3.

Line 320-321: The sentence "Although column concentrations of both sulfur dioxide and sulfates present strong interannual variability, they do not show significant changes on an annual basis" is not clear. Please rephrase.

Line 331: I do not understand why the anomaly is calculated only after 2015. Please explain.

Line 334-337: why the NH3 emissions based on IASI observations could be impacted by changes in SO2 and NOx emissions only after 2015? In Lachatre et al., 2019, the study you are citing line 337, the changes in SO2 at least are seen before 2015. This is also the case in your Figure S2. Please strengthen this discussion.

Line 352: please deeply detail why the fact that northern India has been previously identified as a hot-spot region for ammonia explains the differences between the emission datasets.

Line 335: Please verify the species indices

Line 356: the ammonia emissions remain mostly constant at the global scale. Is it still true at continental scale?

Line 357: “The total calculated ammonia emissions": which one?

Line 360-363: could you please provide statistics (average and standard deviation) for South American and European emissions as well as for the global budget?

Line 363-364: “Based upon IASI retrievals, Liu et al. (2019) showed an increase of surface NH3 concentrations trend of more than 0.2μgNm-3yr-1”: I do not understand the link with the previous sentence.

Line 365: “Ammonia emissions derived over China in this work are among the highest worldwide (Figure S1)”: is this already the case in the EDGAR and EGG bottom-up inventories or is this a new feature?

Line 370: please precise “The comparison of the annual ammonia NE emissions. . .” In general, you should specify the inventory or the sensitivity test you are referring to, it would help for the reading and for the understanding of the study.

Line 377: I would add “in these regions” at the end of the sentence. Indeed, the impact of the different lifetimes seems to be slight over the other regions of the world.

Line 385-386: is this contradictory with the sentence “European emissions are practically identical in all datasets” in line 361?

Line 460: consist in?

Line 461-470: The description of the different inventories and of the different performed

simulations should occur before in the text. I would have placed this paragraph at the end of the introduction.

Figure 4: you should number the different graphs. It would be easier to reference them in the text. Please better describe the NH3 emission dataset in the legend.

Line 532: there is an empty bracket.

Section 4.2: Does the evaluation against CrIS done at the global scale? It is not specified. If it is the case, it is not comparable with the surface evaluation done at the regional scale. It would be very interested to do it also at regional scale for the analysis, as in Figure 5, 6 and 7 and particularly over hot-spots as explained in the major comments.

Line 599: the word “already” is misplaced in the sentence.

Figure 9: the colors of the scale should be changed: when the uncertainty is high, the borders on the map are not clearly visible.

Line 612: what are the regions with “changing balance between nitrate and sulfate abundances”? Please detail in the text.

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