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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.

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- Overall, the paper is well written and provides new information to the literature on global NH3, which has not been well characterized previously. The paper is rather long and could condense it down to a tighter paper that is more focused on key results and conclusions.

Response: We appreciate reviewer's comments and his willingness to improve this manuscript. We have made all the changes requested by the 2 reviewers and we are willing to further work to shorten the manuscript, if additional detailed comments are to be requested.

- Comparing model predictions at the coarse level presented here (2.5 degrees or 250 km) to ground monitors and discussing "hotspots" may not well represent the spatially variable nature of NH3 emissions. Averages over these large cells could misrepresent key features of NH3 distributions. However, the spatial resolution in Figure 4 appears to be finer than 2.5 x 1.3 degree. Was a particular plotting technique used to show the NH3 levels that might be making gradient interpolations or is the data in Figure 4 actually 2.5 x 1.3 degree resolution?

Response: We acknowledge reviewer's observation here and we admit this was misleading. We have now clarified in section 2.3 (see Track Changes at p.7-8). What we have done was to process the IASI column concentration measurements onto a grid of $0.5^{\circ}\times0.5^{\circ}$ using the IDW method that we describe in section 2.3. Then, since the resolution of the CTM model that we used bilinear interpolation classic method to convert to the model resolution ($2.5^{\circ}\times1.3^{\circ}$).

- The authors estimated emission fluxes using a lifetime parameter from the CTM. Was there some reason a traditional assimilation approach (e.g., like Alvarado is doing) was not included in this assessment? The authors should consider a comparison of the column predictions of the CTM simulation using the estimated emissions back to the IASI measurements. If the lifetime approach is accurate, the CTM should accurately predict the IASI columns when using the scaled emissions. If this was done it is not clear from the text. Further, it did not seem like the seasonal NH3 lifetime estimated by the CTM provided a substantively different result than the 0.5VD constant assumption.

Response: There was not special reason for not using a classic assimilation method here. Our idea was to try to calculate emissions from IASI column NH3 measurements. For this, we needed a metric of the lifetime of NH3. We used a constant lifetime of 0.5 d everywhere, as well as a gridded one, calculated from a model, which we thought it is more realistic, as ammonia cannot have the same lifetime everywhere (see section 3.1). Finally, we wanted to see if the calculated emissions have a significant impact on surface concentrations. For this, we compare with measurements from EMEP, EANET

and AMoN. The goal of this paper is not to validate the CTM against IASI column ammonia. However, an example of how the column NH3 in the model compares to IASI column NH3 is now given in Supplementary Figure S11 (of the manuscript). The model is continuously validated by the LSCE group (see relevant papers here: https://www.lsce.ipsl.fr/en/Phocea/Vie_des_labos/Ast/ast_groupe.php?id_groupe=94&voir=p
We rather want to prove that for very short-lived species such as NH3, a simple approach like the one we describe in section 2 is enough to constrain the main source of NH3 in the atmosphere, a chemical species that is difficult to be quantified with classic inverse modelling approaches, due to its heterogeneous chemistry. The goals of this manuscript are explained in detail in the last paragraph of the introduction.

The reviewer states that ", it did not seem like the seasonal NH3 lifetime estimated by the CTM provided a substantively different result than the 0.5VD constant assumption". We only show the lifetime calculated by the CTM in Figure 1d (of the manuscript), which basically shows values between 10 – 12.5 hours, whereas in VD0.5 a constant lifetime of 12 hours (0.5 d) was used everywhere. The difference in the emissions using a variable versus a constant lifetime for NH3 are shown in Figure 3 (of the manuscript) and they are as high as 29.4 Tg/y (on average), or 15% different, which we do think it is substantially different; both in absolute numbers, but also in the spatial distribution of the emissions. The impact on the surface concentrations against observations is shown in both as time-series plots in the Supplements and as scatterplots in the main text. The IASI-constrained emissions, at least in the North America and Southeastern Asia, capture realistically atmospheric concentrations (see linear scale in x- and y-axes).

- Ammonia has a strong diurnal profile. Does the assumption for diel profile impact any of the results presented in this paper or does the diurnal nature of NH3 emissions have no impact on these products?

Response: Indeed, NH3 has a strong diurnal cycle, and the CTM uses a means to account for a diurnal cycle. However, we have not assessed how the diurnal cycle

in the present setup affects the results. The reason is that, although IASI NH3 are measured twice a day, only morning measurements were used in the present study, due to the larger thermal conditions that lead to smaller uncertainties. Accordingly, we have used daily model outputs for concentrations and monthly mean lifetimes from the model. In addition, all measurements used here to evaluate modelled concentrations have a temporal resolution of 1 or 2 weeks. Therefore, no further effort to deal with the diurnal cycle of NH3 was made and rather assumed that it should not affect much our results. Of course, we have to admit that a bias in the overall assessment could be realistic, although no data to prove this were available.

- When taking a closer look at Figure 6, is it surprising that VD0.5, NE, and VDgrid emisisons used in a model result in very few model estimations of ammonia below 0.5 and ECG rarely has a prediction above 0.5. Some of the calculated performance metrics may suggest "good" model performance but the shape of the model-observed NH3 in Figure 6 shows some features that suggest they many of these approaches can not replicate the range of NH3 levels measured.

Response: We rather think this is normal. As one can read in section 3.4, "North American annual ammonia emissions over the 10-year period were averaged 1.1+-0.1 Tg yr-1 (average+-sd). These values are over two orders of magnitude higher than those in EGG (0.062+-0.0013 Tg yr-1). Note that his estimate is three times lower than those reported in VD0.5 (3.1 Tg yr-1) or in VDgrlf (3.4+-0.5 Tg yr-1)." Therefore, we see smaller MFB values (=0.32) in Figure 6 (of the manuscript) than those of VD0.5 (=0.52) and VDgrlf (=0.54) and much higher than those in EGG (=-0.28). Another view of the modelled-observation mismatches can be seen in supplementary Figures S7-S9.

- Please provide some more clarity on the vertical profile used for NH3 for IASI retrievals. Is this constant and not variable with changes in altitude? Does the vertical profile conform to profiles measured as part of aircraft measurement campaigns and seem realistic?

Response: We have not used any vertical profile for IASI NH3. As we explain in detail in p.7-L.316 “IASI total column ammonia measurements were interpolated onto . . .” a grid with the method described in section 2.3. Then, a box-model was used to calculate gridded emissions of NH3, as described in section 2.4 (L. 368-370, p. 8): “It takes into account the gridded column concentrations of ammonia that were calculated with the IDW interpolation method and all the potential removal processes of ammonia occurring in a hypothetical atmospheric box. . .”.

- Line 581: What are large sources of anthropogenic NH3 in central USA?

Response: We explain this, two paragraphs before this point. Please check manuscript with Track Changes (l.599-618): “First, a small region in Colorado, Central US, which is the location of a large agricultural region that traditionally releases large ammonia emissions. . .”. Then, we continue explaining main sources in Central US “is the state of Iowa (home to more than 20 million swine, 54 million chickens, and 4 million cattle), northern Texas and Kansas (beef cattle) . . .”. We think it is a repetition to mention again and again something that has been explained a few lines before.

- Figure 8 is very hard to interpret. The authors should consider alternative colors or another way to present these results.

Response: We have chosen to use the Gaussian kernel density estimation (KDE) method due to the large amount of data that we had to process, and we thought we should avoid overplotting. Another way to show the improvement of the results would be simple scatterplots that present annual data from all 4 simulations (Fig. 1). The reviewer/editor can possibly decide which one shows better. We would rather prefer the KDE method.

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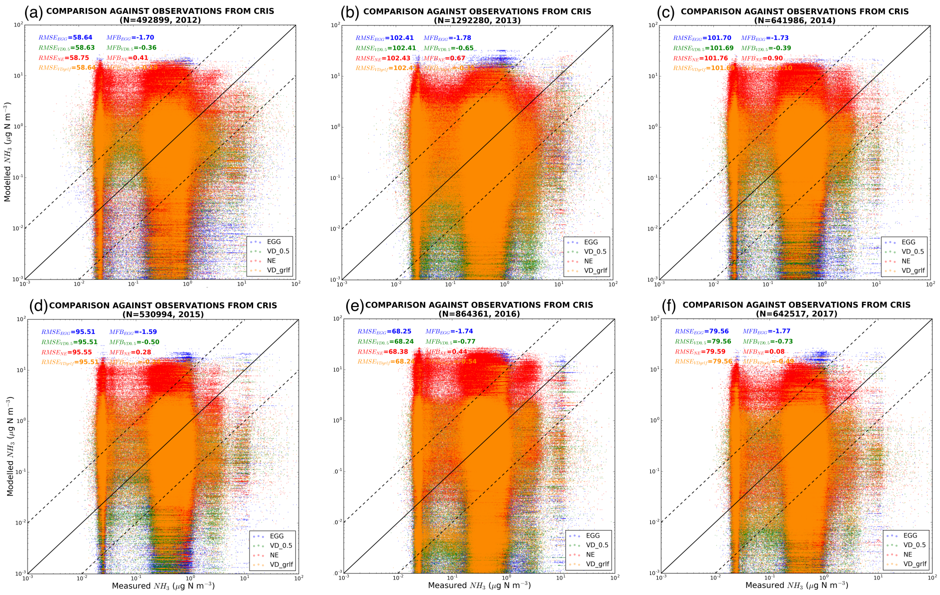

Fig. 1. Annual scatterplots of modelled versus CrIS NH3 surface concentrations from the relevant four simulations using emissions from EGG, VD0.5, NE and VDgrlf.