

Atmos. Chem. Phys. Discuss., referee comment RC2
<https://doi.org/10.5194/acp-2022-216-RC2>, 2022
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Comment on acp-2022-216

Anonymous Referee #2

Referee comment on "Estimating global ammonia (NH₃) emissions based on IASI observations from 2008 to 2018" by Zhenqi Luo et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2022-216-RC2>, 2022

Ammonia emissions estimates constructed from bottom-up inventories currently have large uncertainties. Top-down estimates of ammonia emissions using ammonia retrievals from satellite-borne instruments have the potential to greatly refine these bottom-up estimates. This paper uses ammonia retrievals from IASI to construct a global top-down estimate of ammonia emissions. The authors build on the work of Evangeliou et al., 2021, offering several improvements. These new emissions estimates yield much fewer emissions globally than derived in of Evangeliou et al., 2021, but significantly larger than the bottom-up estimates.

While a number of studies have recently been conducted that use similar methods to derive ammonia emissions regionally, currently few studies have provided top-down ammonia emission estimates globally. As such, this is a timely and scientifically relevant paper. Overall, the presentation of this paper is good, but some re-organization should be considered to improve the paper's readability. Additional analysis is also required before publication, as outlined below.

Major comments:

- I could not see any comparisons with an independent set of observations (such as surface observations). Validation with another set of observations needs to be added. For this, calculations of FB, R², and RMSE should be examined (as done in Table S2).
- Section 2.1 should contain some discussion of the uncertainties associated with the IASI NH₃ retrievals. There is a reference to using the relative errors that are reported with the retrievals in Section 2.4, so maybe part of line 183 could be moved to Section 2.1? What are typical uncertainties on the IASI retrievals?
- The information related to the sensitivity analysis seems out of order, as this information is spread out over different sections. Also, by referring to Table S1 in Section 2.1 and 2.4, it seems to present results of the emissions inversion before the main results are presented. I would move the sentences starting with 'To reduce uncertainty' on line 94 to the end of line 97 to either Section 2.3 or 2.4. I would

consider moving the sentence starting with 'We also test' on line 160 to Section 2.4 as well. Also, I think Table S1 should be moved from the Supplement to the main body of the paper. I also think it would be better to move the second paragraph in Section 2.4 that start on line 186 to the end of Section 2.3. I cannot see much discussion of the results of the sensitivity analysis in the 'Results and discussion' section. More discussion of the sensitivity analysis should be included. In addition to discussing the range of results of the global emissions from the sensitivity analysis, it would be good to add results/discussion/figures for the sensitivity analysis for emissions on regional scales. For example, would it be possible to add a plot like Fig. 2b, but for it to show the deviation from the TDE results instead (maybe as a percentage of the TDE emissions) when the parameters in Table S1 are varied?

- At the end of Section 2.2, you mention that averaging kernels are not provided in the retrieval product used. Including the averaging kernels in the calculation of the columns could potentially significantly change the column values. Is there reason to think the effect of the averaging kernel on the column is small? If so, could you provide some details on this? If not, is it possible to make a rough estimation of the averaging kernel and see how much this changes the results? Also, the two sentences starting with 'To compare' on line 129 until the reference to Van Damme et al. 2017 on line 131 should be moved elsewhere (maybe to Section 2.3 or a new subsection with the new information requested).
- More discussion of the rationale for using the formulation in Eq. (1) is necessary. Different mass balance methods use different methods to determine the proportionality constant between the columns and emissions. For instance, in the finite difference mass balance (FDMB) method, this proportionality is determined by comparing two different model runs: one run using the a priori emissions and one run with perturbed emissions. In the FDMB method, if you assume that you have a simplified model that assumes a steady state and no transport, the proportionality constant will be $1/\tau$, as in Eqn. (1). However, in general these two different methods will differ. So I'm curious what the rationale in choosing this method over the FDMD method is. Do you expect that the two methods would give similar results? Alternatively, an inversion/assimilation method could be used instead, where the proportionality constant would instead be given by the Kalman gain, which takes into account the uncertainties of both the observations and the a priori emissions estimates. Could you describe the advantages and/or disadvantages of using the Kalman gain instead of a mass balance method? In this context, I'm assuming the Kalman gain would be a scalar just like τ , not a (large) matrix that would be employed in a Kalman Filter method such as an EnKF that would obviously be very computationally expensive.
- On line 143, you mention that you use the lifetime for NH_x instead of NH_3 . The lifetimes of the two will differ, but I didn't quite follow why you would want to use the lifetime of NH_x if the estimation is for the emissions of NH_3 . Could you add more explanation why this makes sense conceptually?
- In regards to Eqn. (3), if the concern is that SO_2 emissions are underestimated in the bottom-up emissions, and you would like to make a correction for this, why not just increase the bottom-up emissions estimates by the same amount (i.e. ω)? Is the method used easier to implement? If the increase in SO_2 emissions are fed into GEOS-Chem, then ISORROPIA-II can work out the details of the NH_3 - NH_4^+ partitioning, i.e. when NH_3 is in excess, etc...
- One line 186, you mention that 'we perform GEOS-Chem full chemistry simulations in selected years'. What was the GEOS-Chem mode run initially for the full 2008-2018 period? This should be moved to Section 2.2 and expanded on, i.e. give a brief description of the simulation modes and their difference, why you chose to use the full chemistry mode for only these three years, etc... Also, with the way the paper is currently organized, it is a bit confusing figuring out which simulation mode was used for what. I assume that the non-full-chemistry mode was used with Eqns. (1) and (3), but I can't see where this is specified in the paper. Please clarify in Section 2.2 exactly what the output of each simulation mode is used for. A table might make this clearer to

the reader.

- In regards to the calculation of fractional bias of IASI as compared to GEOS-Chem, if the IASI observations were already used in the estimates of the emissions through comparisons to GEOS-Chem (Eqns. (1) and (3)), what is the goal of presenting these FB values? This comparison uses the full chemistry mode (I assumed that Eqns. (1) and (3) were used in non-full chemistry mode, is that correct?), but I'm not clear on what this comparison is telling you. Add more explanation of what this is being used for. If comparisons with an independent observation data set are added, as suggested above, is this comparison still necessary?
- On line 263, it says that the 'large post-2013 increase is inconsistent with flat or even declining fertilizer input and manure amount (Fig. 4).' Do the plots in Fig. 4 include emissions from livestock? If not, it may be hard to really tell if the trends found by using Eqn. (1) are really inconsistent with the data in Fig. 4.
- On line 275, it described that SO₂ trends from OMI and OMPS are used with Eqn (3). The estimates of SO₂ trends from OMI and OMPS will have uncertainties associated with them. Could these uncertainties be incorporated into your analysis? i.e. could you include another sensitivity test for the value of the SO₂ trend used?

Minor comments:

- Line 31: 'particulate matters' to 'particulate matter'
- Line 33: 'These particulate matters also affect' to 'Particulate matter also affects'
- Line 35: '... and wet processes, and therefore are associated with ...' remove the word 'therefore'
- Line 50: 'depending' should be changed to 'dependent'
- Line 136: Would be good to remind readers here that $C_{\{NH_3,obs\}}$ are the monthly mean observations, not an individual observation.
- Line 180: Replace 'related to the number of IASI measurements ($\delta_{\square\square\square}$) and their measurement errors:' by 'given by'
- Line 187: 'top-down estimate (TDE) and prior emissions (BUE1)' the acronym TDE was already defined on line 134, while the acronym BUE was used prior to this line but not defined until this line. Change so that these acronyms are defined at their first appearance in the paper.
- Section 3.2 starts by introducing Fig. 2 and then introduces Fig. 3 in the next sentence. Try to reorganize this section by first discussing Fig. 2 and then move on to discussing Fig. 3 instead of introducing them at the start of the section together.
- In regards to Fig. 2, I'm a bit confused about the colour bars and the colour scale. Fig 2c shows the difference between Fig 2a and 2b, so the units should be in $g\ m^{-2}\ a^{-1}$, but Fig 2c has blue in it, which isn't in the colour bar in the top row that has these units. The bottom row colour bar has blue in it, but it has the wrong units for 2c ($g\ m^{-2}\ a^{-1}$ per decade). Could you clarify which colour bar goes with Fig 2c? Could the figure be rearrange or changed in some way to make this more clear?
- In the description of Fig 3, it says 'Shadings represent the range derived from uncertainty analyses' Table S1 describes a number of different sensitivity tests. Does the shading refer to the upper and lower bounds over all tests? Please specify in the main text.
- Line 210: 'Both the satellite and model do not find significant trends in NH₃ concentrations over India (absolute value less than 1 % yr⁻¹).' Looking at Figs. 1(c) and (d), it looks like the trend over India are not insignificant. Could you confirm this?
- Line 261: Why is Eqn. (1) 'based only on NH₃ column measurements' if Eqn. (1) blends information from the observations with a priori information of the emissions?

- Line 295: Is the lifetime of 21.2 +/- 3.8 h the lifetime of NH₃ (as stated) or is it the lifetime of NH_x?
- In Table S1, 'TDE' isn't really a 'parameter perturbed'. This line should probably be something like 'None' or 'None (TDE)'.