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comment

Interactive comment on "Estimating global surface ammonia concentrations inferred from satellite retrievals" *by* Lei Liu et al.

Anonymous Referee #2

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This is a useful, but also rather coarse and preliminary, study using IIASI satellite retrievals to provide information about surface NH3 concentrations and trends. The study uses coarse resolution GEOS-Chem vertical profile information with column information retrieved from IIASI. Overall there is convincing correspondence of the IIASI retrievals with surface concentrations- although more robust statistical analysis beyond showing correlation and bias is precluding understanding how good (or bad) the correspondence really is. How many observations were within a factor of two of the satellite derived values, is there a difference between rural, semi-urban, urban values, etc.

One of the issues that have always remained a bit of mystery to me with regard to the satellite retrievals is the importance of using a-priori profile information for the retrievals, given the lack of sensitivity close to the surface and possibly also saturation effects

where concentrations are high. This paper adds another question mark: when on the one hand the retrieval uses profile information, and in a second step the method uses the same model to calculate surface information is there a potential for using twice the same information? Also unclear to me how the vertical resolution of GEOS-CHEM can resolve the strong vertical gradients that are likely to exist in source regions. The authors should indicate 1) the vertical structure of the model 2) the measurement characteristics of the surface observation (including height), 3) how this information is used to calculate surface concentrations. Even if the authors will not resolve all issues, a thorough discussion and way-forward discussion are needed about this.

For these reasons I recommend major revisions to his paper. I would ask the authors to already put on-line the vertical model information- for use by other readers.

Detailed comments.

l. 24 Not only for dry deposition, also for modelling of the formation of ammonium nitrate.

l. 32: I am wondering what a high correlation of satellite/surface obs taken all regions together is really telling? I think it mostly depends on getting the levels of the 'high' concentrations correct, which is confirmed by the numbers given later. Consider removing this statement

l. 65. They have not been established to measure NH3 by itself, but as one of the parameters of a larger range of pollutants.

l. 110 -116. It seems a selling point to suggest that vertical profile information for NO2 and SO2 have been useful for modelling, and therefore also for NH3. The issues is more complex for NH3, with sources almost entirely close to the surface, and a complex mix of source and sinks, which will make the model profile more dependent on the mixing characteristics of the CTM. For NO2 and SO2 the sources and locations are better known, most of SO2 is nowadays emitted also well above the surface, which

makes interference with the dry deposition process less sensitivity to errors.

l. 134: GEOSCHEM vertical profiles were used in the retrieval and subsequently used to derive surface concentrations=>explain

l. 147. Please summarize some characteristics. E.g. error statistics, characteristic of sites (urban, rural, vicinity to direct sources) height of observations that we need to know to understand the ability of a course resolution model to represent those sites. For all 3 networks!

l.153 Need to explain why temporal averaging to 2 weeks is done.

l. 157 measured systems=>measurement systems

l. 164 Please give the characteristic heights of the layers in the boundary (mixing) layer. And how were emissions distributed?

l. 160 Any spin-up considered?

l. 174: how does this compare to the widely used HTAP2 emissions for 2010? What changes to EDGAR result from substituting with regional inventories. Any seasonality applied and what would be the reference for it?

l. 186 some clarification is need as regards the use of 'local' times, which I think are not considering any shift in legal times (i.e. winter/summertime).

l. 206. It is not clear why this more complex fitting procedure is needed. What was the problem, and how is solved by this new fitting. In figure S2 vertical profiles are shown- but I do not have the information to understand if GEOSCHEMs vertical resolution would be able to resolve such profiles.

l. 223 like mentioned earlier, the correlation is merely due to the fact of having very high concentrations in one region, versus low in other regions. If one would compare the low concentration range a factor of 5 or so difference in concentrations belong to a single column value would be found. And maybe in reality even more, depending on a whole

lot of things. l. 234 also this seems to be a rather error sensitive approach. I am mostly concerned about possible co-variance between the intra-day emission variations and the limited sampling at 9 O'clock. Are there observations that can be used to explore this issue?

L 242 Something wrong with reference E et al. And Van et al. And maybe other references as well.

l. 241-249 I am confused about the Figure S1 (with typical maxima of about 20-40 meter) and the statement that models and aircraft measurements can be used to verify them. I think that high towers like the one at Cabouw (NL), and observations at several levels, are probably the more reliable verification, but unfortunately there are not many of these.

L 250 I have no idea what heights the first and fifth layer are corresponding to.

l. 284 It is true that NH3 can be more accurately be retrieved in one region than another depending on the thermal contrast. But it is not clear to me why this would be so much better in China than e.g. in the US? I guess it is also just a matter of detection limits? It could also be related to more reliable simulation of mixing, depending on sufficient observational input into the parent weather model.

l. 296 the fixed profiles=>fixed profiles (language)

l. 304-312 I believe that there can be a large difference between 40-60 meters, but as the authors explain this is all in the same geos-chem layer. I fail to understand how this information is then used to interpret geos-chem profiles.

l. 323 this is mostly a confirmation that crop mask used by the regional and global emission inventories correspond to the MODIS one. And that the fertilizers used in those countries indeed end up on these fields.

l. 355 According to inventories, in Europe about 20 % of NH3 emissions is related to the use of mineral fertilizer, and 80 % to manure managements. So it would be more

relevant to determine the correspondence of those emissions (mineral is often used to top up what wasn't provided by manure).

364 Van et al. Please check this reference. It seems to be a problem of you reference manager.

370 It is probably opportune to refer to the paper by Pozzer et al, a 1x1 model study that has more extensively studied the role of NH3 emission for aerosol. It is also important to critically assess ammonium nitrate measurements, which are notoriously difficult at higher concentrations. Would it be an option to use the model to estimate (equilibrium) ammonium sulfate and nitrate concentrations associated with the 'retrieved' surface ammonia?

421-430 Biomass burning can be an important source of NH3, especially in the smoldering phase. Therefore, I have some doubts that active fire products are the best proxy for Nh3 emission. Did you consider burnt area products instead?

495 leaded=>led?

504: why inconsistent? It could be rather consistent, as you explain in the following sentences.

Supplementary:

Figure S3: Please indicate what heights approximate correspond with the first and fifth layer boundaries.

Figure S4: no idea what heights these layer correspond to.

Figure S5: the figure caption is not self-explaining.

Figure S6: first PM2.5 and then NO2 in caption.

Figure S9: describe upper panel as well.

Figure S10: trends of what (annual concentrations?) and for what period.

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