Review of “Estimating agricultural ammonia volatilization over Europe using satellite observations and simulation data

Summary

This paper sets ambitious goals: to estimate current ammonia volatilization over Europe from satellite data and to predict how it will change as Europe warms. These are interesting ideas, that could potentially provide useful data to the air quality community, due the strong connection between ammonia concentrations and PM$_{2.5}$ amounts. However, given the many uncertainties in the calculation of the soil emission potential (such as the mass transfer coefficient), the work in this paper is more of a roadmap on how this might be done rather than a set of reliable estimates. Nevertheless it is still a valuable paper for the community, as it demonstrates the methods and the types of datasets that are needed to achieve its stated goal.

The paper is overall well organized, though a few sections are confusing and need rewriting. Once the suggested revisions are made, the paper should be accepted for publishing.

Technical comments

Section 3.1:

The authors attribute some of the difference between IASI and GEOS-CHEM to different sampling times: IASI only measures NH$_3$ at 9:30 am, while the GEOS-Chem output was averaged over an entire month. Wouldn’t it be possible to eliminate this disparity by sampling GEOS-Chem only at 9:30 am? Please explain why this was not done.

Section 3.2

The authors state that the ammonia lifetime in New Aquitaine is high due to air stagnation. Why is this region prone to stagnation in March?

I do not think the authors should state that the GEOS-Chem lifetime estimates agree with the Evangeliou results, since the former range from 1 to 13 hours and the latter from 10 to 13. Please rewrite this statement.

Can the authors explain why the loss to transport in England is lower than in Ireland, even though it is also affected by the Gulf Stream?

Section 4

The text between lines 369 and 390 is extremely confusing, in part because Figure 5 is referenced before the differences between the four plots are explained. The calculation of the fixed k is also a bit hard to follow. Finally, in the caption for figure S3 it is stated that cases 1, 2
and 3 have identical soil emission potential, which is not true, since case 3 uses IASI rather than GEOS-Chem NH3. Please reorganize and rewrite this section and make it clearer.

The comparison between the soil emission potential from IASI and GEOS-Chem for the cases of England and the Po Valley uses percentages that are not consistent with the values listed in Table 1. Please explain how they were calculated. And what are the three values of Tskin listed in Table 1?

The discussion of Figure 6 (lines 435 to 448) mentions the average temperature and states that in the Po Valley Tskin from ERA5 is twice as high as this average temperature. Why is this relevant? Don’t cases 1, 2 and 3 use very similar temperatures (ERA-5 or MERRA)? There is no mention of average temperature in description of the cases. Please explain. The statement that the inter-variability between the cases does not depend on the lifetime does not seem to be true. Maybe which inter-variability needs to be defined?

Section 5

On line 472 the author state that current and future ammonia columns are calculated assuming that the emission potential is unchanged. If the whole point of the future climate modeling exercise is to look at effect of changing temperature on the volatilization of ammonia, and the emission potential is strongly dependent on temperature, this sentence does not make sense.

Appendix

The referencing of multiple equations in the appendix was wrong. I have corrected them, but please check and make sure my changes make sense. Finally, please also confirm that the calculation of the first constant in (A-9) (2.75e9) is correct. I was unable to reproduce this value, but that could be an error at my end.

Minor edits (suggested changes are in bold)

Line 42: ... amounted to

Line 43: ... a very reactive base, and constitutes ...

Line 47: ... total ammonia gas is believed ...

Line 60: ... as shown in the Appendix.
Line 61: ...of ammonia in the water in the soil is a function of soil acidity (pH) and temperature ...
Line 62: ...and controlled by the dissociation ...

Line 63: ...exists in the gas phase, and therefore Henry’s law can be used to describe ....

Line 84: ...during the 2003-2019 period ...
Line 85: ...(2022), leading to increased volatilized ammonia, (due to increase in both nitrogen ...

Line 87/88: ... Between the years 2008 and 2018, the ... columns is estimated to be ...
Line 107: ... ammonia to provide regional ...

Line 116: where

Line 120: ...c’ is 100

Line 123: ... It is a function of the roughness length ...

Line 125: The sentence starting with “It can be explained by ...” is unclear. Are the authors stating that a resistance model is used to calculate k?

Line 177: ... in areas where fertilizers are applied.
    Show the emission potential where?

Line 204 : ... we use the ECMWF European Earth Consortium climate model ...

Lines 216-219:... two scenarios: the SSPP2-4.5, a “middle of the road” socio-economic scenario with a nominal 4.5W/m2 radiative forcing level by 2100, similar to the RCP-4.5 scenario, and the SSP5-8.5, the upper edge of the SSP scenario spectrum with a high fossil-fuel development use the 21st century.

Line 238: ...2011), which marks ...

Lines 239-240: ...The differences are likely due to sampling issues: only cloud-free data are used to retrieve ammonia and different sampling times: IASI ....

Line 248: ... Therefore, assuming that meteorological ...

Line 259: What does [not shown here] mean?

Line 275: The lifetime of ammonia () is shown in Figure 2d.

Line 285: ... air stagnation in that area
and (AQEG, 2012), and these PM2.5 particles can dissociate, releasing ammonia. Considered the loss ...

Adopted here ...

Literature. Note that ammonia transfer coefficients are not available for all land types.

In grey in Figure 4.

Therefore, this value was assigned to croplands...

The sentence starting with “These values “ should maybe be rewritten as:

These values obtained by using MODIS land cover types and published estimates of k values represent our best effort to realistic mass transfer coefficients, and therefore realistic soil emission potentials.

Are the authors extrapolating or aggregating by averaging over each GEOS-Chem grid box? Please make this clear.

The k value assigned for forests represents the SO2 exchange in high croplands; this value may be very different for ammonia, since NH3 can easily dissolve in the water film on leaves under conditions of high humidity.

Again I think the authors mean aggregate not extrapolate. Which several grids? Isn’t the MODIS grid just being aggregated to the GEOS-Chem grid?

Using a land type ...

What is the sentence starting with “Based upon …” supposed to convey?

The emission potential does not agree in value with that of GEOS-Chem.

England, northern France, northeastern Spain and Poland....

Potential with values ranging from ....

Are croplands different from agricultural lands? If not, the sentence starting with “Our values” seems unnecessary.
In this study, lower values than those measured in the field are expected. Therefore, we consider our results to be in good agreement with the values in Personne et al. (2015), since ours reflect a 31 day mean of an average of over a large area (55 x 70 km²).

Therefore, we consider our results to be in good agreement with the values in Personne et al. (2015), since ours reflect a 31 day mean of an average of over a large area (55 x 70 km²).

Caption of Figure 6: … are explained in the discussion on Figure 5.

more severe in eastern Europe …

up to +50%...

… facing big challenges in air (??) or downwind of large agricultural regions …

…where there is no ammonia …

Appendix

where \( H_{NH3} \) is Henry’s constant, which can be …

Substituting Eq.(A-7) into (A-8) we get:

Since in …

where \( \text{Flux}_{NH3} \) …

Eq. (A-11) can be written as ..

using Eq. (A-9) in (A-13) we get: