

Reviewer #3

We wish to thank the referee for his/her helpful comments. The full reviews are copied hereafter and our responses are inserted. The comments of the reviewer are in normal black and our answers in bold.

GENERAL COMMENTS

The manuscript submitted by Fortems-Cheiney et al. assesses different methods to determine NH₃ emissions that are used in the scientific community. The authors compare three state-of-the art approaches for NH₃ emission retrievals and discuss their spatial and temporal differences over a period in Spring 2011, which also includes the time of fertilizer application. While the analysis was performed for France, the discussed discrepancies between methods is also of high interest to researcher focusing on other regions. Due to the general high uncertainties in NH₃ emission estimates, the presented study is of high relevance for improving chemical transport models. The manuscripts is well and clearly written and the analysis and interpretation of results is sound. Given the relevance of the topic for air quality, nitrogen deposition and atmospheric chemistry in general, I suggest approving the manuscript for publication in Atmospheric Chemistry and Physics after addressing the following comments.

SPECIFIC COMMENTS

L. 34-35: I suggest to clarify that the given values are the cumulative emissions from March to May. It is mentioned above, however, it would be good to restate here to not confuse it with an emissions flux.

We have changed the sentences to clarify this: “The total spring budgets, from March to May 2011, at the national level are higher when calculated with both alternative inventories than with the reference one, the difference being more marked with CADASTRE-CIT. NH₃SAT and CADASTRE-CIT inventories both yield to large NH₃ spring emissions due to fertilization on soils with high pH in the northeastern part of France”.

L. 123: I assume the study period was from 1 March to 31 May? Still, the day should be included in the period description.

The days have been included.

L. 217-219: Above it is stated that only morning overpass measurements are used. This may introduce a systematic bias to the NH₃SAT emissions as emissions may be larger during midday due to higher temperatures or increased activity (e.g. mobile emissions). If I understand it correctly, the effect also depends on the TNO-GEN input data for CHIMERE, i.e. if monthly or a diurnal profile is applied to that. While I agree with the decision to only use morning overpass data, this potential bias should be explained and how it may impact the findings (e.g. could it explain why NH₃SAT emissions are lower than those of CADASTRE-CIT in northeast, where fertilizer emission are high?).

Indeed, only morning overpass measurements are used, when the emissions are not at their maximum. These morning IASI data are compared with NH₃ simulated columns at the same hours in the morning, using the diurnal profile used in the TNO-GEN inventory. This is introducing an uncertainty to the NH₃SAT emissions, but not a systematic bias.

One perspective for this question would be an inter-comparison using both IASI and CrIS (with an overpass in the beginning of the afternoon). Unfortunately, CrIS was not flying during spring 2011, our focused period.

L. 219-221: Could one also take only those periods where the IASI quality criteria are met and omit the other periods for the analysis instead of using the TNO-GEN emissions? How would that change the average correction factors presented in this study?

As a test, we calculated a mean daily corrective factor for each region A, B, C, D and we applied this corrective factor at the grid-cell scale when IASI super-observations are not available. This results in the daily variability shown in light orange below, showing for

example higher emissions in the northeastern part of France the second week of March. Nevertheless, the small number of IASI pixels taken into account for this corrective factor may be partially contaminated and may not be sufficiently robust. We prefer not to integer this test in our study.

L. 246-247: From this paragraph alone it is not clear whether the manure field spreading is included the organic fertilizer application, which I would assume. If that is the case, you may add here “..., which is part of the organic fertilizer emissions” or similar.

The formulation is possibly incorrect: all husbandry effluents are considered, whether they are liquid or solid, what we thought was contained in the term "manure" as opposed to "farm yard manure". The authors have modified the sentence to remove any ambiguity into:

“For livestock emissions, with the exception of the stage of effluent spreading in the field, the less detailed inventory of the French Interprofessional Technical Centre for Studies on Air Pollution CITEPA is used.”

L. 314: In the Results & Discussions section the authors describe the differences between the different approaches and also give here and there some suggestions why these differences occur. Still the discussions can be extended at the end of the section, e.g. by elaborating more on the uncertainties of each approach, leading to recommendations for future research directions and at which part each of the approaches should be improved (e.g. need for implementation of bidirectional exchange module in CHIMERE?). Although the paper is targeted on France, the findings are also insightful for users of NH₃ emission estimates in other parts of the world. Therefore, relating the findings in the discussion also to inventories/approaches used in other regions would in my opinion make the manuscript stronger and attract more interest by a wider community.

Thank you for this suggestion, which allows highlighting the implications of our study. As already answered to referee 1, we added a couple of sentences at the end of the Conclusion section:

“Yet, current results of our study have important implications for air quality modelling over Europe. The important changes in the spatial distribution of NH₃ emissions as a function of soil properties is of general concern not only for France, but for whole Europe. Soils are alkaline or neutral (pH>6) not only over North-Eastern France, but also over large parts of Italy, eastern Spain, or eastern Germany [Reuter, 2008]. Over these regions, our study suggests potentially larger NH₃ emissions than with a constant emission factor treatment, with impacts then on fine particle formation. These features should be included in “operational” emission inventories used for air quality modelling.”

Reuter, H.I., Lado, L.R., Hengl, T. and Montanarella, L.: Continental-scale digital soil mapping using European soil profile data: soil pH, *Hamburger Beiträge zur Physischen Geographie und Landschaftsökologie* – 92 Heft 19/2008, pp. 91-102, 2008.

L. 428: I suggest restating here in short the hypothesis from the introduction.

We rather have removed the reference to the hypothesis from the introduction in this sentence.

L. 405-408: Would market gardening be included in the TNO-GEN inventory? If not, an underestimation in that region would also apply to the TNO-GEN inventory.

To our knowledge, market gardening is indeed not included in the TNO-GEN inventory. We have changed the sentences: “Over the southeastern part of France, CADASTRE-CIT is about 23% lower than NH₃SAT (28 and 37 ktNH₃, respectively, Table 2). One hypothesis to explain the lower NH₃ emissions in CADASTRE-CIT is that market gardening is

important in this area and not taken into account in the CADASTRE-CIT inventory [Ramanantenaso et al., 2018; Génarmont et al., 2018]. Nevertheless, market gardening is not included, to our knowledge, in the TNO-GEN inventory. TNO-GEN and NH₃SAT inventories being in quite good agreement in terms of budget (35 and 37 ktNH₃, respectively, Table 2), further work is required to understand these discrepancies.”

TECHNICAL COMMENTS

Some words are accidentally merged, for example in lines 38, 50, 51, 93, 139, 196,197, 207, 209, 210, 211, 272, 287, 291, 320, 371, 381, 400, 433, 466, 475 and 478.

We apologized for the inconvenience. It has been corrected.

The description and spelling of regions like “North-Eastern France” vs. “northeastern France” should be consistent throughout the manuscript.

This has been corrected and the description of the different regions are now consistent throughout the manuscript.

L. 26: The sentence structure can be improved as it can be misleading what the actual methods are which the authors refer to. For example, I suggest inserting “:”, “namely”...or something similar after “emissions”.

The sentence has been changed: “In this study, we compare NH₃ emissions in France during the spring 2011 from one reference inventory, the TNO inventory, and two alternative inventories that account in different manners for both the spatial and temporal variabilities of the emissions: (i) the NH₃SAT satellite-derived inventory based on IASI NH₃ columns and (ii) the CADASTRE-CIT inventory that combines NH₃ emissions due to nitrogen fertilization calculated with the mechanistic model VOLT’AIR on the database of the CADASTRE_NH₃ framework and other source emissions from the CITEPA.”

L. 292: I am not familiar with this terminology but there might be better terms than “desegregation” and “reagregation” in English.

“Desegregation” and “reagregation” are dedicated terms that refers to the fact of distributing the emissions obtained at an initial scale to another entity and then reallocating them to different scales (here to grids compatible with the CHIMERE model, according to the distribution of crops in that grid cell). We do not know alternative terms and we kept them in the text.

L. 379: I suggest using “In bold are marked...”.

We have changed the sentence: «Regions for which the inventories NH₃SAT and CADASTRE-CIT propose the same sign of relative differences are marked in bold»

L. 401: A closing parenthesis is missing.

It has been corrected.