

Interactive comment on “Contributions of natural and anthropogenic sources to ambient ammonia in the Athabasca Oil Sands and north-western Canada” by Cynthia Whaley et al.

Anonymous Referee #1

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Contributions of natural and anthropogenic sources to ambient ammonia in the Athabasca Oil Sands and north-western Canada By Cynthia Whaley, Paul A. Makar, Mark W. Shephard, Leiming Zhang, Junhua Zhang, Qiong Zheng, Ayodeji Akingunola, Gregory R. Wentworth, Jennifer G. Murphy, Shailesh K. Kharol, and Karen E. Cady-Pereira

The topic of the paper is about improving modeling representation of NH₃ concentrations in the Alberta Oil Sands region, by running the GEM-MACH model with new parameterizations for bidirectional exchange of NH₃ and NH₃ emissions from fires.

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The results are based on previous simulations proposed by Shepard et al. (2015) and are compared to surface, aircraft and satellite measurements.

General comments:

The subject of study (evaluating the model capability to reproduce NH₃ concentrations) is interesting, and the paper is easy to follow. The structure is well organized. Some sentences need rephrasing, and some explanations are confuse (see specific comments).

Citations of non available papers are given (cited as “this issue” but non available in the special issue). This stands for example for description of measurements (Wentworth et al), deposition of N and S (Makar et al), parameterizations (Akingunola et al), fire emissions (Zhang et al.).

NH₃ concentrations are really low in the region of study and the reader may wonder why this region is worth studying, in the light of what is written in the introduction, (NH₃ may be harmful for air, water quality, or ecosystem and human health). A sentence or 2 on the relevancy of studying regions where concentrations remain low for the moment would be useful.

Some citations are given for satellite measurements of NH₃ concentrations at a large scale, but nothing is said about orders of magnitude. Do the model results of this study match with previous satellite measurements? More values of ambient concentrations should be cited. This is also true for bidirectional exchanges: some papers are cited, but only as a list of papers, and no quantified values are given to be compared with what is found in this study. The papers should be cited with precise examples of measured fluxes is the same area or in regions with the same type of ecosystems.

A meteorological description of the site would be useful all along the study. Indeed, emissions have their impact on NH₃ concentrations, but wind speed, humidity, temperature have also a significant impact on exchange fluxes. This part of dynamical

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interpretation on deposition fluxes is missing. A discussion about why the addition of bidirectional flux is so important in improving the model is missing in terms of processes. The discussion is only about ppb and %, and not about processes.

1- Does the paper address relevant scientific questions within the scope of ACP?: yes
2- Does the paper present novel concepts, ideas, tools, or data?: not really, bidirectional exchanges of NH₃ is already known to be important, tools and data have already been used in Shepard et al (2015)
3- Are substantial conclusions reached?: conclusions are reached
4- Are the scientific methods and assumptions valid and clearly outlined? yes
5- Are the results sufficient to support the interpretations and conclusions? yes
6- Is the description of experiments and calculations sufficiently complete and precise to allow their reproduction by fellow scientists (traceability of results)? yes
7- Do the authors give proper credit to related work and clearly indicate their own new/original contribution? yes
8- Does the title clearly reflect the contents of the paper? yes
9- Does the abstract provide a concise and complete summary? yes
10- Is the overall presentation well structured and clear? yes
11- Is the language fluent and precise? Not always
12- Are mathematical formulae, symbols, abbreviations, and units correctly defined and used? yes
13- Should any parts of the paper (text, formulae, figures, tables) be clarified, reduced, combined, or eliminated? Yes, mentioned in specific comments
14- Are the number and quality of references appropriate? Not always

Specific comments

Line 42: As you write that NH₃ is a contaminant, precise in what order of concentration it has negative effects.

Precise somewhere in your introduction that despite negative effects of high concentrations, low concentration regions are also worth studying.

Line 44: Modeling provides. . .: this could be true if inventories are correct and with fine resolution, which is hardly feasible in most models. Remove this first part of the sentence.

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Line 55: reformulate your sentence because reading “the AOSR is a large source of air” is a bit weird.

Line 58: why are NH₃ concentrations so low, despite local pollution?

Line 61: give mean concentrations of cited agricultural areas.

Line 66: give values of the fraction of deposited NH₃ compared to NO₂ and HNO₃.

Line 114: precise what species are used from the inventories.

Line 121: A word about the importance of carefully design stack parameters would be useful to understand why this part is so important for your study.

Line 167: give reference values of realistic NH₃ concentrations consistent with findings from the literature, and explain why you use low end values.

Line 191: what do you mean by “major point emissions?”

Line 220: what is ECCC?

Line 259: you mention +/- 15% of uncertainty for measurements, with a mean measured value of 0.63 ppb (line 59). The model/measurements bias is 0.3 ± 0.85 for bidi, what is the real impact on modeled concentration? What is the range of possible concentration? Is the measured concentration included in this range?

Line 299: Figure 3 should not be placed here. It is only used later in the text, and should be included before figure 9. Furthermore, if placed here, it is not understandable why these three dates are chosen (not explained in the caption).

Line 324: You mention fig 5 and then you talk about fig 4. Place fig 5 after fig 4 and give a description on interpretation of it.

Line 326: what are the background times?

In figure 4, NH₃ is in ppb, but NH₄⁺, NO₃⁻ and SO₄²⁻ are not in ppb. Please correct the caption

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Line 335: what is the increase of concentration with the influence of a local plume? 0.5 ppb seems to be very very low, and more included in the measurement noise than in a local pollution signal.

Line 337: 0.08 ppb is less than the 15% measurement uncertainty. Is it really significant?

Line 339: $R=0.2$ and 0.4 . Are these coefficients significant? Could you give a significance (p -value) of your correlation calculation everywhere it is necessary?

Line 340: Fig 6a: what is the unit of NH_3 concentration?

Line 368: SO_4^{2-} is influenced by anthropogenic emissions, why not by fire emissions?

Line 375: Why did you choose this precise flight? By the way, it would be useful to give some average values of meteorological conditions when describing the area of study (mean temperature, humidity, rainfall, wind speed, etc... all parameters that have a possible influence on NH_3 concentration)

Line 376: can you explain why fire+bidi does not improve the results compared to bidi?

Line 404: Figure 3 is used in this paragraph. It should appear in the text at this time, and not before.

Line 430: specify at the lowest level.

Line 446: remove "that" before the fire+bidi model

Line 451: You suppose that the conversion of NH_3 to NH_4^+ is underestimated: did you have a look at the NH_4^+ pool in that case?

Line 453 to 459: this explanation is very confused. Please rephrase.

Line 507: this sentence is not useful. Obviously if bidirectional parameterization is used it will balance deposition with emission and not increase deposition fluxes

Line 517 and elsewhere: the unit for the flux is not appropriate. Please homogenize

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throughout the paragraph and use preferably $\text{ngN.m}^{-2}.\text{s}^{-1}$. Lines 584 and 587 use another unit which is not a flux unit. In this paragraph needs bibliography values need to be included for equivalent ecosystem or region. Figure 12 and 13 are redundant. Figure 13 is not necessary in my opinion.

Line 539: again this sentence is not useful. Obviously wet deposition is only deposition. Again flux units are not correct and should be homogenized.

Line 550: how do wet deposition fluxes compare with literature?

Line 570-571: this sentence has already been written above. The conclusion should mention the possible influence of meteorological conditions on NH_3 concentration, as well as in the text.

Line 574: "miniscule": please quantify.

Technical comments

Line 16: remove , aftertime period.

Line 108: write covers instead of covering, twice in the line.

Line 109: remove "And" at the end of the line.

Line 117: remove (before Zhang and put (before 2017.

Line 163: replace "in" by "from" before Wen et al.

Line 228: Time period "from" . . .

Line 296: a verb is missing in the sentence.

Line 321: include "to" after compared.

Line 352: remove italics for "should", same line 488 for "total" and line 507 for "more"

Line 454: a) is missing after 10c

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Line 493: problem with the sentence, please rephrase

Line 507 and 509: remove , after 11.

Line 526: replace × by times

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