Review of Burrows et al., “Impact of modelled particle characteristics on emissions inferred by inversion of tracer transport”

Summary

The authors report on a method to estimate uncertainty in emissions and atmospheric transport of bacteria on the global scale. They combine simulations of the atmospheric chemistry transport model ECHAM5/MESSy-Atmospheric Chemistry (EMAC) with a Monte Carlo Markov Chain source estimation method to derive uncertainties in emissions and selected processes within the atmospheric chemistry model. They compare uncertainties found to observational uncertainties and find that model process uncertainties can contribute substantially to overall uncertainty, even for very uncertain measurements such as the ones of bacterial aerosols.
The manuscript covers two interesting topics: the source estimation of very uncertain but non-negligible (novel) types of aerosols, and secondly the estimation of uncertainty in models describing the atmospheric fate of these particles. Unfortunately there are some major deficiencies in how the results are presented. The paper is not well structured. Frequent references to the appendices which contain information required to understand the results makes it unnecessarily difficult for the reader to grasp the findings. Figures are prepared in a sloppy manner and some are clearly not publication ready (missing/overlapping labels, duplicate information, panels with different sizes).For these reasons is suggest publication only after major revisions.

Main comments

1. Almost all of the information in Appendices A1 and A2 is repeated within the text (4.2.1, 4.2.3, 4.2.4). Further, mathematical symbols used in the main text (e.g. Likelihood function “|D”, “N”, Theta) are not explained - one has to refer to the appendices. Remove A1 and A2 and incorporate the additional information into the main text.

2. Structure: the classical way to write a paper (Introduction, Methods and Data, Results, Discussion, Conclusion) might help to disentangle some confusion arising in this work:

    - Section 2: remove sub-caption 2.1, rename to “Observations” or “Bacteria observations”, and move 2.2 into model description.
    - Section 3: create section 3.1 “Model description” or similar, describing the model and its components (that is the current 3.1 + the old 2.2). Create new section 3.2 “Simulations and sensivity studies” describing all simulations made (including the modeling part of section 3.3 for SENS_COLD, SENS_MIXED, LIM_COLD, LIM_MIXED, and the information that is given later in 4.2.5). State the number of simulations made between 1um and 10um particles (I assumed 10, per 1um?).
    - Remove the respective descriptions of the model runs made from the following section and only describe the results in a new (sub-)section.

- Section 4:
  - 4.1 can be removed once there is no need to refer to the Appendices anymore.
  - add remaining information from A1 and 2 to 4.2 subsections
  - 4.3 make clear that 4.3.1 deals with the per-ecosystem fluxes, while 4.3.2 deals with the global total. Easily achieved by renaming section 4.3.1. Move “Sensitivity of global and regional emissions to particles size” to the beginning of 4.3.2. You describe figures A3, A4 in 4.3.1, so the logical consequence is to go on with this paragraph, and only after that discuss the global integral (4.3.2).
  - “Comparison with previous work” → move to 6. Discussion and conclusion.

3. This study can indicate errors in sources and transport, but also identify missing sinks of bacteria. It is not given (and actually rather unlikely) that the wet and dry deposition schemes implemented accurately represent reality. This problem is dealt with in the two NO-PRIOR and PRIOR-POS studies, where PRIOR-POS forces positive values for fluxes (i.e. emissions). The authors are, however, quick to discard the NO-PRIOR simulation and focus on the PRIOR-POS simulation, which I find unfortunate. They state themselves that “the typical posterior distribution has the shape of a Gaussian distribution [...]”, which in the case of PRIOR-POS is “[...] abruptly cut off at zero”.
   There is no reason (given) why the model system should only overestimate emissions and the loss processes should be correct. Also, I do not understand what the benefit of “decoupling the emissions” (last sentence in paragraph 4.3.1) is, and why it should reason the choice of PRIOR-POS over NO-PRIOR.
   Figure 2 is then the central figure showing emission estimates, and it is based on PRIOR-POS, making the implicit assumption that your loss processes are correct, potentially overestimating emissions.
   I suggest using NO-PRIOR in 4.3.2 and try to find an explanation on why certain ecosystems seem to work rather as sink than as source (precipitation? dry deposition? boundary layer mixing?...), as you partly did it in p 4405, “Sensitivity of global and regional emissions to particle size”.
   Overall, PRIOR-POS should at most be a sensitivity study, if not removed completely. If you still feel this is better than NO-PRIOR, you will have to justify this much better.

4. The study of Lelieveld et al. (2012) used in 3.1 to ascertain that the model has some skill to represent large-scale transport of aerosols is a risk assessment and does not focus on the accuracy of the underlying modeling system to quantitatively simulate tracer transport. While they cite several other studies that comprehensively evaluated parts of the modeling system, the only assessment of tracer transport accuracy made in the study cited by the authors is a qualitative comparison against a map of accumulated deposition, where Lelieveld et al. (2012) state themselves that “This qualitative agreement is quite satisfactory, especially because quantitative agreement cannot be expected [...]” (Lelieveld et al. (2012), p. 4248). This does not “establish the validity” as the authors claim.

The authors should cite other literature sources that comprehensively deal with the accuracy of the model. I deem this an overall comment, because it is used here to imply that the model does well overall and only some parameters are uncertain.This needs to be justified correctly to exclude underlying problems in the model which invalidate your sensitivity studies.

5. What was the reasoning for selecting the sensitivity parameters to study? It looks like a somewhat arbitrary selection. What about ice sedimentation velocity (and consequently removal of scavenged mass from a grid box)? Scavenging (or not) during convective ascent (entrained/detrained particles, e.g. in Croft et al. (2012))? Model resolution? Model timestep? I concede that the number can be endless, but I would expect some more justification for this set of parameters.

6. I guess it is out of scope for this work, but what would happen if you apply this method to a source of aerosols that are better constrained than bacteria? Does this mean that model uncertainties will overwhelm? Discuss.

Specific comments

p 4393 l 18: This sentence does not make sense: “While efforts to quantify emissions in models depend greatly on observations, these efforts can be classed...”. Should rather be something like “Two broad groups of efforts to quantify emissions can be distinguished:”

p 4397 l 2: simulated yr -> simulated years

p 4397 l 8: why “nearly all”? Explain (in the manuscript)

p 4398 l 10, Table 2: the table is superfluous and can be removed. The processes and (the only) difference can be summarised in a sentence.

p 4398 3.3: it does not read like you do consider the change in size distribution due to aerosol cloud processing (e.g. Hoose et al. (2008)). I guess it will be difficult to implement as you explicitly state you simulate monodisperse aerosols. However, this process will affect atmospheric lifetime as (and you show that yourself) bigger particles have shorter lifetimes. You should at least mention that this is not considered in your uncertainty estimates.

p 4398 l 7-8: “is treated as follows. For” -> “is treated as follows: for”

p 4399 l 1-8: modeling description part that would go into new simulations section.

p 4399 l 9-13: results part into new results section.

p 4399 3.4: part of results section.

p 4402 l 10: some symbols (Theta, |D, N) are not explained. Will be resolved once you include the appendices.

p 4403 l 10: explain “lowest acceptance rate”. Why “ca. 15 %”? Is given in the Appendix, but could be summarized in one sentence here and would help the reader.

p 4403, 16: more robust indication than what? Median? Mean?

p 4403, 4.3.1: Appendices represent ancillary material that further strengthens a point made in the main paper, but the manuscript itself should be complete without the Appendix.
Figures A3, A4, A1, A2 are a central to this paragraph. Hence they should not be in the Appendix. Either rephrase or move A3 and A4 to main text. Further, A3 and A4 are mentioned before A1 and A2, please reorder.

p 4405 “Sensitivity...”: Can you derive a measure to define the reliability of the estimates for the different source regions then? Low emissions have large errors it seems, so more caution must be taken there, no?

p 4405 l 23: Should be Figure A2, not Figure 2. Again - essential Figures need to be in the main text, not the Appendix.

p 4406 5.1: On which inversion setup are these results based? They should be based on the NO-PRIOR setup. If not, please change and (if substantially different) include PRIOR-POS as further “sensitivity”.

p 4406 l 9-11: (very minor comment) Two subsequent sentences that start with “We quantify”. Rephrase.

p 4408 5.2: please rephrase this paragraph. Start out with the fact that Fig. 3 shows the result you will discuss in the following. Then, state the different observations. Finally (something that is currently missing) - discuss possible reasons for these observations. That larger particles need a stronger source because the sink is stronger as well is obvious. Why does the relative uncertainty stay constant? What would be a process / model error that would change this? Is the effect of CCN activation decreasing with particle size because bigger particles are activated anyway (size-dependent activation in SCAV)?

p 4408 5.3: The uncertainty ranges you use to normalize your results are mostly reasoned in a previous section, where you gave sources for e.g. activated fraction in mixed-phase and cold clouds. Only the particle size is given without reference. Here you state now that it can easily be twice as large. This seems much more reasonable than the rather unfounded 1um.
What is the reasoning for a 1um uncertainty? I would expect a reference good enough to counter the reasoning in 5.3 l 20-28. Otherwise, please repeat analysis with at least 2 um uncertainty in

size and rewrite paragraph.

p 4409 first paragraph: Discuss what a negative normalized uncertainty means.

p 4409 13-15: remove Fig. 6, explain in simple terms what the method of Fox does and what exactly is significant. What would be an insignificant interaction? Not much of a correlation?

p 4409 16-21: move this paragraph up, as it apparently belongs to what you discussed before the digression into statistical significance. Fig. 5 can go into the Appendix or be removed.

p 4410 rewrite conclusions depending on the outcome of the changes in previous sections.

Table 2: remove and describe processes in 1 sentence on p 4398, l 10.

Table 3: Aren’t there 4 sensitivity setups, not 3?

Table 4: citation for each uncertainty would be helpful - or reference to section in text where this is discussed.

Figure 1: rephrase caption: “Global mean lifetimes of particles in the BASE model setup as function of aerodynamic diameter, emission ecosystem, and CCN activity. Left panel: CCN-ACTIVE. Middle: CCN-INACTIVE. Right: ratio of CCN-INACTIVE to CCN-ACTIVE lifetimes.” Also, please indicate with points where you actually made simulations, as this is not a continuous function but based on discrete intervals.
Make all three panels the same height. Labels need to have same font size. Use same scale in left and middle plot.

Figure 2: please choose to show either box plots or distributions, but not both. Also, please show NO-PRIOR instead of (or additional to) PRIOR-POS.

Figure 3: y-axis on left plot has no units (absolute uncertainty).

Figure 5: suggest to move into appendix of remove.

Figure 6: remove, discuss briefly in text.

Figures A1 and A2: what is the benefit of showing correlations between ecosystems? Remove or discuss more in text.

Figures A3 / A4: more care should be put into preparing these Figures. Overlapping labels of the x-Axis should be removed, for the NO-PRIOR scenario the position of 0 (deposition <> emissions) should be clearly marked, ticks on y-axis are not labeled anyway so they can be removed, and x-axis titles could be rotated to fit without overlapping.

References

Croft, B., Pierce, J. R., Martin, R. V., Hoose, C., and Lohmann, U.: Uncertainty associated with convective wet removal of entrained aerosols in a global climate model, Atmos. Chem. Phys., 12, 10725-10748, doi:10.5194/acp-12-10725-2012, 2012.

Hoose, C., Lohmann, U., Bennartz, R., Croft, B., and Lesins, G.: Global simulations of aerosol processing in clouds, Atmos. Chem. Phys., 8, 6939-6963, doi:10.5194/acp-8-6939-2008, 2008.

Lelieveld, J., Kunkel, D., and Lawrence, M. G.: Global risk of radioactive fallout after major nuclear reactor accidents, Atmos. Chem. Phys., 12, 4245-4258, doi:10.5194/acp-12-4245-2012, 2012.