Response to Anonymous Referee #1 on “Impact of modelled particle characteristics on emissions inferred by inversion of tracer transport” by S. M. Burrows et al.

We thank the referee for the constructive review. Our detailed responses to the referee's comments follow. We have adopted most of the referee's suggestions, including moving the material from the appendix into the main text and revising the title to better reflect the domain of the study.

General comments

Referee comment (RC) 1: The title could better reflect the study by including the type of particles examined, i.e. biogenic aerosols (or the particle type for which the study is applicable, see also comment 2).

Author response (AR) 1:

Following the referee's suggestion, we will change the title to: “Estimating bacteria emissions from inversion of atmospheric transport: Sensitivity to modelled particle characteristics”

We will also more explicitly state in the abstract that the study has been applied to the case of bacterial aerosol by adding:

“Source estimation via Monte Carlo Markov Chain is applied to a suite of sensitivity simulations and the global mean emissions are estimated for the example problem of bacteria-containing aerosol particles.”

We will also revise the text:

“Uncertainty due to CCN activity or to a 1 μm error in particle size is typically between 10 % and 40 % of the uncertainty due to observation uncertainty”

to:

“For this example, uncertainty due to CCN activity or to a 1 μm error in particle size is typically between ...”

RC 2: The authors introduce this manuscript as a “case study”. However, biogenic aerosols and more specifically, bacteria, is a somewhat special case in that size distribution is largely unknown. To present this as a case study, the authors should explain why bacteria aerosols were chosen? What are the properties of bacteria aerosols compared to other biogenic and non-biogenic aerosols? Furthermore, how applicable are the results of this study, i.e. for bacteria aerosols, to other types of aerosols?

AR 2:

We will add a sentence to the Model Description:

“We assume that to a first approximation, bacteria-containing aerosol particles are transported similarly to other insoluble aerosol particles such as dust, i.e. they have the same size-dependent rate of removal in the model as other particles.”

In the “Discussion and Conclusions”, we will add the following more detailed discussion of the areas in which the results of this study are and are not more broadly applicable to other aerosol types:

“Since transport and removal processes are the basis for the correct simulation of the distributions of any atmospheric aerosol, some of the results of this study also are broadly applicable to other coarse atmosphere aerosols. The dependence of atmospheric residence time (attributable to transport and removal) on size and emission region is a result that holds generally for particles emitted from those regions (fig. XX). The important contribution of uncertainty in particle size to uncertainties in simulated particle residence time will also be applicable for other coarse aerosols, particularly as size approaches 10 µm (fig. YY). The activation of particles as CCN is better constrained for many other atmospheric particle types than for bacterial aerosol, so the contribution of CCN uncertainty may be reduced in other cases. For fine (submicron) aerosol particles, the relative contributions of different uncertainty types are likely to differ from the results presented here, as the efficiency of removal processes changes with size.”

RC 3: The authors define particle size as a model error, however, particle size could also be considered to be an observation error, since observations of particle size are completely lacking. The authors should mention this as it is relevant when addressing the question of how the source estimates may be improved, which is the central motivation for this study.

AR 3: Observations of particle size are indeed quite limited, but some information is available. We treat this as a model error, because the particle size is a model parameter that is fixed at an assumed value in each realization of the model. However, we appreciate the referee's point that size could also be considered an observational error. This is already mentioned in the last paragraph of the Discussion and Conclusions. We will emphasize this more by adding the following sentence in the Discussion and Conclusions: “The uncertainty in particle size could also be reduced by further observations of the size of bacteria-carrying particles.”

RC 4: Why were the particle properties of CCN activity and ice nuclear scavenging chosen for the sensitivity studies? Could the authors please add some justification for this choice.

AR 4: We will add the following text to the methods section to explain better our choice of parameters:

“We focus here on uncertainties related to physical and chemical properties of the transported particles, i.e. particle size, CCN activity and IN activity.”

We agree with the referee that many other important sources of uncertainty exist, including many that are unrelated to particle characteristics. Some of these arise from model numerics and resolution, the formulations of parameterizations, etc.

We will add text to the discussion section that points out some additional expected sources of model error:
“In this study, we focused on the impact of model parameters describing particle characteristics that affect simulated aerosol removal processes, within the context of a particular realization of a global chemistry-climate model. However, tracer transport in global chemistry-climate models can also be sensitive to many other aspects of the model that are beyond the scope of this study, such as model resolution, the use of prescribed meteorology (especially for stratosphere-troposphere exchange and exchange of stratospheric tracers between hemispheres) and tracer lifetime (Aghedo et al., 2010), numerical formulations of atmospheric dynamics (Rasch et al., 2006), and the formulation of the parameterization of deep convective transport (Mahowald et al., 1995; Tost et al., 2006; Lawrence and Salzmann, 2008). The sensitivity of modeled transport to particle characteristics as simulated here could potentially change if these or other aspects of the model were changed.”

RC 5: The explanation on how the ensemble and posterior error distribution are calculated, which is described in Appendix 3, should go into the main text, e.g. in section 4.2.5.

AR 5:

Following the suggestion of the referees, we have moved the material from the appendix (including the description of how the ensemble and posterior distribution are calculated) into the main text.

RC 6: There is no discussion of the posterior emission estimates found for each ecosystem. Furthermore, figures A3 and A4, which show the posterior probability distributions for each ecosystem, are not discussed at all the text. The authors should add some description of these results to the main text.

AR 6:

Following the suggestion of the referees, we have moved the material from the appendix (including figures A3 and A4) into the main text. These figures are already discussed briefly in the ACPD manuscript in Section 4.3.1.

In our revised manuscript, the discussion of these two figures will appear at the beginning of a subsection titled "Mean annual flux estimated per ecosystem":

"The posterior distributions of the estimated fluxes for each particle size and source ecosystem are shown as histograms in Fig. 2 and Fig. 3. In each case, the typical posterior distribution of flux estimates for each ecosystem has an approximately Gaussian shape, which results from the assumption

that the observation uncertainty has a Gaussian distribution. The histograms appear most irregular in the wetlands region, which is also the most pooly constrained by observations (Tab. 1).

As particle size increases, not only do estimated emissions increase estimated emissions for individual ecosystems typically increase as well. This can be seen in the histograms of the ecosystem emission estimates, especially when the emissions in each ecosystem are constrained to be positive (Fig. 2). When emissions are not constrained to be positive, this pattern is less clear (Fig. 3), especially for wetlands and coastal regions. These regions are poorly constrained by the observations due to their relatively small contribution to simulated concentrations: even large changes in the emissions in these regions have only a small influence on the concentrations in other regions, or on global emissions."

Specific comments

RC 7: p4393, l15: "a large fraction", could the authors please provide an estimate of (or range for) this fraction.

AR 7: We will add the following text: "For example, observations of particles with radius greater than 0.2 μm between 2000 and 2008 in Mainz, Germany found that between 5% and 50% of particle volume was composed of primary biological aerosol particles; at Lake Baikal, Russia, an average of ca. 0% of particles were observed to be PBAP (by number and volume; particles with radius > 0.2 μm) (Jaenicke et al., 2007). Measurements in the Amazon rainforest found that 40% of submicron and up to 80% of supermicron particles were primary biological particles (Graham et al., 2003). Although the biological fraction reported depends on the definition and on the measurement technique used, on which there has been little consensus (Després et al., 2012), it is clear that biological particles can be important contributors to the atmospheric aerosol population, particularly in the supermicron size range."

RC 8: p4393, l25: after "to optimally match observations" add that this is within the range of uncertainties for the observations and prior emission estimates.

AR 8: Here, we had written: "In contrast, inverse modelling approaches use observed concentrations in conjunction with a model of atmospheric transport, and apply mathematical techniques to infer the necessary emissions required for the model to optimally match observations."

We will revise this text to: "In contrast, inverse modelling approaches use observed concentrations in conjunction with a model of atmospheric transport to estimate emissions. This is achieved by applying mathematical techniques to infer the necessary emissions required for the model to optimally match observations, accounting for estimates of the uncertainties in observations, and for prior information about the emission."

RC 9: p4394, l14: specify that this is the transport model

AR 9: We have inserted the word "transport" to specify this.

RC 10: p4401, l16: by "model parameters" do the authors mean the emissions in the each of the 10 ecosystem classes? This should be made clearer.

RC 10: The integration of the material from the appendix into the main text should help make this clearer. We will also replace the phrase "model parameters" with the symbol **m** here, which should be clearer.

RC 11: p4401, l20: if the model underestimates removal, then smaller emissions would be possible in order to explain the observed aerosol concentrations, however, the net emissions in each ecosystem still has to be positive. Unless the authors propose that a given ecosystem could have a net removal of aerosols through dry deposition?

RC 12: section 4.3.1: (see also above comment) the authors should mention the physical meaning of the negative emission estimates in the test NO-PRIOR, i.e. net removal of aerosols from the atmosphere. The negative emissions may also only be due to the fact that the variables are poorly constrained, and the strong negative correlations between variables would suggest this.

AR 11,12: Indeed, we fully agree with the referee on this point. The inferred total global flux will always be positive, but not necessarily in each ecosystem. If the model underestimates removal in one ecosystem by an amount that exceeds emissions in that ecosystem, then a negative inferred flux might be required to match observations. The referee correctly notes that this would mean that a given ecosystem could have a net removal of aerosols, through the combined effects of dry and wet deposition.

To explain this better in the manuscript, we will revise the text in the original section 4.3.1 to include the following:

"In NO-PRIOR, negative fluxes are allowed, and in some regions the most likely estimate of the flux is negative. Negative fluxes can occur for statistical or physical reasons. In many atmospheric tracer inversions (Gurney et al., 2002) the data are insufficient to constrain each emission individually. Posterior flux estimates are characterized by dipoles where the combination is well-constrained but fluxes from individual regions can take on large values of opposite sign. Physically, negative emissions imply a misspecification of deposition processes: The model cannot, with the deposition rates assumed in the transport matrix, simulate the concentration gradients observed between ecosystems.

In the NO-PRIOR case, flux estimates in different regions are highly cross-correlated (fig. ##). These high correlations suggest that the negative inferred emissions in NO-PRIOR may be statistical and can be better explained by the weakness of the observational constraints on the inversion than by an underestimate of emissions.

For this reason, we introduced a positive prior constraint on each inferred flux (PRIOR-POS). In

PRIOR-POS, the typical posterior distribution has the shape of a truncated Gaussian distribution. This is because negative fluxes are disallowed, and the correlations between flux estimates in the different regions become very small: the additional constraint has the effect of somewhat decoupling the emissions from different regions (fig. ##). We will focus on the results of the better-constrained PRIOR-POS inversion in the remainder of this paper."

We will also add additional text to the discussion and conclusions:

"We performed both an unconstrained inversion (NO-PRIOR) and an inversion constrained by a prior assumption that fluxes must be positive (PRIOR-POS). The NO-PRIOR inversion leaves open the possibility of an underestimate of deposition that can result in a negative inferred flux. However, the high correlations between the inferred fluxes in different ecosystems indicate that negative inferred fluxes can be better explained by the problem being poorly constrained."

RC 13: p4404, point 1: In fig. 2 the distributions are distributed so that there are fewer high values than low values, the distribution is skewed but in the opposite direction to what the authors state.

AR 13: Our original text is correct. It is possible that the referee has been misled by the unaccustomed presentation of the histograms in fig. 2, which are rotated by 90 degrees. The boxplots in the upper panel clearly also show that there are more high outliers than low outliers.

RC 14: p4404, point 2: perhaps make this point clearer by adding that since only one observation is given for each aerosol source type independently of particle size, and since small particles have longer residence times, less emissions of small particles are required to match the observed concentrations compared to large particles, which have shorter residence times.

AR 14: Following the referee's suggestion, we will revise this point to include the following text: "Estimated emissions increase with increasing particle size. Since the observed concentrations are defined independently of particle size, and since smaller particles have longer residence times, lower emissions of small particles are required to match the observed number concentrations compared to large particles, which have shorter residence times."

RC 15: p4408, l27: could the authors clarify how the uncertainty in emissions due to particle size in fig. 4 is calculated? Is this the normalized uncertainty considering all particle sizes (1 to 10 microns)?

AR 15: Yes, this is the normalized uncertainty considering all particle sizes, but it has been calculated by comparing pairs of inversions that differ in particle size by 1 micron, with all other parameters held constant. The model uncertainty is then calculated according to Eq. 6 (of the ACPD manuscript), with the "parameter uncertainty" equal to 2 microns (+- 1 micron). This is discussed in the section "Definition and calculation of normalized model uncertainty", p. 4407, lines 18-20.

RC 16: p4408, l27: Why was the 1 micron uncertainty chosen and not 2 microns? How would the uncertainty in emissions due to particle size increase if a range of 2 microns were used – would this double the uncertainty contribution?

AR 16: We agree that this was not very well explained in the manuscript and we will add text to explain our reasoning, which we feel does justify the ±1 μm range. An argument could also be made in favor of a range of ±2 μm, which would approximately double the uncertainty contribution from particle size.

In the section "normalized model uncertainty", we will add the following text:

"Limited observations of the size range of bacteria containing particles indicate that they are in the size range of about 1 – 5 µm, with smaller particles observed at a coastal site (count median diameter 2.4 µm and 95% confidence interval 3.1 – 1.6 µm) and larger particles observed at inland sites (count median diameters near 4 µm and 95% confidence intervals ranging from 3.2 to 5.0 µm) (e.g., Shaffer and Lighthart, 1997, see also Tong and Lighthart, 2000; Wang et al., 2007). We therefore consider a uncertainty range of ±1 µm to represent the approximate range of size uncertainty per source."

In the discussion, we will add:

"As discussed in Sect. 5.5, the few measurements of the size of bacteria-containing particles that are available suggest that particles bearing culturable bacteria have diameters in the range of about 1–5 µm, with ranges of ca. ±1 µm from the median at each individual measurement location (e.g. Shaffer and Lighthart, 1997; Lighthart, 2000; Tong and Lighthart, 2000; Wang et al., 2007), so the uncertainty range in particle size could plausibly be considered to be as large as ±2 µm, rather than the ±1 µm used in this study. Doubling the uncertainty in particle size would approximately double its contribution to the model parameter uncertainty in the inversion."

Technical comments

RC 17: p4395, l23: "such as CO2"

AR 17: Thanks, we have fixed this.

RC 18: p4396, l2: replace "lumped eco-systems" with e.g. "eco-system classes"

AR 18: Thanks, we have fixed this.

RC 19: p4397, l2: replace "yr" with "years"

AR 19: Thanks, we have fixed this.

RC 20: p4398, l20: "releases"

AR 20: The original text is correct ("... cloud droplets, which upon evaporation release ...")

RC 21: p4404, l4: the correct term for a "cut-off" Gaussian distribution is "truncated Gaussian"

AR 21: We have changed this.

RC 22: fig. 3: left plot add units to y-axis

AR 22: Thanks, we have fixed this.