Response to Anonymous Referee #2 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. We have adopted most of the referee's suggestions, including making improvements to the structure of the manuscript that will make it easier for readers to follow, and making improvements to some of the figures. Our detailed responses to the referee's comments follow.

Main comments

Referee comment (RC) 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.

Author response (AR) 1: Following the referee's suggestion, we have incorporated the remaining information from the appendices into the main text, and removed A1 and A2.

RC 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.

AR 2: We have made the requested changes. The number of simulations between 1μm and 10μm particles is in fact ten, (in 1μm increments), we will state this more clearly in the section describing the simulations. The paragraph “Sensitivity of global and regional emissions to particles size” has been moved to the end of the old section 4.3.1, rather than the beginning of 4.3.2 as the referee suggested. We also move the subsection “Partitioning of uncertainty in total global emissions” so that it immediately follows the subsection “Global annual mass emissions”. In addition, we changed the title of the Section called "Source inversion by Monte Carlo Methods" to "Source inversion and error analysis", and moved the description of the nomalized uncertainty calculation into a subsection at the end of this section called "Definition of normalized model uncertainty". We have retitled the related subsection in the "Results" section from "Normalized model uncertainty results" to simply "Normalized model uncertainty."

RC 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.

AR 3: We agree with the referee that, in general, a negative inferred emission can indicate a missing sink. More precisely, a negative inferred emission can indicate that the loss processes in that region are underestimated by an amount that exceeds the source, after averaging across the ecosystem and over time.

Negative inferred fluxes can also arise in the inversion for non-physical reasons, i.e. when an overestimation of the source in one region is compensated by a negative estimate in another region. The high cross-correlations between flux estimates in different regions that arise in the NO-PRIOR case (fig. A1) demonstrate that precisely such a mechanism is in fact occurring. Emissions in deserts, for example, are almost precisely anti-correlated with emissions in shrubs (correlation coefficient of -0.99).

This is most likely caused by the atmospheric mixing of these signals so that the observing network cannot separate them. There are two possible ways to proceed from here. The first is to apply post hoc considerations of "reasonableness" to the results. This includes consideration of the posterior statistics such as the correlation analysis above. The other is the inclusion of prior information.

Because of the high correlations, we are not confident that the negative emissions have physical meaning. Instead, they reflect the fact that the problem is poorly constrained. To better constrain the problem, we have introduced prior information: our judgment that the inferred fluxes should be positive. We chose to present some results both with and without the positive constraint in order to give the reader a better understanding of how this assumption affects the outcome, but we felt that the results with the positive constraint were a better estimate of the likely real-world fluxes.

We will better explain this in the manuscript, by adding text to Section 4.3.1 (which discusses the histograms of emission estimates and the correlations between regions):

“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).”

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 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.

AR 4: To address the referee's concerns, we will replace the text:

“The validity of the EMAC model for studies of large-scale aerosol transport and deposition has been established by comparison of simulated and observed deposition patterns of radioactive particles following the Chernobyl nuclear meltdown (Lelieveld et al., 2012).”

with the following text:

“EMAC simulates a realistic climate similar to that produced by other global chemistry-climate models (Lamarque, et al., 2013), and the climate is similar in free-running simulations and simulations nudged with meteorological data (Klinger, 2011).

The EMAC model, in a similar configuration to the one used here, has been shown to produce satisfactory simulations of the observed deposition patterns of radioactive particles following the Chernobyl nuclear meltdown (Lelieveld et al., 2012).

In somewhat different configurations (e.g. including simulation of aerosol microphysics), the EMAC model has also been shown to be capable of producing satisfactory simulations of the atmospheric dust life cycle (Gläser et al., 2012; Astitha et al., 2012), of dust and black carbon simultaneously (Aquila et al., 2011), and of multiple interacting natural and anthropogenic aerosol species (Pringle et al., 2010). The ability of the model to predict aerosol optical depth (AOD) is has been evaluated in detail for the Mediterranean region (de Meij et al., 2012) and globally (Pozzer et al., 2012)."

In addition, 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: 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.

AR 5: 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 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.

AR 6: The sizes of the model uncertainties are a function of various factors that would change for a different aerosol source. One of these is the geographic and temporal distribution of the emissions. For example, the transport of particles emitted in tropical desert regions will be less sensitive to wet removal efficiencies than the transport of particles emitted from rainforests or in the ocean's storm tracks, where precipitation is more frequent. Another factor is the size of the particles in question: atmospheric residence time varies non-linearly as a function of particle size, so the sensitivity of removal rates to changes in particle size is itself a function of particle size. (Kunkel et al. 2012 also sheds light on both of these issues)

Furthermore, the size of the model uncertainty depends on the uncertainty range for the parameter in question, which may also vary between classes of particles. For example, perhaps the CCN efficiency of sulfate aerosol is comparatively well-known, while the CCN efficiency of black carbon aerosol is much less well-quantified.

Specific comments

RC 7: 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:"

AR 7: Thanks, we changed this following the referee's suggestion.

RC 8: p 4397 l 2: simulated yr -> simulated years

AR 8: fixed this.

RC 9: p 4397 l 8: why "nearly all"? Explain (in the manuscript)

AR 9: In the revised text, we have removed this phrase, and simply explain that "CCN-ACTIVE particles are additionally removed by cloud droplet nucleation on the particles and subsequent precipitation." The reason for the original wording was that the size-dependent parameterization of nucleation scavenging efficiency asymptotically approaches 1 (100% nucleation) with increasing particle size (Tost et al, 2006), such that at this size, it is effectively (but not quite) unity.

RC 10: 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.

AR 10: We have done this.

RC 11: 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.

AR 11: The referee is correct that this was not considered in our simulations. We will add this text to the discussion section:

“We did not consider changes in the aerosol size distribution due to aerosol microphysics and/or cloud processing, which could introduce additional uncertainty into the aerosol transport and residence times.”

RC 12: p 4398 l 7-8: “is treated as follows. For” -> “is treated as follows: for”

AR 12: We have corrected this, following the referee's suggestion.

RC 13: p 4399 l 1-8: modeling description part that would go into new simulations section.

AR 13: Done.

RC 14: p 4399 l 9-13: results part into new results section.

AR 14: We think that this is part of the description of simulations and have integrated it into the new simulations section.

RC 15: p 4399 3.4: part of results section.

AR 15: We have moved this to the results section, following the referee's suggestion.

RC 16: p 4402 l 10: some symbols (Theta, D, N) are not explained. Will be resolved once you include the appendices.

AR 16: Following the referee's suggestion, we have integrated the appendices into the main text, resolving this issue.

RC 17: 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.

AR 17: Following the referee's suggestion, we have added this text and a reference to the section with more complete information: “For each realization of **G**, we performed an MCMC inversion with one million trial solutions, which were either accepted or rejected as members of a posterior solution ensemble following the Metropolis rule (Section ##).”

RC 18: p 4403, 16: more robust indication than what? Median? Mean?

AR 18: Yes, more robust than the median or mean. We will indicate this in the text.

RC 19: 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.

AR 19: We have moved these figures into the main text and reordered them.

RC 20: 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?

AR 20: A measure for the reliability of the estimates for the different source regions could be obtained from the spread of the ensemble for each region, but we focus mainly on the global flux, since it is less uncertain. We did not show these results in the paper, but we found that the relative uncertainty in the total global flux is smaller than the relative uncertainties in the individual ecosystems. This is to be expected, since changes in the emissions in different regions can compensate each other.

In the revised text, we will point out that this is a common occurrence in such inversions: “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.”

For this reason, we chose to focus most of our presentation on the global flux, rather than the regional flux.

RC 21: p 4405 l 23: Should be figure A2, not figure 2. Again - essential figures need to be in the main text, not the Appendix.

AR 21: We have fixed this.

RC 22: 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”.

AR 22: These results are based on the PRIOR-POS setup. As explained above (Author Response 3), we believe the PRIOR-POS inversion setup is the more appropriate focus for the results of the paper. We will add a clarifying sentence in Section 4.3.1 following the discussion of the NO-PRIOR and PRIOR-POS results: “We will focus on the results of the better-constrained PRIOR-POS inversion in the remainder of this paper.”

RC 23: p 4406 l 9-11: (very minor comment) Two subsequent sentences that start with “We quantify”. Rephrase.

AR 23: Following the referee's suggestion, we have revised this paragraph to avoid the repetition.

RC 24: 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)?

AR 24:

The uncertainty attributable to CCN activation remains constant across the size range in absolute terms, it only decreases relative to the median global flux estimate as the particle size increases. The total relative uncertainty remains constant because the decrease in relative uncertainty due to CCN activity happens to be roughly compensated by an increase in CCN activity due to particle size. This is a coincidental result, which might be change with a different dependence of the various loss processes on particle size.

We have revised this section following the referee's suggestion. The revised section reads:

“In fig. ##, we show the magnitude of the uncertainties arising from each of the sources considered: observational uncertainty, particle size, CCN activity, mixed-phase scavenging and cold ice scavenging. Overall, the estimated total global mass emissions increase with increasing particle size, due to the shorter particle residence time in the atmosphere. In parallel, the overall uncertainty in the global emissions increases in absolute terms but remains near 150% of the median global flux estimate.

The uncertainty contribution from CCN activity is approximately constant in absolute terms across the size range of interest. This is expected because particle wet removal as parameterized in SCAV is effectively independent of particle size in this size range.

The uncertainty contribution from CCN activity decreases relative to particle size as particle size increases and median global emissions increase. Because this decrease is compensated by a growth in the relative uncertainty contribution from particle size, the total relative uncertainty remains approximately constant. The contributions from ice scavenging parameters are comparatively small, although mixed-phase ice scavenging contributes more uncertainty than particle size for particle diameters from 1—4 µm."

RC 25: 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.

AR 25:

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."

RC 26: p 4409 first paragraph: Discuss what a negative normalized uncertainty means.

AR 26: We will add this text: "The normalized model uncertainty can have a negative value; this indicates that the median of the distribution has shifted in the opposite direction from the expected one, which can occur as the result of randomness in the Monte Carlo method."

RC 27: 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?

AR 27: We think figure 6 quickly conveys information about the magnitude and statistical significance of the marginal relationships between the model particle characteristics and the estimated model parameter uncertainty. In order to more clearly communicate the method used to generate the figure and the main points it illustrates, we will expand the caption and revised the text in the section "Normalized model uncertainty results".

The new caption reads (using old figure number):

“Fig. 6. Illustration of the effects of the marginal effects of model parameters on the model uncertainty, shown in linear effects plots following Fox (1987, 2003). Each plot shows the values predicted by a generalized linear model across the range of values of a main predictor variable, while other predictor variables are held constant at an average value. Top: effect of particle size on CCN uncertainty, controlling for IN sensitivity case. Middle: effect of CCN activity on size uncertainty, controlling for IN sensitivity case. Bottom: effect of sensitivity case on CCN uncertainty, controlling for CCN activity and size. Red dashed lines indicate the 95 %-ile confidence interval.”

The revised paragraphs in the main text read (using old figure numbers):

“The sensitivity to the CCN activity of the particles is high for particles of 1 μm diameter, decreasing to only moderate sensitivity for particles around 10 μm diameter (fig. 5). The sensitivity to particle size is small to moderate for particles around 1 μm diameter, and increases to a large sensitivity for particles around 10 μm diameter. The sensitivity to particle size is higher for CCN-INACTIVE particles than for CCN-ACTIVE particles, particularly at particle sizes closer to 1 μm (fig. 5).

All effects shown in fig. 5 and discussed above are statistically significant by the Student’s t-test ($p < 0.01$). Other significant effects include: a reduction in the normalized uncertainty from CCN activity in the LIM MIXED simulation (after controlling for the effect of particle size; $p < 0.01$), and an increase in the normalized uncertainty from particle size as particle size increases ($p < 0.01$). Three of the statistically significant interactions between model parameters and the normalized model uncertainties are illustrated in the linear effect diagrams shown in fig. 6. Each diagram shows the marginal effect of one of the three predictor variables (particle size, CCN activity and IN sensitivity case), while the others are held constant (Fox, 1987, 2003).”

RC 28: 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.

AR 28: Thanks, we have moved the paragraph as the referee suggests. However, we would like to keep figure 5, which shows the interactions between the different factors in affecting the model uncertainty. We think it is useful to point these out. We have expanded the discussion of figures 5 and 6 in the text, see AR 27.

RC 29: p 4410 rewrite conclusions depending on the outcome of the changes in previous sections.

AR 29: We believe the referee is referring to the focus on PRIOR-POS results and the use of +- 1 μm as size uncertainty. We have now provided further explanation and justification for these choices (see AR 3, AR 25, AR 32), and we hope the referee will agree that these are reasonable choices. Therefore, there is no need to change the conclusions.

RC 30: Table 2: remove and describe processes in 1 sentence on p 4398, l 10.

AR 30: Following the referee's suggestion, we have removed this table and described the processes briefly in the text in description of the simulations.

RC 31: Table 3: Aren’t there 4 sensitivity setups, not 3?

AR 31: Thanks, we've fixed this.

RC 32: Table 4: citation for each uncertainty would be helpful - or reference to section in text where this is discussed.

AR 32: We have chosen to address this comment by adding a few sentences in the text explaining the reasoning for choosing each range of variables, with citations and references as appropriate. The following sentences will appear where the table is first referenced in the main text.

“The values used for the parameter uncertainty ranges and the observation uncertainty are summarized in Table XX. We define the observation uncertainty as the spread in the posterior distribution, given by the middle 90 % range. The denominator in Eq. 18 is the average of the spreads in the two sensitivity cases. The model parameter uncertainties were chosen to approximately span the range of possible values. 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. Because it is unclear to what extent bacteria are CCN-ACTIVE in the atmosphere, we consider the limiting cases: particles that are completely inactive and particles that are always active as CCN. The ice scavenging parameters span the ranges of values observed for cold and mixed-phase clouds (Section 3.4, Henning et al., 2004)."

RC 33: 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.

AR 33: We have rephrased the caption following the referee's suggestion, except that we write "residence times" instead of "lifetimes". We have made the requested corrections to the figure panels, and we have also changed the color palette to one that is designed for accessibility to color-blind readers. For better legibility, we have increased the size of the axis labels and made the lines thicker. We also edited the figure caption to note that these are values for 1 µm particles, and that "points indicate the locations of the simulations performed."

RC 34: 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.

AR 34: We have produced a new version of the figure that includes only the histograms, however, we have kept the focus on the PRIOR-POS results. Our reasoning for choosing to focus on the PRIOR-POS case is explained in AR 3.

RC 35: figure 3: y-axis on left plot has no units (absolute uncertainty).

AR 35: Thanks, we have fixed this.

RC 36: figure 5: suggest to move into appendix of remove.

AR 36: We would like to keep figure 5, which shows the interactions between the different factors in affecting the model uncertainty. We think it is useful to point these out. We have expanded the discussion of figures 5 and 6 in the text, see AR 27.

RC 37: figure 6: remove, discuss briefly in text.

AR 37: See AR 27.

RC 38: figures A1 and A2: what is the benefit of showing correlations between ecosystems? Remove or discuss more in text.

AR 38: This has been addressed in Author Response 3.

RC 39: 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.

AR 39: We have made the suggested improvements to the figure. In the NO-PRIOR case, the position of zero flux is marked by a vertical dashed line.

In addition to the improvements to the figures suggested by the referee, we have eliminated unnecessary white space around several of the figures, and we have combined the two correlation diagrams into two panels of a single figure.

References

Aghedo, A. M., Rast, S., and Schultz, M. G.: Sensitivity of tracer transport to model resolution, prescribed meteorology and tracer lifetime in the general circulation model ECHAM5, Atmospheric Chemistry and Physics, 10, 3385–3396, doi:http://dx.doi.org/10.5194/acp-10-3385-201010.5194/acp-10-3385-2010, http://www.atmos-chem-phys.net/10/3385/2010/, 2010.

Astitha, M., Lelieveld, J., Kader, M. A., Pozzer, A., and de Meij, A.: Parameterization of dust emissions in the global atmospheric chemistry-climate model EMAC: impact of nudging and soil properties, Atmos. Chem. Phys., 12, 11 057–11 083, 2012.

Aquila, V., Hendricks, J., Lauer, A., Riemer, N., Vogel, H., Baumgardner, D., Minikin, A., Petzold, A., Schwarz, J. P., Spackman, J. R., Weinzierl, B., Righi, M., and Dall'Amico, M.: MADE-in: a new aerosol microphysics submodel for global simulation of insoluble particles and their mixing state, Geosci. Mod. Dev., 4, 325–355, doi:http://dx.doi.org/10.5194/gmd-4-325-201110.5194/gmd-4-325-2011, http://www.geosci-model-dev.net/4/325/2011/, 2011.

de Meij, A., Pozzer, A., Pringle, K. J., Tost, H., and Lelieveld, J.: EMAC model evaluation and analysis of atmospheric aerosol properties and distribution with a focus on the Mediterranean region, Atmos. Res., 2012.

Gläser, G., Kerkweg, A., and Wernli, H.: The Mineral Dust Cycle in EMAC 2.40: sensitivity to the spectral resolution and the dust emission scheme, Atmos. Chem. Phys., 12, 1611–1627, doi:http://dx.doi.org/10.5194/acp-12-1611-201210.5194/acp-12-1611-2012, http://www.atmos-chem-phys.net/12/1611/2012/, 2012.

Klinger, C.: Quantitative evaluation of ozone and selected climate parameters in the chemistry-climate model EMAC, Ph.D. Thesis, Master thesis, Ludwig Maximilian University (LMU), Munich, Germany, 2011.

Kunkel, D., M. G. Lawrence, H. Tost, A. Kerkweg, P. Jöckel, and S. Borrmann (2012), Urban emission hot spots as sources for remote aerosol deposition, Geophys. Res. Lett., 39, L01808, doi:10.1029/2011GL049634.

Lawrence, M. G. and Salzmann, M.: On interpreting studies of tracer transport by deep cumulus convection and its effects on atmospheric chemistry, Atmos. Chem. Phys., 8, 6037–6050, 2008.

Lamarque, J.-F., Shindell, D. T., Josse, B., Young, P. J., Cionni, I., Eyring, V., Bergmann, D., Cameron-Smith, P., Collins, W. J., Doherty, R., Dalsoren, S., Faluvegi, G., Folberth, G., Ghan, S. J., Horowitz, L. W., Lee, Y. H., MacKenzie, I. A., Nagashima, T., Naik, V., Plummer, D., Righi, M., Rumbold, S. T., Schulz, M., Skeie, R. B., Stevenson, D. S., Strode, S., Sudo, K., Szopa, S., Voulgarakis, A., and Zeng, G.: The Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP): overview and description of models, simulations and climate diagnostics, Geosci. Mod. Dev., 6, 179–206, doi:http://dx.doi.org/10.5194/gmd-6-179-201310.5194/gmd-6-179-2013, http://www.geosci-model-dev.net/6/179/2013/, 2013.

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:http://dx.doi.org/10.5194/acp-12-4245-201210.5194/acp-12-4245-2012, http://www.atmos-chem-phys.net/12/4245/2012/, 2012.

Mahowald, N. M., Rasch, P. J., and Prinn, R. G.: Cumulus parameterizations in chemical transport models, J. Geophys. Res.-Atmos., 100, 26 173–26, 1995.

Pringle, K. J., Tost, H., Message, S., Steil, B., Giannadaki, D., Nenes, A., Fountoukis, C., Stier, P., Vignati, E., and Lelieveld, J.: Description and evaluation of GMXe: a new aerosol submodel for global simulations (v1), Geoscientific Model Development, 3, 391–412, doi:http://dx.doi.org/10.5194/gmd-3-391-201010.5194/gmd-3-391-2010, http://www.geosci-model-dev.net/3/391/2010/, 2010.

Rasch, P. J., Coleman, D. B., Mahowald, N., Williamson, D. L., Lin, S.-J., Boville, B. A., and Hess, P.: Characteristics of atmospheric transport using three numerical formulations for atmospheric dynamics in a single GCM framework, Journal of Climate, 19, 2243–2266, 2006.

Tost, H., Jöckel, P., and Lelieveld, J.: Influence of different convection parameterisations in a GCM, Atmos. Chem. Phys., 6, 5475–5493, doi:http://dx.doi.org/10.5194/acp-6-5475-200610.5194/acp-6-5475-2006, http://www.atmos-chem-phys.net/6/5475/2006/, 2006.