Review of Burrows et al. (2013), Impact of modelled particle characteristics on emissions inferred by inversion of tracer transport

Synopsis

This manuscript presents an inversion method for the estimation of aerosol sources using atmospheric observations and an atmospheric transport model and examines the contribution of uncertainties in the modelled particle properties to the uncertainty in the source estimates. In particular, the authors examine the influences of uncertainties in CCN activity, ice nucleation scavenging and particle size, and compare these to the influence of observational uncertainty. This study looks specifically at aerosols of biogenic origin, namely bacteria, which comprise a significant part of the total atmospheric aerosol load but for which the sources are poorly known. Overall, the methods used are sound and the manuscript is well written. However, there are parts of the methods and results that should be expanded and/or clarified. Therefore, I recommend this manuscript for publication after moderate revisions.

General comments

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).

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?

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.

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.

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.

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.

Specific comments

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

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

p4394, l14: specify that this is the transport model

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

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?

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.

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.

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.

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)?

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?

Technical comments

p4395, l23: “such as CO_2”

p4396, l2: replace “lumped eco-systems” with e.g. “eco-system classes”

p4397, l2: replace “yr” with “years”

p4398, l20: “releases”

p4404, l4: the correct term for a “cut-off” Gaussian distribution is “truncated Gaussian”

Fig. 3: left plot add units to y-axis