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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Atmospheric aerosols in the earth system: a review of interactions and feedbacks" *by* K. S. Carslaw et al.

Anonymous Referee #1

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This paper is a timely overview of the links between aerosols, atmospheric chemistry and climate change science. Much of the science is very well dealt with, with up-to-date references, and a balanced discussion of conflicting studies. Some aspects receive too little attention though, and some repetition could be avoided. After modification to reflect the following comments, it should make a good contribution to the literature.

A general comment is that the paper is thin on discussions of the nitrogen cycle, giving most attention to organic aerosols and DMS. As a first example, Figure 1 shows contributions of biogenic gases and carbonaceous aerosol to the atmospheric aerosol, but no anthropogenic sulphates or nitrates. These fall out of the sky as acid deposition, but their source is not indicated. Nitrates have long been neglected in global modelling studies, but awareness is increasing of their important role as both aerosols and in

terms of their interactions with ecosystems. These topics are touched upon briefly, but since the topic of the paper is "aerosols", the nitrogen story should be given more weight. (The organic story is rather over-represented here). More comments on this below.

The Tables and figures provided are also rather qualitative. This review covers many subjects, and it is difficult for the reader to keep the various components and arguments in perspective. It would be good to use Tables to summarise the results presented in the text, even if strong caveats are needed in footnotes or comments. If the authors can provide a more up-to-date emission table than that of Andreae & Rosenfeld (below), then provide this - it would also help with the overview.

A relevent review paper which has appeared recently is Andreae & Rosenfeld (Aerosol-cloud-precipitation interactions. Part 1. The nature and sources of cloud-active aerosols Earth-Science Reviews, 2008, 89, 13-41). This review contains valuable discussions on the important factors affecting aerosol radiative forcing, and on their sources, often going into more detail than the present manuscript can attempt. I suggest that the authors update their manuscript in the light of this paper.

As this article is a reivew, I was also expecting to see more discussion of the issues linking meteorology and aerosols, e.g. as raised in: Rosenfeld, D. et al., Flood or drought: How do aerosols affect precipitation? Science, 2008, 321, 1309-1313.

Specific comments:

p11089, line 14 - the Andreae & Rosenfeld paper has alternative (more up-to-date) emission estimates.

p11089, line 28. The ice-cores have provided evidence of changes in many components, e.g. Legrand et al. (JGR, 2007, D23S11) and Fagerli et al (JGR, 2007, D23S13)

p11093, line 5. Although the authors are in widespread company in quoting the estimate of Guenther et al. to three significant figures, the factors 2-3 uncertainty on this

estimate are worth mentioning.

p11093, line 9. The Goldstein+Galbally estimate was re-done by Hallquist et al.. Although that paper is not formally accepted yet, the referees found no fault with the calculation, and this is an important new estimate which is worth citing.

p11093, line 15. Strictly, the Zhang et al paper deals with OOA rather than SOA.

p11094, line 5. As the Hallquist et al article is still at the ACPD stage, it would be good to give at least a few published references also.

p11094, lines 13-16. The results of models are cited without criticism. In the SOA field models are notoriously unreliable, as the paper of Volkamer (GRL, 2006, L17811) made clear. These uncertainties should be reflected earlier in the discussions (they are briefly mentioned later)

The sections 2.1.2 to 2.1.3.1 cover much the same ground several times, repeating that temperature increases emissions and CO2 may reduce them. These arguments should be condensed into one presentation.

p11096, line 27. Although the paper cited (Bäck and Hari 2008) was not immediately available to me, I find it hard to believe that leaf area can increase by a factor 2 by the year 2100. This cannot be a globally relevant number, but refer to some part of the boreal forest perhaps? The authors should be more explicit (and critical) in what they mean here.

p11099, line 9. Add "At least in smog-chambers"... before this sentence. The jury is still out on the main source of SOA.

p11099, section 2.1.3.2 The estimates of terpene emissions (and indeed isoprene) are still very uncertain. See Arneth et al. (ACP, 2008, p4605-)

p11100-11101. Experimental evidence that changed oxidation can result in changed OC can be found in the ice-core studies of Legrand et al. (JGR, 2007, D23S11).

p11101, lines 12-13. The evidence surrounding the importance of sulphate for SOA is still unclear, so it would be safer to say "may increase" than just "increases".

p11102. Winiwarter et al. (AE, 2009, 43, 1403-) have also provided estimates of PBAP emissions.

p11103. Wildfires generate emissions of many compounds, not just the OC and BC as discussed here. Give the overview.

p11109. Section 2.4.2 - is one example where the N-cycle is discussed in a superficial way (especially compared to all the pages spent on organic aerosols).

Deposition of nitrate particles may lead to increased biomass growth and carbon sequestration in some areas (the amount is hotly disputed), but also to increased emissions of N2O. Even sulphate deposition can have a fertilising effect in some parfts of the globe. If reviewing the impact of aerosols on climate such factors need some attention.

p11110, line 14. The Magnani paper is cited uncritically (and here wrongly). Actually, here the paper is mistakenly cited as Mencuccini et al., but the title given in the reference list has the title and page numbers of the Magnani et al article which caused headlines around the world. This paper suggested that nitrogen deposition could cause a very high sequestration of CO2. This analysis has several severe flaws however, as has been made clear by (among others):

de Vries, W. et al., Ecologically implausible carbon response? Nature, 2008, 451, E1-E3.

and Sutton, M. et al., Uncertainties in the relationship between atmospheric nitrogen deposition and forest carbon sequestration Global Change Biology, 2008, 14, 1-7

(In summary, Magnani found a relation between growth and nitrogen deposition, and ascribed the whole difference to the nitrogen. They didn't account for the fact that the high N areas were also areas conducive to growth for many other reasons. Forests in

Germany do grow better than those in Finland, and nitrogen is only a small part of the reason.)

p11124, section 3.2.1. Dry deposition is also a very hard process to parameterise for sea-areas, and few studies have addressed this.

p11126, line 22. Provide references for the NMHC emissions. (e.g. Guenther et al. 1995 estimated this)

p11135, section 5.2.3. Dust also acts as sink of nitrate, affecting the fine/coarse distribution of these compounds (e.g. Dentener et al. JGR, 1993), and hence the radiative forcing.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 11087, 2009.