

Interactive comment on “Organic aerosol and global climate modelling: a review” by M. Kanakidou et al.

Anonymous Referee #6

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Review of “Organic aerosol and global climate modeling: a review,” by M. Kanakidou et al., submitted to Atmos. Chem. Phys.

This paper reviews the current state of knowledge of issues related to organic aerosols, including measurements, theory, and modeling. The paper is an excellent review and should definitely be published. I would like to make the following suggestions for the authors to consider:

1. Section 2.2.2. “Reported NMVOC emissions have been decreasing.” It would be useful to identify whether certain types of NMVOCs are decreasing more than others. For example, what are the relative rates of decrease of alkanes versus alkenes, aromatics, etc.?
2. Section 2.2.3. It would be useful if organic emission estimates were broken down

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into anthropogenic versus natural. For example, when discussing the relative formation of SOA versus POA in paragraph 25, what fractions of SOA and POA are anthropogenic versus natural?

3. Similarly, in Section 2.2.4, what percent of total SOA precursor emission is natural versus anthropogenic?

4. Section 2.3. “Uncertainties.” It would be better to describe this section as “Uncertainties and Research Needs” or to have separate subsections for each of these two categories.

5. Section 2.3. Are sesquiterpenes the most efficient natural or anthropogenic SOA precursors or just the most efficient natural precursors?

6. Section 3, introduction. “The processes leading to SOA formation” It seems that aerosol microphysics and cloud processing are missing from the chain.

7. Section 3.1.1, “Experimental evidence indicates that high molecular weight compounds containing the -OH functionality are particularly important in this respect.” It may be useful to note, however, that the higher the molecular weight, the greater the Kelvin effect, and the more difficult it is to homogeneously nucleate, so there must be a limit with respect to how high the molecular weight can be.

8. Section 3.2, “The authors argued that ozonolysis is probably the only atmospherically relevant organic source for new particle formation because of the low atmospheric concentrations of these biogenic compounds.” Does this statement apply to natural nucleation only? What about nucleation of anthropogenically-emitted organic gases from combustion sources?

9. Section 3.2., “The absence of nucleation events suggests that nucleation of purely SOA compounds in the atmosphere may not be that frequent.” Might that be due to the fact that most low-vapor-pressure organics have high molecular weight, but high-MW species have a strong Kelvin effect at nucleation sizes?

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10. Section 3.3. Gas-particle partitioning is not a time-dependent physical process in the same manner as is nucleation or condensation or coagulation. Instead, it is a convenient method of estimating the relative concentrations of species in gases and particles. As such, I think it would be more logical if Section 3.3. were moved following Section 4, “Aerosol dynamics”

11. Section 3.3. Please define “organic phase.” Organics can be either liquid or solid or mixed phase or they may be dissolved in aqueous solutions.

12. Section 3.3, page 5890, Question 2. “Is gas-aerosol equilibrium for the condensing organic species established rapidly or are the kinetics of condensation/evaporation important?” This question is raised before condensation is discussed (Section 4.2), which is another reason why Section 3.3 should follow Section 4.

13. Section 3.3, p. 5890. “Table 5 presents a summary of recent global modeling studies that include SOA and their main assumptions.” A paper by Jacobson (JGR 2004, Vol. 109, doi:10.1029/2004JD004945) states that it uses a “Scheme which is also used to condense organic gases onto size-resolved aerosols. The model treats dissolutional growth of NH₃, HNO₃, HCl, and soluble organics to all size bins” This is a method of solving organic growth that appears to differ from those listed in the table.” According to an earlier paper by Jacobson, he agglomerates the condensable organic gases into one or more groups that condense that are condensed onto or dissolved into particles of different size.

14. Section 4.1. “Based on actual knowledge, in global models organic aerosol mass is assumed to be distributed mainly in the accumulation mode.” This statement might be modified to state, “in most global models” since the paper above appears to be an example of where this is not done.

15. Section 5.2. “The most important process by which organic aerosols are scavenged is formation and removal of cloud condensation nuclei.” This may be true for aerosol mass but not necessarily number. One study found, “washout may be

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a more important in-plus-below-cloud removal mechanism of aerosol number than rainout (the opposite is true for aerosol mass).” (abstract of Jacobson, JGR 108, doi:10.1029/2002JD002691, 2003).

16. Section 5.2. “Capture of aerosols by snow and ice is still poorly described in general circulation models.” This is true, but the method described in the paper given under comment 15, which has been used in a global model (comment 21) differs from the use of scavenging efficiencies.

17. Section 6.1, “Presently, models are applying first order conversion kinetics of hydrophobic to hygroscopic BC and OC₂” This is true for the most part but not so for the study of Jacobson (2001a) (referenced in the current paper) in which the evolution of particle mixing state was calculated with size-resolved microphysical and chemical processes.

18. Section 6.2, “Hence, when the urban plume starts to disperse on regional scale, most of the conversion has already happened.” What is the approximate turnover time in that case?

19. Section 10. Equation 18 is correct only for a single particle. For a population of particles, the efficiency for each particle size and wavelength must be weighted by the extinction coefficient (scattering or absorption) at that size and wavelength, so Equation 18, as written, does not apply to a population of particles.

20. Section 10.1. “Only a single study exists on the complex index of refraction of various organic aerosol components and mixtures at concentrations representative of atmospheric conditions.” The complex refractive index is a property of a compound; thus, it is independent of the concentration of the compound. The sentence above implies that the complex index of refraction of an individual particle varies with the concentration of particles, which does not appear to be physical. Please clarify.

21. Section 12. “Recently, first-principles approaches to predicting cloud droplet

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number have emerged” The studies listed do not appear to treat the size-resolved collision-coalescence process or the ice-crystal process of precipitation formation from aerosol particles. Global studies that treat these processes include Jacobson (107, D19, 4410, doi:10.1029/ 2001JD001376 and the papers described under comments 13 and 15).

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