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Atmospheric Chemistry and Physics Discussions

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Interactive Comment

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

Anonymous Referee #5

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This is a timely review on the state of knowledge on the issue of atmospheric organic aerosol. The review is very comprehensive, up to date and very well written. I especially appreciate that the authors' highlight some of the standing issues in each topic. This will be very helpful for readers of this review, who undoubtedly will have diverse and broad backgrounds. I think that the review fits very well the readership of Atmospheric Chemistry and Physics and should be published following some modification.

My main comment is that the authors do not discuss relevant work that has been conducted in the last several years by groups working on laboratory experiments that characterize the processing of organic aerosol. A review of some of these studies was recently published (Y. Rudich, Chemical Reviews 2003), but additional studies have since been published. Specifically, the reactions of ozone and OH with organic surfaces and aerosols have been thoroughly studied, most notably on oleic acid aerosol particles and multicomponent particles. These studies used well-defined laboratory models to identify the kinetics, reaction mechanisms and implications for the CCN activity of organic aerosols and should be mentioned in this review in the discussions about aging processes and hygroscopicity changes (see partial list of references below). In addition, recent paper by Katrib et al (2004) suggests new reaction pathways for the formation of large molecular weight products in the ozonolysis reactions of oleic acid. This new reaction pathway involves reactions of Criege intermediates in the condensed phase with aldehydes, ketons and alkenes and it does not involve acidity. This is also a topic that is relevant to the current in the HULIS discussion. Some references on the topics include:

Katrib, Y., S.T. Martin, H.M. Hung, Y. Rudich, H.Z. Zhang, J.G. Slowik, P. Davidovits, J.T. Jayne, and D.R. Worsnop, Products and mechanisms of ozone reactions with oleic acid for aerosol particles having core-shell morphologies, J. Phys. Chem. A, 108 (32), 6686-6695, 2004.

Moise, T., and Y. Rudich, Reactive uptake of ozone by aerosol-associated unsaturated fatty acids: Kinetics, mechanism, and products, J. Phys. Chem. A, 106 (27), 6469-6476, 2002.

Morris, J.W., P. Davidovits, J.T. Jayne, J.L. Jimenez, Q. Shi, C.E. Kolb, D.R. Worsnop, W.S. Barney, and G. Cass, Kinetics of submicron oleic acid aerosols with ozone: A novel aerosol mass spectrometric technique, Geophys. Res. Lett., 29 (9), 2002.

Smith, G.D., E. Woods, T. Baer, and R.E. Miller, Aerosol uptake described by numerical solution of the diffusion - Reaction equations in the particle, J. Phys. Chem. A, 107 (45), 9582-9587, 2003.

Smith, G.D., E. Woods, C.L. DeForest, T. Baer, and R.E. Miller, Reactive uptake of ozone by oleic acid aerosol particles: Application of single-particle mass spectrometry to heterogeneous reaction kinetics, J. Phys. Chem. A, 106 (35), 8085-8095, 2002.

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Discussion Paper

Thornberry, T., and J.P.D. Abbatt, Heterogeneous reaction of ozone with liquid unsaturated fatty acids: detailed kinetics and gas-phase product studies, Phys. Chem. Chem. Phys., 6 (1), 84-93, 2004.

de Gouw, J.A., and E.R. Lovejoy, Reactive uptake of ozone by liquid organic compounds, Geophys. Res. Lett., 25 (6), 931-934, 1998.

There is also a paper by Abbatt's group showing activation of oleic and linoleic acid particles to cloud droplets following ozonolysis. The paper, now accepted in JGR-atmosphere is relevant to the review and puts the lab work as an important tool for understanding aging of organic aerosols.

Formation of Cloud Condensation Nuclei by Oxidative Processing: Unsaturated Fatty Acids. Keith E. Broekhuizen, Troy Thornberry, P. Pradeep Kumar, and Jonathan P.D. Abbatt

Other relevant studies focused on reactions of other radicals (such as OH, NO3, Cl and Br) with organic layers that lead to changes in the hydrophobicity of these surfaces (in the same context as the ozone reactions), as measured by contact angle and microbalance methods:

Demou, E., H. Visram, D.J. Donaldson, and P.A. Makar, Uptake of water by organic films: the dependence on the film oxidation state, Atmospheric Environment, 37 (25), 3529-3537, 2003.

Eliason, T.L., S. Aloisio, D.J. Donaldson, D.J. Cziczo, and V. Vaida, Processing of unsaturated organic acid films and aerosols by ozone, Atmospheric Environment, 37 (16), 2207-2219, 2003.

Usher, C.R., A.E. Michel, D. Stec, and V.H. Grassian, Laboratory studies of ozone uptake on processed mineral dust, Atmospheric Environment, 37 (38), 5337-5347, 2003.

Mmereki, B.T., and D.J. Donaldson, Direct observation of the kinetics of an atmospher-

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ically important reaction at the air-aqueous interface, J. Phys. Chem. A, 107 (50), 11038-11042, 2003.

Bertram, A.K., A.V. Ivanov, M. Hunter, L.T. Molina, and M.J. Molina, The reaction probability of OH on organic surfaces of tropospheric interest, J. Phys. Chem. A, 105 (41), 9415-9421, 2001.

Moise, T., and Y. Rudich, Reactive uptake of Cl and Br atoms by organic surfaces - a perspective on the processing of organic aerosols by tropospheric oxidants, Geophys. Res. Lett., 28 (21), 4083-4086, 2001.

Moise, T., R.K. Talukdar, G.J. Frost, R.W. Fox, and Y. Rudich, The reactive uptake of NO3 by liquid and frozen organics, J. Geophys. Res., 107 (D2), art. no. 4014, 2002.

Rudich, Y., I. Benjamin, R. Naaman, E. Thomas, S. Trakhtenberg, and R. Ussyshkin, Wetting of hydrophobic organic surfaces and its implications to organic aerosols in the atmosphere, J. Phys. Chem. A, 104 (22), 5238-5245, 2000.

Moise, T., and Y. Rudich, Reactive uptake of ozone by proxies for organic aerosols: Surface versus bulk processes, J. Geophys. Res.-Atmos., 105, 14667-14676, 2000.

Thomas, E., Y. Rudich, S. Trakhtenberg, and R. Ussyshkin, Water adsorption by hydrophobic organic surfaces: Experimental evidence and implications to the atmospheric properties of organic aerosols, J. Geophys. Res.-Atmos., 104 (D13), 16053-16059, 1999.

Wadia, Y., D.J. Tobias, R. Stafford, and B.J. Finlayson-Pitts, Real-time monitoring of the kinetics and gas-phase products of the reaction of ozone with an unsaturated phospholipid at the air-water interface, Langmuir, 16 (24), 9321-9330, 2000.

Benz, R.W., F. Castro-Roman, B.L. de Groot, D.J. Tobias, and S.H. White, Molecular dynamics (MD) simulations of a DOPC bilayer: Comparison with experimental data, Biophysical Journal, 86 (1), 368A-368A, 2004.

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Vieceli, J., O.L. Ma, and D.J. Tobias, Uptake and collision dynamics of gas phase ozone at unsaturated organic interfaces, J. Phys. Chem. A, 108 (27), 5806-5814, 2004.

Thomas, E.R., G.J. Frost, and Y. Rudich, Reactive uptake of ozone by proxies for organic aerosols: Surface-bound and gas-phase products, J. Geophys. Res.-Atmos., 106 (D3), 3045-3056, 2001.

Poschl, U., T. Letzel, C. Schauer, and R. Niessner, Interaction of ozone and water vapor with spark discharge soot aerosol particles coated with benzo a pyrene: O3 and H2O adsorption, benzo a pyrene degradation, and atmospheric implications, J. Phys. Chem. A, 105 (16), 4029-4041, 2001.

An important topic also uncovered in this review is the role of the organic matrix in sequestering pollutants at the interface. This topic may be less relevant to the climatic issues, but certainly relevant to issues such as health effects and transport of pollutants in the troposphere. Some laboratory studies of this issue have been conducted by Donaldson et al

Mmereki, B.T., S.R. Chaudhuri, and D.J. Donaldson, Enhanced uptake of PAHs by organic-coated aqueous surfaces, J. Phys. Chem. A, 107 (13), 2264-2269, 2003.

Mmereki, B.T., and D.J. Donaldson, Laser induced fluorescence of pyrene at an organic coated air- water interface, Phys. Chem. Chem. Phys., 4 (17), 4186-4191, 2002.

There is also substantial lab work on the thermodynamic properties of organic aerosol. See for example

Bilde, M., and S.N. Pandis, Evaporation rates and vapor pressures of individual aerosol species formed in the atmospheric oxidation of alpha- and beta-pinene, Environ. Sci. Tech., 35 (16), 3344-3349, 2001.

Bilde, M., B. Svenningsson, J. Monster, and T. Rosenorn, Even-odd alternation of evaporation rates and vapor pressures of C3-C9 dicarboxylic acid aerosols, Environ. Sci. Tech., 37 (7), 1371-1378, 2003.

I suggest that the authors will consider to relate to this large body of laboratory studies by mentioning some of these studies and by providing some references for those readers who would like to learn more about the laboratory studies which are every relevant for the topic.

There are addiitonal minor points to be mentioned:

1. Section 3.1.3: Claeys et al revised their initial mechanism about the OH oxidation leading to methylerythriol (atmospheric Environment 2004). This should be mentioned here (it is mentioned somewhere else in the review as well).

2. Section 9.2: it is unknown if the mixtures suggested by Fuzzi et al for presentation of WSOC really represent the chemical reactivity of organic aerosol. They may replicate some physical properties such as surface tension. Russell et al are also working on finding good mixtures that will represent the organics in aerosols. (see

Maria, S.F., L.M. Russell, B.J. Turpin, and R.J. Porcja, FTIR measurements of functional groups and organic mass in aerosol samples over the Caribbean, Atmospheric Environment, 36 (33), 5185-5196, 2002.

Russell, L.M., S.F. Maria, and S.C.B. Myneni, Mapping organic coatings on atmospheric particles, Geophys. Res. Lett., 29 (16), 10.1029/2002GL014874, 2002.

3. Section 11. The word “optic” should probably be “optical”

4. section 13.3. “pre-fired” should probably be “heated”

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 5855, 2004.