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

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Received and published: 1 March 2005

First of all we would like to thank referee #5 for his/her constructive comments/additions to the review.

We have incorporated in the text (page 5906 line 8): “Rudich (2003) has published a comprehensive review of some of the laboratory studies that have been conducted the last years to characterize the chemical processing of organic aerosol. Since then more thorough studies have been performed particularly on the reactions of ozone and OH with organic surfaces and aerosols, 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 (Moise and Rudich, 2002; Smith et al., 2002; 2003; Katrib et al., 2004; Vieceli
et al., 2004; Molina et al., 2004).”

Later on in page 5907 line 12 we added: “Molina et al. (2004) investigated the OH-initiated oxidation of solid organics in the presence of O2, NOx and H2O and detected small gas phase products including CO, CO2, CH2O, CH3CHO, CH3OH, and HCOOH that escape from the aerosol phase by volatilisation.”

Comment on the Katrib et al work has been added in page 5907 line 19.

Comment on Abbatt’s group work that shows activation of oleic and linoleic acid particles to cloud droplets following ozonolysis is added and the paper is referenced.

The following paragraph has been added in page 5907 line 19:

“Katrib et al. (2004) suggest 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 Criegee intermediates in the condensed phase with aldehydes, ketones and alkenes and it does not involve acidity. Kawamura et al. (1995; 1996; 2005) and Narukawa et al. (2003) provide experimental evidence that Br chemistry in the Arctic transforms unsaturated diacids to smaller saturated acids. Ozonolysis of oleic and linoleic acid is shown to contribute to activation of particles to cloud droplets (Broekhuizen et al., 2005). Reactions of OH, NO3, Cl and Br radicals with organic layers have been found to change the hygroscopicity of the surfaces as measured by contact angle and microbalance methods (Demou et al., 2003; Eliason et al., 2003; Rudich et al., 2000; Moise and Rudich, 2001).”

At the end of the same section we refer to the recently published paper by Maria et al. (Science, Dec. 2004) that provides a flavor of the importance of these reactions:

“Recently, Maria et al. (2004) on the basis of observational growth rates of atmospheric particles, evaluated that consideration of such surface- and volume- limited chemical reactions should increase the carbonaceous particle burdens in climate models by 70%.”
The role of the organic matrix in sequestering pollutants at the interface is definitely a very interesting point but not straightforward linked to the focus of the paper.

A paragraph has been added (in page 5884 after line 19) summarizing the work on the thermodynamic properties of organic aerosol: Few experimental values of the vapour pressures of secondary organic aerosol components exist. The vapour pressure of five mono-carboxylic acids (C14-C18) and two di-carboxylic acids (glutaric and adipic acid) were measured by Tao and McMurry (1989) over the temperature range 283-323 K using a Tandem Differential Mobility Analyzer (TDMA) system. Vapor pressures of the mono-carboxylic acids were in the range 3x10^{-7} to 10^{-4} Pa at 296 K and vapor pressures of the di-carboxylic acids were 8x10^{-4} and 10^{-5} Pa for glutaric and adipic acid at 296 K. Heats of formation were in the range 163-191 kJ mol^{-1} for the mono-carboxylic acids and 102 and 117 kJ mol^{-1}, for glutaric and adipic acid, respectively. Additional measurements of the vapour pressures of C3-C9 dicarboxylic acids were reported by Bilde et al. (2003) Hallquist et al. (1997) using a Knudsen effusion apparatus, measured the vapor pressures of two oxidation products of a pinene and D3 carene, namely pinonaldehyde and 2,2-dimethyl-3-(2-oxypropyl)-cyclopropaneacetaldehyde (in the paper referred to as caronaldehyde), respectively. Vapour pressures were measured over the temperature range 255-276 K and the values at 296 K were 4.2 Pa for pinonaldehyde and 2.3 Pa 2,2-dimethyl-3-(2-oxypropyl)cyclopropane-acetaldehyde. Using the Tandem Differential Mobility Analysis (TDMA) technique Bilde and Pandis (2001) measured the vapour pressures of glutaric acid, trans-norpinic acid and pinic acid particles. Additional measurements of the volatility of the various SOA components are needed for a comprehensive description of SOA formation in atmospheric chemical transport models.

References


Bilde, M., B. Svenningsson, J. Monster, and T. Rosenorn, Even-odd alternation of


We followed the recommendation of the referee to mention some of the laboratory studies and the list of references in the reviewer’s comment is also available for further reading.

The Claeys et al (2004b) paper is indeed mentioned a few lines further when the proposed mechanism is discussed. To avoid misunderstanding we removed the ‘OH-initiated’.

The work by Russell et al. (2002) is already referenced together with the Decesari et al (2000) study in page 5931 line 26. For clarity we have added at the end of the section the sentence: “The ultimate aim of these studies is to represent the hygroscopicity, the chemical reactivity and the physical and optical properties of OA with the same model compounds and mixtures.”

Typing errors have been corrected.