

## Uptake of ambient organic gaseous mixtures to laboratory generated aerosols

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The formation of secondary organic aerosols (SOA) in the atmosphere has been an area of significant interest due to its climatic relevance, its effects on air quality and human health. Due largely to the underestimation of SOA by regional and global models, there has been an increasing number of studies focusing on alternate pathways leading to SOA. In this regard, recent work has shown that heterogeneous and liquid phase reactions, often leading to oligomeric material, may be a route to SOA via products of biogenic and anthropogenic origin (Kroll et al., 2005; Kalberer et al., 2004).

Although oligomer formation in chamber studies has been frequently observed, the applicability of these experiments to ambient conditions, and thus the overall importance of oligomerization reactions remain unclear. Specifically, the importance of aerosol acidity, the reversibility of reactions upon neutralization, and the effect of realistic gas phase composition and concentration on the uptake is uncertain and will in part determine the importance to SOA formation.

In the present study, ambient air is drawn into a 2m<sup>3</sup> Teflon smog chamber and exposed to acidic sulfate aerosols, which have been formed in situ via the reaction of SO<sub>3</sub> with water vapor. The aerosol composition is continuously measured with a High Resolution Aerodyne Aerosol Mass Spectrometer (HR-ToF-AMS), and particle size distributions are monitored with a scanning mobility particle sizer (SMPS). The use of ambient air and relatively low inorganic particle loading potentially provides clearer insight into the importance of heterogeneous reactions. Particularly, the effect of aerosol neutralization arising from the ammonia present in the ambient air, and the reversibility of the organic uptake under such circumstances can be studied.

Results of experiments, with a range of sulfate loadings show that there are several competing processes occurring on different timescales. A significant uptake of ambient organic gases to the particles is observed immediately, possibly associated with the condensational uptake of ambient organics. The increase in organic aerosol mass due to this process increases as a function of the original input SO<sub>4</sub> mass (Figure 1A).

On the other hand, during a period of several hours there is a shift in the organic mass spectrum towards fragments greater than 300 amu, indicating that higher molecular weight products (possibly

oligomers) are being formed through a reactive process. This process continues regardless of the fact that the particles are continuously being neutralized.

Measurements with the HR-ToF-AMS during this time also indicate that there is a slow volatilization of organic mass from the aerosols possibly due to the formation of volatile by-products of oligomer forming reactions. This volatilization is primarily associated with hydrocarbon fragments (C<sub>x</sub>H<sub>y</sub>) (Figure 1B), resulting in an overall reduction in the molar ratio of Carbon to Oxygen (C/O).

The results suggest that both condensation and heterogeneous reactions can occur with ambient organic gases, resulting in significant increases in organic mass, which can occur even in the presence of ammonia. This may have significant implications to the ambient atmosphere where particles may be fully neutralized after their formation.

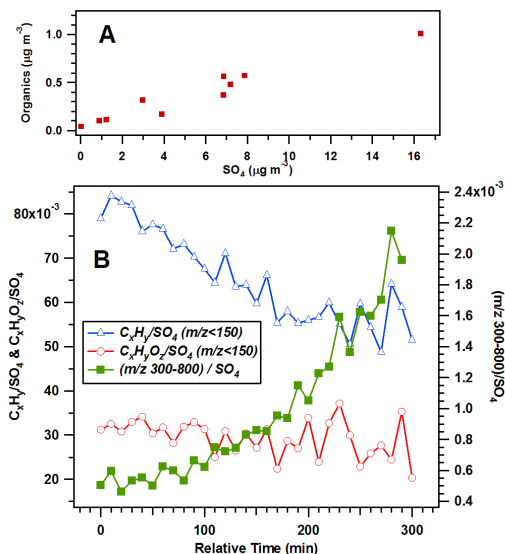


Figure 1. **A.** Immediate organic uptake relative to added sulphate. **B.** Normalized organic fragments as a function of time from sulphate addition.

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