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INSTITUTIONEN FÖR KEMI OCH MOLEKYLÄRBIOLOGI

Thesis for the degree of Doctor of Philosophy in Natural Science, Specialization in Chemistry

Secondary Organic Aerosols:
Composition, Gas-to-Particle Partitioning and Physical Properties

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Abstract

Atmospheric aerosols influence our climate and air quality. Aerosol particles in the atmosphere are transformed through many different physical and chemical reactions. A substantial fraction of the particles in the atmosphere are of secondary origin, formed as a result of gas to particle conversion. The formation process of secondary organic aerosols (SOA) from oxidation of volatile organic compounds (VOC) is currently not fully understood. The objective of this thesis is to contribute to the understanding of factors important for secondary particle formation by simulating certain atmospheric processes in a flow reactor and by measurements of organic compounds in the ambient atmosphere. This work focuses on the formation of secondary organic particles via gas to particle conversion, their chemical composition and the volatility of the compounds. These factors are important for understanding the formation and evolution of secondary particles in the atmosphere, which in turn is important for making predictions about our future climate.

The chemical composition of SOA was studied using a chemical ionization high-resolution time-of-flight mass spectrometer connected to a Filter Inlet for Gases and Aerosols (FIGAERO-ToF-CIMS). The analysis was performed on samples from three sites: a boreal forest in Europe, a temperate forest in North America and a semi-urban location near a major city in Asia.

In order to model SOA and thus be able to predict its impact on society, in particular relating to climate change and health issues, accurate models for SOA formation are needed. The basis for such models includes understanding gas to particle partitioning and the factors that influence this partitioning. In addition, knowledge of the compounds in the particles is needed. The work revealed ways in which anthropogenic pollution could affect the partitioning and consequently the formation of SOA. It was shown that equilibrium phase partitioning behaves as predicted under some circumstances, such as when the air was not affected by anthropogenic pollution. However, when the air masses were affected by anthropogenic pollution, equilibrium phase partitioning does not behave as expected, due to restrictions in uptake and the aerosol not being in equilibrium. This effect was especially seen for highly oxygenated compounds.

Keywords: gas to particle conversion, volatility, secondary organic aerosols, FIGAERO, CIMS, monoterpenes, isoprene, SOA, BVOC.