Aerosol source analysis approach for a rural background site – identifying the chemical fingerprints of anthropogenic and biogenic aerosols

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The aerosols' effects on climate, human health and visibility are dependent not only on their size but to a large extent also by their chemical properties. Particle mass loadings in various size classes are routinely and widely monitored, but less attention is generally paid to the particles' chemistry. However, the developments in measurement instrumentation and analysis methods, in the past ten years or so, have made possible direct online measurement of aerosol chemical composition and a detailed source analysis of the results using factor analysis. In this study an application of the new analysis methods to mass spectrometric measurement datasets from a boreal forest research site is presented.

The SMEAR II atmospheric research site (Hari & Kulmala, 2005) at Hyytiälä, Southern Finland is a well-known example of a rural background measurement station, influenced often by clean air masses from sparsely populated areas of Northern Scandinavia and the Arctic Ocean, but also experiencing pollution episodes when the incoming air masses originate from continental Europe or the industrial regions of Russia. Also aerosols from the local and regional pollution sources, such as sawmills, heating and traffic from nearby large cities are frequently observed. The aerosol loadings and their chemical composition are therefore extremely event-driven and very dependent on wind directions and trajectories. From the perspective of characterizing the various aerosol types, the SMEAR II aerosol datasets are an intriguing opportunity to test out the functionality of some of the new mass spectrometric data analysis methods. The main scientific aim of this study is to identify the "chemical fingerprints" of various anthropogenic aerosol sources, that we expect to show less chemical variation over time, and to use them as an input for resolving the more subtle biogenic aerosol components and their expected seasonal variation. We further aim to utilize trajectory clustering to solve the geographical source areas for the different aerosol types.

The available measurement data consist of 3 one month long measurement campaigns in 2008-2009, in connection to the EUCAARI project (European integrated project on aerosol cloud climate air quality interactions. The main focus of this study is on the Aerosol Mass Spectrometer (C-ToF AMS, Drewnick et al., 2005) data, but there is also a variety of aerosol chemical properties, meteorological, radiation, VOC and trace gas data available for the measurement periods.

The analysis of the datasets will utilize application of the positive matrix factorization (PMF) method and its constrained variation, ME-2 (Multilinear Engine 2), that gives the user more freedom to incorporate available knowledge of the known aerosol properties. Although this part of the analysis is still ongoing, we have already identified several distinct anthropogenic aerosol types, such as local sawmill pollution, mixed urban pollution (traffic, heating, industrial) from the nearby city of Tampere, regional pollution from the urban areas of Southern Finland, and long range transported, highly oxidized pollution from the industrial areas of Europe. One example of a sawmill pollution episode is given below (fig. 1 and 2).

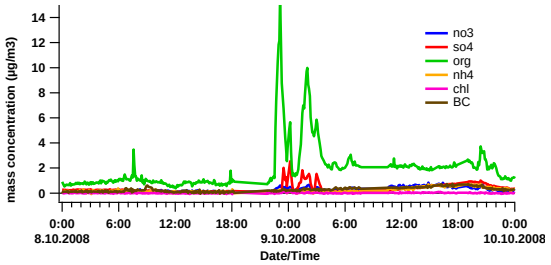

Figure 1. AMS chemical species time series over a night with a sawmill aerosol pollution event on October 8th to 9th of 2009. In addition to a strong increase in organics, the sawmill aerosol also contains sulphates and nitrates.

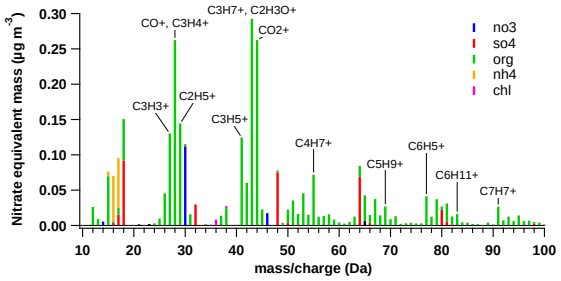

Figure 2. Average AMS mass spectra from the time of the sawmill pollution event. The mass spectra resembles that of the biogenic background aerosol, and contains typically hydrocarbon-like features such as strong alkyl ion fragment series at peaks 27, 29, 41, 43, 55, 57, 69, 71, 83 and 85 Th.

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