



Aircraft Measurements of Aerosol Partitioning and Aging during EUCAARI-LONGREX

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The chemical composition of the atmospheric aerosol burden has significant implications for its climate impacts. Specifically, it determines the scattering or absorbing nature of the aerosol and its affinity for water uptake. Measurements of aerosol chemical composition are presented here from the UK Facility for Airborne Atmospheric Measurement (FAAM) BAe-146 research aircraft from May 2008. The BAe-146 operated out of Oberpfaffenhofen, Germany during the European Integrated Project on Aerosol Cloud Climate Air Quality Interactions (EUCAARI) LONG Range EXperiment (LONGREX). A primary goal of the study was to examine the effects of atmospheric aging on aerosol chemical, physical and optical properties. Science flights were conducted across Northern Europe, during a period of anticyclonic circulation in clear sky conditions.

The aircraft employs a suite of aerosol instruments, which measure the chemical composition, microphysical, optical and hygroscopic properties of the in-situ aerosol population. An Aerodyne compact Time-of-Flight Aerosol Mass Spectrometer (cToF-AMS) measured the mass of volatile and semi-volatile particulate chemical constituents, as a function of size. These components included organic matter, nitrate, sulphate and ammonium. The spatial distribution and chemical evolution of these components will be presented.

The chemical nature of the organic aerosol component is examined via Positive Matrix Factorisation (PMF). The factor analysis of the organic aerosol component revealed the dominance of Oxygenated Organic Aerosol (OOA) over Hydrocarbon-like Organic Aerosol (HOA). OOA is analogous to Secondary Organic Aerosol (SOA), whilst HOA is strongly associated with Primary Organic Aerosol (POA) derived from fossil fuel combustion. Two components were commonly resolved for the OOA component; an aged, more oxidised factor (reflecting the regional organic aerosol background) and a fresher (less aged) component, which exhibited less oxidation than the regional fraction. Close to major source regions in Western Europe, such as the Netherlands and Belgium, the less oxidised (fresher) component was strongly correlated with nitrate. This is indicative of the less oxidised component being semi-volatile, in common with the behaviour of ammonium nitrate. The aircraft data was compared with a ground-based site in the Netherlands. The abundance of nitrate and the less oxidised organic component was increased at the top of the boundary layer when comparing with ground-based measurements. Such features are attributed to partitioning of gas phase precursors to the particle phase, due to reduced temperature and enhanced relative humidity.

Subsequent missions downwind of the source regions were conducted in the Eastern Atlantic in order to investigate the aging of the aerosol burden. The aged regional organic aerosol component was enhanced in such regions, relative to the less oxidised component. The strength of the relationship between nitrate and the less oxidised component decreased with distance from the major source regions. Conversely, downwind of the major source regions, nitrate was increasingly associated with the more aged regional organic component. This is attributed to chemical aging of the fresher organic component, which increasingly resembles the more aged regional organic component. Consequently, the principal loss mechanism for the fresher organic component is via chemical aging. Hence, nitrate is more closely coupled to the more oxidised component under such conditions.

Such phenomena significantly perturb the radiative equilibrium of the climate system and the ecological influence of atmospheric aerosols. The addition of secondary particulate mass increases the Aerosol Optical Depth (AOD) of the atmospheric column, increasing the direct scattering of solar radiation by aerosol particles. The relative enhancement in the concentration of the inorganic and organic components will determine the hygroscopic nature of a mixed inorganic-organic system. This is exacerbated in areas exhibiting high relative humidity, such as the top of the boundary layer. Consequently, wet growth of the particulate matter substantially increases the AOD. Furthermore, chemical aging of the aerosol burden potentially alters the hygroscopic nature of the aerosol. Also, clouds forming at the top of the boundary layer entrain air from beneath them into the cloud. Enhanced aerosol concentrations in such air will lead to a modification of the microphysical properties of the cloud. Such modifications are known to modify the amount, lifetime and radiative impact of clouds, the so-called indirect effect of aerosols. The impacts of such phenomena upon the AOD will be further investigated via comparison with the European surface-based AErosol RObotic NETwork (AERONET). The sub-saturated and super-saturated properties of the aerosol will also be investigated based upon in-situ measurements from the aircraft.