

IN-SITU CHARACTERISATION OF SUBMICRON AEROSOL PHYSICAL, CHEMICAL AND HYGROSCOPIC PROPERTIES AT THE SUPERSITE HORNISGRINDE DURING THE COPS FIELD CAMPAIGN

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INTRODUCTION

Aerosols have both a direct and indirect effect on climate. They alter the Earth's radiation balance by direct scattering and absorption of solar and terrestrial radiation [Haywood and Boucher, 2000]. Aerosol properties also have a large impact on cloud properties, as particles around 100nm diameter and larger are potential Cloud Condensation Nuclei [McFiggans, et al., 2006]. The impact of aerosol properties on clouds includes alterations in cloud optical thickness, cloud lifetime, cloud top height and precipitation suppression (Lohmann et al 2007).

The uncertainty associated with the influence of particles on cloud properties was identified as one of the largest sources of uncertainty in global radiative forcing [Forster, et al., 2007].

FIELD STUDY

The Convective and Orographically-induced Precipitation Study (COPS) is an international field campaign initiated within the Quantitative Precipitation Forecast (QPF) program funded by the German Research Foundation (DFG). The study

aims to improve the accuracy of precipitation forecasts through the use of in-situ and remote sensing instrumentation, advanced high resolution models optimised for use in complex terrain, and data assimilation/ensemble prediction systems.

The region under observation during COPS was in Southwest Germany/Eastern France. This area is prone to frequent thunderstorms in summer, with the skill of numerical weather forecasts in the area being low. The main observation period within COPS was conducted from 1st June until 30th August 2007.



Figure 1: COPS Study Region

Instrument	Measured property
CPC (TSI 3010)	Total particle number concentration (10nm > D > 1000nm)
DMPS	Number size distribution (20nm > D > 600nm)
GRIMM OPC	Number size distribution (250nm > D > 4000nm)
HTDMA	Hygroscopic growth factor @ 90% RH (43,85,127,169,211 and 254nm)
CCNC	Activated particle number concentration
HR-ToF-AMS	Size resolved non-refractory chemical composition (40nm > D > 700 nm)
MAAP	Equivalent total black carbon mass concentration

Table 1: Aerosol instrumentation deployed and properties measured in-situ at the Hornisgrinde hilltop site during COPS-UK.

A contribution to COPS from the United Kingdom (COPS-UK) was made in the form of the deployment of various in-situ and remote sensing instrumentation from the ground and an aircraft platform (the UK Facility for Airborne Atmospheric Research, FAAM BAE-146). Results from the in-situ aerosol sampling instrumentation situated on top of the Hornisgrinde (1164m asl) hill top site are presented here. The in-situ aerosol measurements made at this site by COPS-UK are listed in Table 1.

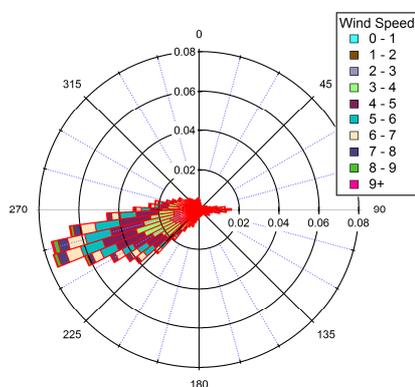


Figure 2: Wind rose from the Hornisgrinde measured during COPS

Measurements were taken from 23rd June and 27th July 2007. Instruments were sampling through an inlet 3m above ground level through a cyclone impactor which removed all particle above 4 μ m diameter. This impactor was used to remove water droplets to prevent instrument damage during in cloud events which occurred frequently throughout the experiment. The correct operation of the impactor was confirmed by the GRIMM OPC which saw no particles above 4 microns diameter. The data presented here are for out of cloud conditions only. Figure 2 show the wind rose measured at the Hornisgrinde during COPS. The wind was nearly always from the South West with typical wind speeds averaging around 5 m/s.

AEROSOL COMPOSITION

The aerosol composition was measured online using a High-Resolution Time-of-Flight Aerosol Mass Spectrometer (HR-ToF-AMS, [DeCarlo, et al., 2006]) which provides mass loadings of Organic, Nitrate, Sulphate and Ammonium submicron aerosol, and a Multi-Angle Absorption Photometer (MAAP, [Petzold, et al., 2005]) to provide a measure of the Black Carbon (BC) mass concentration. The Organic, Nitrate, Sulphate and Ammonium mass concentrations shown here have not been corrected for collection efficiency which would result in a predicted doubling of mass loadings [Drewnick, et al., 2003].

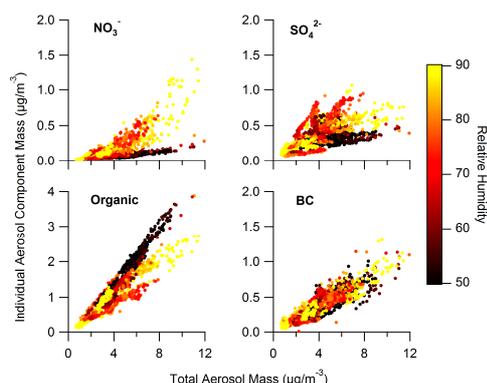


Figure 3: Uncorrected submicron aerosol mass loadings derived from the HR-ToF-AMS (Organic, Sulphate and Nitrate) and MAAP equivalent Black Carbon. Data are coloured according to relative humidity.

The mass loadings of each aerosol component against the total aerosol mass are shown in Figure 3. Organic aerosol contributes the largest amount of material to the total aerosol mass. The relative amount of organic material also seems to be relatively constant. The amount of nitrate in the aerosol, while small in magnitude, exhibits a considerable amount of variability. The highest loadings of nitrate appear during times of high relative humidity. The black carbon and sulphate mass contributions vary in response to these

changes and probably for other reasons including air mass trajectory.

The average organic mass spectrum measured during COPS-UK can be seen in Figure 4. The major mass fragments seen in the spectrum are from m/z 's 18 and 44, both of which are from poly-acidic organics. This suggests the organic aerosol is of a highly aged secondary nature. The mass spectrum compares well with that seen by Alfarra et al [2004] in a clean rural environment. The hygroscopic growth factor of the aerosol measured using the Hygroscopicity Tandem Differential Mobility Analyser was low (typically 1.3) which agrees with the high organic aerosol content. The HTDMA also suggests the presence of a single internally mixed accumulation mode in the aerosol population.

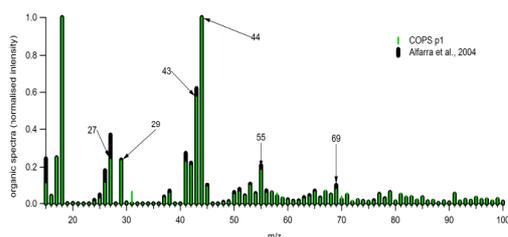


Figure 4: Average organic mass spectrum from COPS and from a rural location [Alfarra, et al., 2004].

CCN MEASUREMENTS

Measurements of CCN concentrations were made using a DMT CCNC which was ran cycling at 5 different super saturation ratios. The instrument measured total and mono-disperse CCN concentrations at alternate hours. The CCNC measured through a DMPS system to measure mono-disperse aerosol. The data presented here are uncorrected for multiple charge effects which may alter the measured CCN spectra. Comparing the measured CCN concentrations with the measured CN concentrations throughout the particle size distribution allow the determination of the

critical diameter (D_{50} , the diameter at which 50% of the particles activate at a given supersaturation). A time series of aerosol composition from the HR-ToF-AMS shown in Figure 5 shows 2 highlighted areas. One is dominated by organics and the other which has significantly more inorganics relative to the organics.

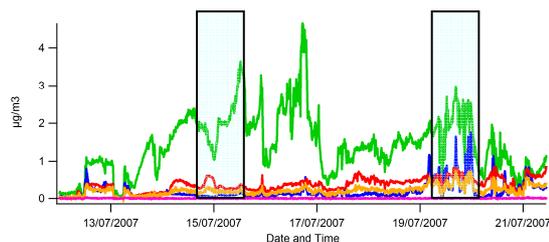


Figure 5: Time series of uncorrected mass loadings of organic (green), nitrate (blue) sulphate (red) and ammonium (orange) aerosol components.

Figures 6 and 7 show the uncorrected CN and CCN concentrations, as well as the particle size being sampled as a function of time. The CCNC was exposing the sample to a supersaturation of 0.07% during these scans. The highly organic aerosol seen on the 15th July and shown in Figure 6 appears to have a D_{50} of around 256nm, with the corresponding D_{50} of the more inorganic influenced aerosol being around 210nm.

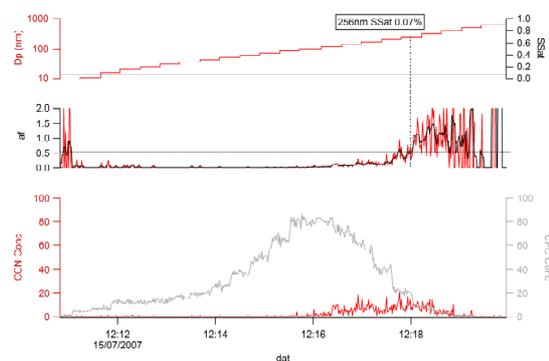


Figure 6: Time series of uncorrected CN and CCN number concentration during mono-disperse sampling in high organic mass fraction aerosol. Also shown are activated fraction and particle size.

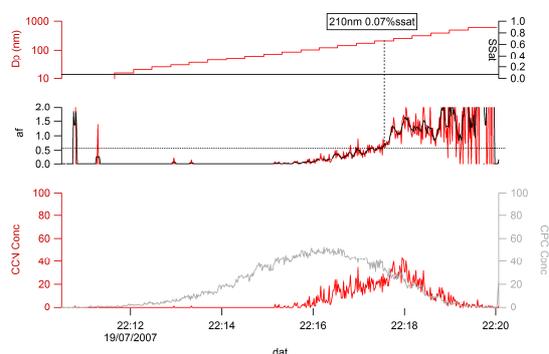


Figure 7: Time series of uncorrected CN and CCN number concentration during mono-disperse sampling in high nitrate mass fraction aerosol. Also shown are activated fraction and particle size.

CONCLUSIONS

Aerosol composition on the Hornisgrinde hill top site during COPS-UK was found to be highly organic in nature. The organic aerosol was also seen to be highly oxidised. The lack of variability in these properties could be due to the central European location and due to the altitude of the field site. Ammonium nitrate was enhanced during periods of high relative humidity. The aerosol measured by the HR-ToF-AMS was found to be pH neutral at all times.

Aerosol particles exhibited low sub-saturated growth at 85% humidity due to the high organic content. The measured D_{50} which dictates the number of CCN formed from a particle size distribution was seen to be large in a period of very of organic to inorganic aerosol mass.

ACKNOWLEDGMENTS

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