CLOUD-PROCESSING AND AEROSOL OPTICAL PROPERTIES AT A POLLUTED CONTINENTAL SITE

J.A. Ogren[1], E. Andrews[1,2], J. Allan[3], K. Bower[3], H. Coe[3], B.Corris[3], M. Flynn[3], D. Liu[3], W. Morgan[3], and P. Williams[3]

[1]Earth System Research Laboratory, NOAA, Boulder, CO 80305, USA
[2]Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO 80305, USA
[3]SEAES, University of Manchester, Manchester, M13 9PL, UK

1. INTRODUCTION

The magnitude, and even the sign, of the climate forcing by aerosol particles is strongly dependent upon both the aerosol single-scattering albedo (SSA), which is the fraction of the aerosol light extinction that is due to scattering, and the angular dependence of light scattering, which can be parameterized by aerosol properties such as the asymmetry parameter or the backscattering fraction (BFR). Long-term monitoring at a variety of surface sites reveals a systematic decrease in aerosol SSA as the aerosol loading decreases, i.e., aerosols are "blacker" in the cleanest air and an increase in BFR, i.e., aerosols are smaller in the cleanest air (Delene and Ogren, 2002).

One hypothesis for this behavior is that clouds preferentially scavenge large and primarily scattering aerosols more effectively than small and/or absorbing aerosols, which is what would be expected if the absorbing component of the aerosol is dominated by hydrophobic black carbon and the scattering component is dominated by hygroscopic species like sulfates. If the clouds precipitate, the aerosol that remains after the cloud dissipates will be smaller and enriched in black carbon relative to the water-soluble species that often dominate aerosol light scattering.

To address this hypothesis, we have been conducting a series of field campaigns in a variety of locations, with a focus on scattering and absorption of aerosol particles in cloud-free air, in interstitial air inside of clouds, and in the evaporated residuals of cloud droplets. The current work emphasizes an experiment in November-December, 2006 at Holme Moss, a field site on the moors about 30 km northeast of Manchester, UK that has been used as a research site by the University of Manchester (UM) for more than a decade for both field campaigns and long term climatological measurements (e.g., Beswick et al., 2003; http://cloudbase.phy.umist.ac.uk/field/).

Long-term climatology suggested that the site, at 525 m asl, would frequently be in cloud (150-200 hrs/month in autumn), and the site was indeed in fog ~22% of the sampling period (fog defined here for simplicity as visibility < 5 km). While the site had potential to receive fresh pollution from Manchester and Leeds, during the campaign the wind was primarily from the southwest meaning Manchester was the main source of aerosol sampled.

2. EXPERIMENTAL APPROACH

Identical instruments for measuring aerosol light scattering and light absorption were operated downstream of two complementary inlets. A counterflow virtual impactor (CVI) provided samples of cloud droplet residuals, i.e., the aerosol particles that remain when a cloud droplet

Corresponding author's address: John A. Ogren, NOAA R/GMD1, 325 Broadway, Boulder, CO 80305, USA; E-Mail: John.A.Ogren@noaa.gov

evaporates, and a radial impactor provided a sample of the interstitial particles smaller than the cloud droplets sampled by the CVI. The interstitial inlet was also used to sample ambient aerosols during cloud-free periods.

The radial impactor size cut was set to 5 μm diameter, i.e., all particles larger than 5 μm were assumed to be cloud droplets and all smaller particles were called interstitial. Based on calculations, the size cut of the CVI was approximately 8 μm, so there was a 3 μm gap in measurements between interstitial and cloud aerosol measurements – likely the particles in this size range were small cloud drops. Measurements of particles from both inlets were made at low relative humidity so that they could be directly compared. Aerosol light scattering coefficient was measured with an integrating nephelometer (Model 3563, TSI, Inc., St. Paul, USA) and light absorption coefficient was measured with a filter-based light photometer (Model PSAP, Radiance Research, Seattle, USA). All the measurements reported here are at a wavelength of 550 nm. Nephelometer data were corrected for truncation errors following the procedures recommended by Anderson and Ogren (1998) and the PSAP data were adjusted using the procedures recommended by Bond et al. (1999). Aerosol light extinction coefficient, σ_{ep}, was calculated as the sum of the corrected scattering and absorption coefficients. Cloud liquid water content (LWC) was measured with tunable diode laser hygrometer (MayComm, Wilmington, USA) downstream of the CVI.

3. RESULTS

The Holme Moss site was a very interesting location to sample aerosol. While observed aerosol extinction was fairly typical for a semi-remote location (median extinction ~20 Mm^{-1}), the aerosol single scattering albedo was consistently significantly lower than most sites at which NOAA had previously made measurements (Holme Moss median SSA ~0. 84). In other words, Holme Moss was an ideal location for measuring absorbing aerosol. The source of the absorbing aerosol is likely the diesel exhaust from vehicular traffic transported to the site from Manchester as well as nearer rural activities (peat burning and residential coal fires).

During cloudy periods at the site, the interstitial aerosol tended to be smaller and more absorbing than the ambient aerosol observed during clear conditions. There was also a decrease in aerosol extinction during cloud events compared to clear conditions, e.g., $\sigma_{ep,interstit} < \sigma_{ep,clear}$. These results are consistent with the systematic variation reported by Delene and Ogren (2002) and with observations made at other sites (Figure 1) where the aerosol properties could be segregated by the presence or absence of cloud (e.g., Ogren et al., 2004). These data suggest that clouds are preferentially scavenging the larger, more scattering aerosol leaving small absorbing aerosol in the interstitial air. Interestingly, cloud water collected at the Holme Moss site showed a distinct grayish tinge suggesting that at least some of the absorbing aerosol was incorporated into the cloud droplets and the single particle soot photometer (SP2, DMT, Boulder, USA) showed that the cloud drop residuals did contain some black carbon.

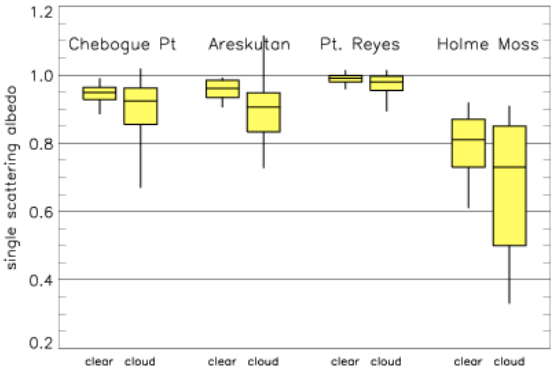

Figure 1: Single scattering albedo measured through inlet with 5 μm radial impactor; clear is SSA when no cloud is present, cloud is SSA for interstitial aerosol.

The extremely windy conditions at the site (wind speeds typically > 10 m/s) resulted in the CVI sampling non-ideally. Based on wind tunnel studies by Noone et al., (1992)

sampling efficiencies for winds > 10 m/s are 45% for 8 um droplets and decrease precipitously for larger drops, e.g. ~20% sampling efficiency for 10 um droplets at 10 m/s wind speed. Based on the poor CVI sampling conditions and the detection limits of the optical instruments, the aerosol optical data needs much more analysis before any cloud drop residual optical properties can be produced. Two more sensitive chemical instruments did show differences in the cloud drop residuals compared to interstitial air. The cloud drop residuals from the CVI tended to have a higher fraction of sulfate (based on aerosol mass spectrometer, AMS, measurements) and lower soot content (based on SP2 measurements).

The suite of instruments deployed at Holme Moss provided some indication of how aerosol particles might interact with water vapor in the atmosphere. Measured aerosol hygroscopicity was surprisingly high (median f(RH)~2.0, where f(RH) is the ratio of light scattering at 85% to the value at 40%) considering the polluted influence suggested by the low values of SSA. For comparison, Sheridan et al., (2001) showed that smoke aerosol from local field fires at a rural continental site in the US was significantly less hygroscopic than the background aerosol at the site. However, when put in the context of AMS measurements, the measured f(RH) at Holme Moss was consistent with the composition dependence of f(RH) described by Quinn et al (2005) (Figure 2).

A cloud condensation nuclei counter (CCN, DMT, Boulder, USA) was used to measure CCN concentrations as a function of supersaturation. The CCN activated fraction (ratio of CCN concentration to total aerosol concentration (CN)) was highly variable, and complete activation was not observed even at the highest supersaturations studied (1.5 percent). The CCN activated fraction appeared to increase with increasing SSA (Figure 3) and decrease with increasing organic contribution to the aerosol.

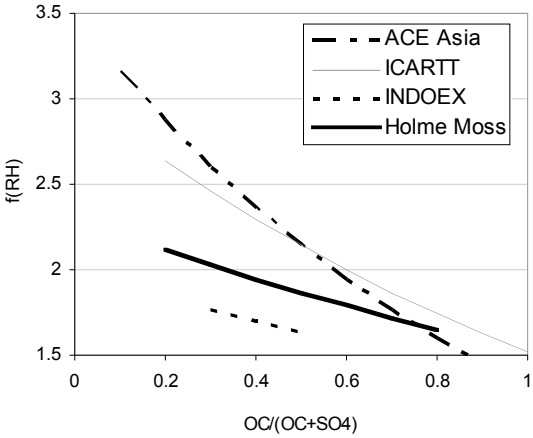

Figure 2: f(RH) as a function of aerosol composition (based on figure from Quinn et al., (2005)); Holme Moss line added for this report.

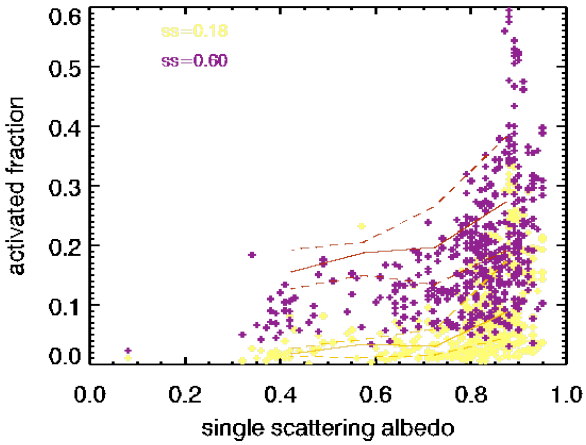

Figure 3: CCN activated fraction (CCN/CN) for two supersaturations (given in percent) as a function of single scattering albedo

4. CONCLUSIONS

Based on the results from Holme Moss and several other field campaigns, we have found that clouds tend to scavenge larger, less scattering aerosol leaving the darker, smaller aerosol in the interstitial air. In terms of intrinsic aerosol properties, this means that BFR increases and SSA decreases in cloud scavenged air and interstitial aerosol. As BFR and SSA are both important factors in aerosol radiative forcing, it follows that the radiative

properties of the cloud-processed aerosol are quite different than the pre-cloud aerosol.

Aerosol composition (as indicated by SSA and aerosol mass spectrometry chemical measurements) allows us to explore how particle composition influences the interactions between water and aerosol particles which has implications for both direct (aerosol hygroscopicity) and indirect (CCN activity) forcing.

Future plans include evaluating the Holme Moss data set in conjunction with several other similar data sets to develop

1) improved parameterization of elemental carbon (EC) processing by clouds

2) better understanding of size and composition dependent aerosol processing by clouds

3) reduced uncertainty in EC cycle and lifetime in atmosphere

4) better understanding of the cloud processing mechanisms affecting aerosol properties

Incorporation of findings from these planned explorations into climate models will reduce the uncertainty in predictive modeling capabilities and improve our ability to identify the amount of aerosol radiative forcing versus other forcing factors such as greenhouse gases.

5. ACKNOWLEDGEMENTS

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