FIELD APPLICATIONS OF AN AEROSOL MASS SPECTROMETER: WHAT ARE WE LEARNING ABOUT AEROSOL CHEMICAL AND MICROPHYSICAL PROPERTIES?

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1. INTRODUCTION

Atmospheric aerosols play important roles in climatology and visibility by absorbing and scattering solar radiation and also adversely affect human health in ways that are not fully understood. Motivated both by climate change issues and by health concerns associated with airborne particulates, there have been increased efforts over the past decade to better characterize aerosol chemical and microphysical properties to help provide guide lines for the legislation of appropriate particulate matter emission standards. Thus, in addition to intensive air quality measurement campaigns, there has also been a focused effort on developing and testing advanced technology aerosol instrumentation.

Presented here are results obtained from national and international deployments of the Aerodyne Aerosol Mass Spectrometer (AMS) system by different research groups. The AMS is capable of providing size and compositionally resolved mass loadings of non-refractory aerosol components in real time. Recent deployments of the AMS include field campaigns in Houston (TEXAQS 2000), New York City (PMTACS 2001), ACE-ASIA (2001), Michigan (PROPHET 2001), Vancouver (PACIFIC 2001), California (ITCT, 2002), Pittsburgh (PAQS 2002), New England (NEAQS 2002), Mexico City (2002 and 2003), Finland (QUEST 2003) and Tokyo. An analysis of aerosol mass, size, and composition trends obtained during these campaigns reveal diurnal variations in the photochemically driven formation of sulfate, nitrate, and oxidized organic aerosols. The chemical composition of these aerosols can be clearly distinguished from those observed during vehicular exhaust or biomass burning pollution events. These results, when combined with airmass back trajectory calculations and ancillary gas phase measurements allow for the characterization of particle sources.

2..AEROSOL MASS SPECTROMETER (AMS)

2.1 Introduction

The AMS is shown schematically in Figure 1. The basic AMS design consists of three main parts: an aerosol inlet, a particle sizing chamber, and a particle composition detection section. Each section is separated by small apertures and is differentially pumped. Particles are sampled from ambient pressure and focused by an aerodynamic inlet into a narrow beam (~1 mm diameter) as they enter into high vacuum (Zhang et al, 2002). Particle aerodynamic diameter is determined by measuring particle velocity via particle time-of flight (p-TOF) over a known flight path. As shown in Figure 2, particle chemical composition is determined via flash vaporization followed by electron impact (EI) ionization mass spectrometry. Each of these components is based on standard technology that is easily constructed and robustly operated making the AMS transportable and suitable for field measurements. The current AMS is self-contained in a single rack (51”H x 24”D x 41”W). It weights ~400 lbs. and consumes ~600 Watts of power (Jayne et al, 2000).

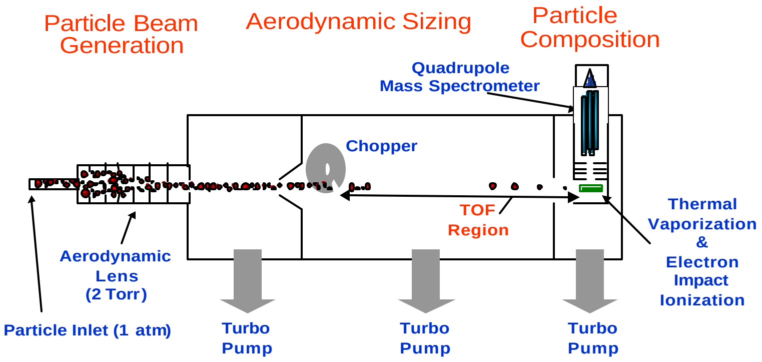

Figure 1. Schematic of the aerosol mass spectrometer (AMS).

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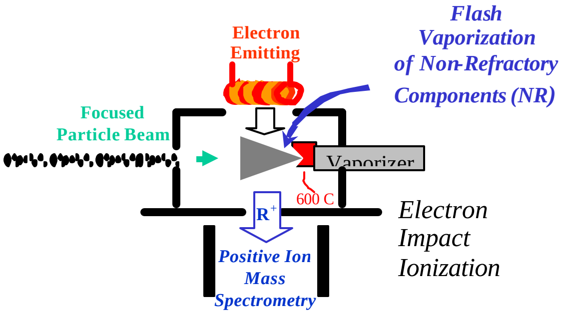

Figure 2. Schematic of AMS Detector

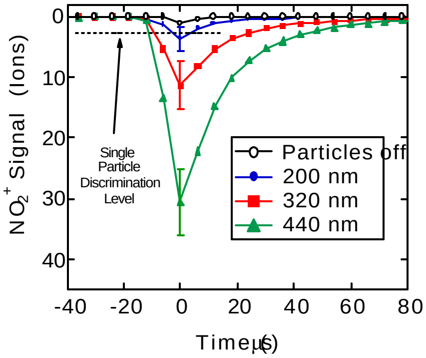

Figure 3. Flash vaporization of NH_4NO_3 particles

The major advantage of the AMS compared to other particle mass spectrometers is the combined quantitative particle collection efficiency from 30 nm to 1 µm (near unity for particles in the size range 60nm to 600 nm) and quantitative chemical detection of volatile and semi-volatile components. The key to the latter is the separation of particle vaporization and vapor ionization steps (Figure 2), which enables linear chemical composition detection even for complex mixtures. Figure 3 shows ion pulses detected from individual sub-micron particles, showing flash vaporization (< 100 µsec) and sensitivity that is linear with particle volume [Jayne et al., 2000]. Sensitivity of ~0.01 µg/m^3 (in a few minutes) for individual chemical components has been demonstrated with the Aerodyne AMS. Moreover, the fast vaporization provides the time resolution for measurement of particle TOF enabling aerodynamic particle sizing.

2.2. Quantitative Chemical Analysis

The vaporized species analyzed by the AMS are best described as non-refractory (NR), defined as all chemical components that vaporize (within a second) at the vaporizer temperature of ~600C. This includes most atmospheric aerosol composition (for example, most organic carbon and inorganics such NH_4NO_3 and $(NH4)_2SO_4$). The major limitation of the AMS detection is lack of sensitivity to refractory substances which include crustal oxides (dust) and elemental carbon. At higher vaporizer temperature s (up to 900C) most mineral salts (including $NaSO_4$ and seasalt) are detectable (Allan et al, 2003c).

Figure 4 shows a mass spectrum of ambient aerosol that represents the total sub-micron mass loading. This mass spectrum is recorded by blocking and unblocking the focused aerosol beam and taking the difference. One of the key concepts in the AMS quantitative analysis is to calculate aerosol loading by summing ion signals at all m/z values. In this way, detected mass spectral signals can be converted directly to aerosol mass loadings using well known EI ionization cross sections. The AMS response is calibrated based on detecting single NH_4NO_3 particles (as in Figure 3), size selected either with a differential mobility analyzer (DMA) or measured with the AMS itself. Quantitative (and chemical) analysis then requires identification of mass spectral ion fragmentation patterns in order to apply ionization efficiencies (IE) (relative to the nitrate calibration) for chemical classes.

A key aspect of this mass spectral analysis is that ion signals for a given chemical class are obtained by summing all the appropriate ions for that class, so that total ion signal is simply proportional to the IE derived from ionization cross sections (Jimenez et al, 2003a; Allan et al, 2003a). Figure 4 shows the chemical classes identified by color (as labeled) for ammonia (m/z15,16,17 for NH^+,NH_2^+,NH_3^+), nitrate (m/z 30,46 for NO^+,NO^{2+}), sulfate m/z (48,64,80,81,98 for SO^+, SO_2^+,SO_3^+,HSO_3^+,$H_2SO_4^+$) and organics, which include most of the remaining ion fragments. The ion fragmentation patterns of these inorganic and organic classes are easily

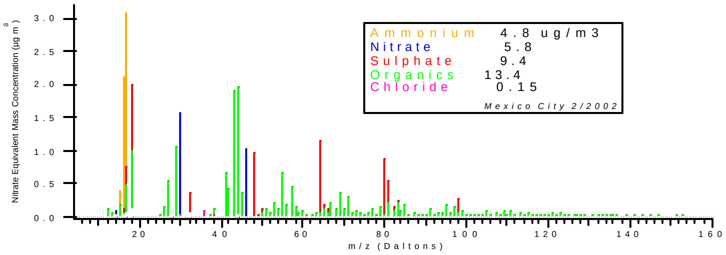

Figure 3. Typical electron impact ionization mass spectrum of ambient aerosol with chemical classes identified by color.

separated (as in Figure 4) though the detailed mass spectral analysis does account for interferences (Allan et, 2003d).

The combination of vaporizer/ionizer geometry (Figure 2) and EI ionization makes the AMS a quantitative (and universal) detector for all chemical species vaporized at temperatures at 600 to 900C. Every vapor molecule passes through the ionizer with an ionization efficiency that can be estimated from previous work. As has been repeatedly demonstrated from comparison with other chemical analyses of ambient aerosol mass loading (see, for example, Drewnick et al, 2003a; 2003b), this approach has an absolute accuracy of better than 20%. That accuracy can be improved for analysis of specific compounds for which detailed laboratory calibration is possible.

2.3 Aerodynamic Collection and Sizing

This quantitative analysis applies to mass loading of sub-micron aerosol particles that reach the detector. At this point the major uncertainty in AMS determination of ambient aerosol mass loading involves understanding the collection efficiency (CE) of particles that reach the detector. For spherical particles, particle focusing in the aerodynamic lens and transmission to the detector are well understood, with 100% collection between 60 and 600nm. However, particle focusing for aspherical shapes is reduced to a degree that is hard to predict. In general, there are two issues when comparing AMS results to other aerosol mass loading measurements: collection of large particles, largely reflecting the fact that most current techniques measure PM2.5 (i.e. particle mass for particles less than 2.5 um in diameter) while the AMS is (at present) actually a PM1.0 measurement; and losses of sub-micron particles in the AMS due to poor focusing of aspherical particles (see discussion in Drewnick et al, 2003a).

Based on many observations, it is now apparent that the sub-micron aerosol focusing depends on the relative humidity (RH) at the AMS sampling inlet. For RH<60-70%, typical values of CE are ~0.5 while CE becomes = 0.8-1.0 for RH>80-90% (the exact value of CE depends on the PM1.0/PM2.5 ratio). This is true, in particular, for sulfate dominated aerosol and is consistent with laboratory work which shows that relatively “dry” particles of $(NH_4)_2SO_4$ aerosol effloresce in the aerodynamic lens (where under vacuum the aerosol dries due to fast evaporation of water vapor). Efflorescence forms solid, aspherical crystals which are poorly focused by the aerodynamic lens. At higher RH, higher aerosol water content leads to better focusing of more spherical particles.

This effect has been observed in the field, either by ambient variation in RH (Delia et al, PROPHET, unpublished results, 2001) or via inlet RH control (Allan et al 2003c; 2003e). More recent work has utilized a beam width probe within the AMS (based on a moveable wire placed in front of the detector) that directly measures the actual focused width of the particle beam, enabling an in situ estimate of CE. Initial results confirm the effect of RH on focusing (Allan et al, 2003e) and also have shown that focusing (and CE) are higher for organic rich particles (Jimenez et al, 2003b, Mexico City). In the future, beam width measurements will be routine for AMS operation.

The AMS measures the vacuum aerodynamic particle diameter (D_{va}) which, for spherical particles is the product of particle density (ρ) and geometric diameter (d), $D_{va} = \rho\ d$. This differs from the classical aerodynamic diameter that is proportional to the $\rho^{1/2}$. For aspherical particles, there are additional factors in D_{va} that take into account particle shape. In fact, future work will combine measurements of physical particle size (e.g. parallel measurement of mobility diameter) and AMS aerodynamic sizing and beam focusing, in order to correlate particle morphology and chemical composition.

3. Results and Discussion

Figure 5 displays typical data from AMS aerosol analysis, in this case, size resolved mass loading of sulfate containing aerosol from one day of sampling in Houston (Canagaratna et al., TEXAQS 2000, unpublished results). The middle panel shows an image plot of the mass distribution ($\log(D_{va})$ versus time, colored by differential mass loading); the top panel shows total sulfate loading (obtained by integrating the mass distribution); while the side panel shows the average mass distribution averaged for that day.
The total sulfate loading is compared to that measured by a PILS ion chromatography instrument (Weber et al, 2001; assuming CE = 0.5 as discussed above).

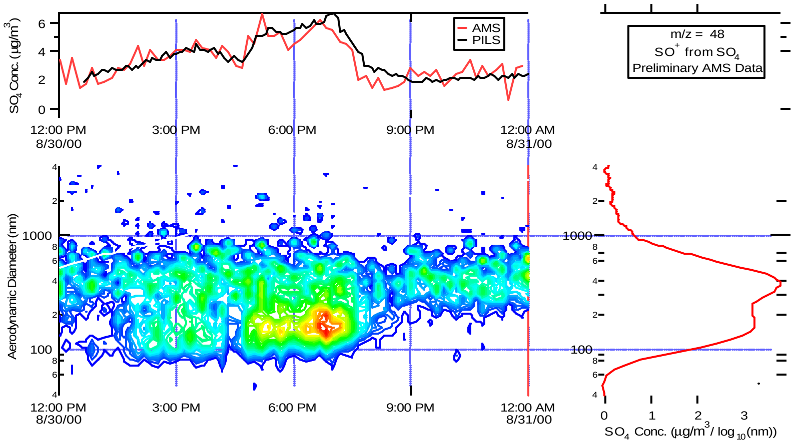

Figure 5. AMS data from TEXAQS 2002 in Houston, TX. Bottom panel shows the size distribution of sulfate versus time, top panel shows the total sulfate loading versus time and the right panel shows the size distribution.

The aerosol time trend is typical of industrialized (urban) regions There is a ubiquitous background (accumulation) mode at D_{va} ~300-500nm that tends to peak in the afternoon (on sunny days) in areas where there is SO_2 transport (associated with power plants). In Figure 5, the afternoon peak was also associated with a small particle mode ~150nm diameter. The appearance of the small mode is coincident with a rise in gas phase SO_2. Both the small and large particle modes also contain organics whose mass loading is roughly equal to that of sulfate.

These results are consistent with photochemical production of secondary aerosol (both from SO_2 and volatile organic compounds, VOCs) as was observed from aircraft measurements in power plant plumes in the Houston region on the same afternoon (Brock et al, 2003). In fact, such growth of smaller particles containing both sulfate and organics has been observed repeatedly at many sites worldwide (Houston, TEXAQS 2000; northern Michigan PROPHET, 2001; PAQS, Pittsburgh, 2002; QUEST, Finland, 2003; Mexico City, 2003). It is important to note that the AMS can only measure particle growth for d>30nm. In most of these cases, physical number density measurements indicate that new particle nucleation was the precursor for the AMS observed particle growth.

Figure 6 summarizes typical atmospheric aerosol chemistry and microphysics (PEMTACS, New York City; Drewnick et al, 2003b). Sulfate (together with organics) forms a large accumulation mode (as in Houston, Figure 5), while in urban areas, the organic mass distribution is distinctly bi-modal with a small mode centered at D_{va}~80-100nm. The small organic mode shows a clear peak in early morning associated with rush hour traffic. The small mode time trends also correlate well with those of NO and CO, indicative of a transportation source (Allen et al, 2003b; Zhang et al, 2003, Pittsburgh 2002; Jimenez et al, 2003b, Mexico City). Moreover, both the size and chemistry (see below) of the small organic mode match that measured from direct emission in diesel bus and truck plumes (Canagaratna et al, 2003). Finally, lab studies have shown that the aerodynamic size distribution centered at D_{va}~80-100nm is representative of aspherical, fractal soot particles (Slowik et al, Boston College, 2003), consistent with mobility and density measurement of ambient and engine particles (McMurry et al, 2002).

The principle difference between regional and urban mass distributions is the absence of the smaller organic mode in the former. For example, in remote areas (e.g., northern Michigan, Finland, outflow from China in Korea) there is no evidence of such a small organic mode. For the accumulation mode, mass loading levels and chemical composition clearly correlate with the degree of anthropogenic (industrial) influence. For example, the accumulation mode organic/SO_4 loadings in New York City show several day duration indicative of regional synoptic transport; the diurnal cycle also shows a peak in the afternoon (much as in Houston) indicative of in situ photochemical processing. In (northern) remote locations (e.g., Michigan and Finland)

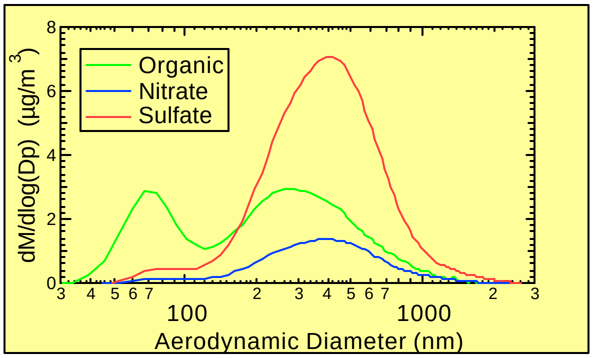

Figure 6. Speciated mass loadings from PEMTACS, New York City, 2002 showing bimodal organic size distribution.

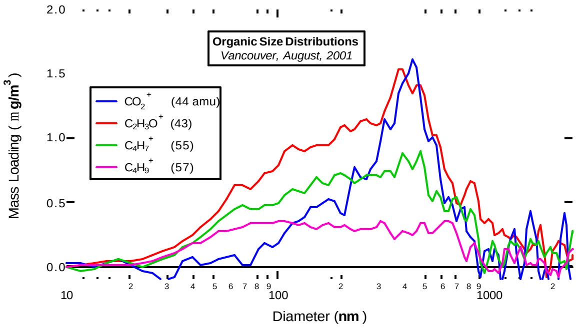

Figure 7. Size distributions for CO_2^+ (m/z 44), $C_2H_3O^+$ (m/z 43), $C_4H_7^+$ (m/z 55) and $C_4H_9^+$ (m/z 57) indicating externally mixed aerosol particles.

there is a clear difference in composition depending on air flow direction; for southerly flow, organic loading tends to be larger than sulfate while for northerly the converse is true, reflecting the degree of anthropogenic influence. The other extreme is outflow from China, measured both on aircraft and at a fixed site on Jeju Island, Korea where the aerosol composition is dominated by accumulated sulfate emissions (Bahreini et al, 2003; Topping et al, 2003). In other urban areas either secondary nitrate or organics (or both) can dominate, depending on the relative magnitudes of different precursor emissions (Jimenez et al, 2003b, Mexico City; Tekegawa et al, unpublished results, Tokyo, 2003). In aircraft flights during the summer of 2002 over New England, all of these phenomena were observed: in free tropospheric air above the boundary layer, organics were larger than sulfate, except for well defined layers indicative of sulfate plumes, both well aged and with evident of new particle growth, while the outflow from urban centers showed evidence of small organic primary emissions (Jayne et al, 2003, NEAQS).

The (operational) definition of organic carbon (OC) is the fraction of mass that evolves CO_2 upon volatilization and combustion of collected aerosol samples. For the AMS, OC is defined as the sum of mass spectral ion fragments that can be classified as organic (e.g., $C_iH_jO_kN_l^+$). In fact, there is significant information about composition available from analysis of detailed ion fragmentation patterns in the total organic mass spectrum. An example, of this is shown in Figure 7, which displays the mass distribution of individual organic ions in an urban aerosol (Vancouver, Canada, Alfarra et al, 2003; see also Allen et al, 2002b). These distributions clearly show that the organic composition is different for the “small” and “large” modes. The small mode is dominated by hydrocarbon fragments (e.g. $C_nH_{2n-1}^+$ and $C_nH_{2n+1}^+$). In fact, the mass spectrum of the small organic mode (measured either from direct diesel emission or from ambient loading during “rush hour”) is nearly identical to that of lubricating oil (Canagaratna et al, 2003; Tobias et al, 2001), indicating that the small organic mode is composed of fractal (black carbon) soot particles with adsorbed oil.

In contrast, the large organic mode is weighted toward oxygen-containing fragments, CO_2^+ (m/z 44) and $C_2H_3O^+$ (m/z 43), which originate from decarboxylation of organic acids and from carbonyls (ketones, aldehydes), respectively (McLafferty and Turecek, 1993). In fact, outside of urban areas, almost every AMS ambient organic mass spectrum shows a peak at m/z 44 which is also correlated with enhanced signals at m/z 43 (as in Figure 4, for example). The extreme example is the observation of aged aerosol in Asia (on Jeju Island, Korea) where highly oxygenated organic aerosol (inferred from high m/z 44 signal; Topping et al, 2003) was compared to analysis of water soluble organic in collected aerosol (see Decesari et al, 2000). In more remote areas, i.e. northerly flow at high latitude acidic aerosol derived from anthropogenic emissions (Delia et al, 2003; Allan et al, 2003e). In a study of the outflow of the regional plume from the northeastern United States, again highly processed organic aerosol was observed, well correlated with photochemical age derived from measured VOC distributions, with clear evidence of organic nitrate aerosol composition (Middlebrook et al, 2003).

In summary, this contrast between smaller organic aerosol (primary combustion emission) and larger, mixed sulfate/organic accumulation (attributed to photochemical processing) aerosol has been observed worldwide (at least in the northern hemisphere). This includes evidence of growth of both new particle and secondary aerosol. The results are based on about 25 deployments of the AMS in North America, Europe and Asia, involving about 10 institutions. Future work will focus on resolving the microphysical and chemical evolution of atmospheric aerosol: What is the fate of small primary aerosol particles, i.e. scavenging and/or chemical processing? What fraction of the oxygenated organic aerosol originated from gas phase (secondary) versus in particle processing of condensed (semi-volatile) organic? What are the processing, transport and deposition lifetimes of atmospheric aerosol?

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