Characterization of Laboratory and Ambient Particles Using the Combination of Aerosol Mass Spectrometry and Light Scattering Techniques

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Introduction

A light scattering module has been incorporated into the Aerodyne Aerosol Mass Spectrometer (AMS). For a given non-refractory particle the combined light scattering-AMS system provides a light scattering signal (function of the particle's geometric diameter and composition), a chemical ion signal, and a vacuum aerodynamic diameter (Dva). Particle density is extracted from the relationship between optical diameter (function of light scattering signal) and Dva. Knowing the density, size distribution, and chemistry of an aerosol plume can lead to a better understanding of how the aerosols were formed and processed. This new information begins to answer the follow questions:

-Are the organic aerosols primary or secondary?
-Is the composition of the aerosol plume purely organic, or a mixture with organic and inorganic species?
-Are the particles internally or externally mixed?

The light scattering module also serves as an internal particle counter for both non-refractory aerosols, which the AMS chemically speciates, and refractory aerosols, which the AMS does not fully detect. Therefore, the light scattering module functions as a diagnostic tool for understanding the collection efficiency of the AMS.

Light Scattering-AMS Schematics

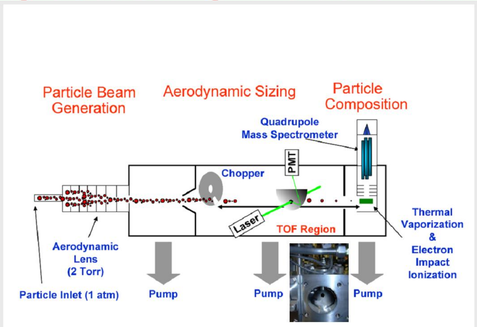

Figure 1a. Light Scattering-AMS Schematic

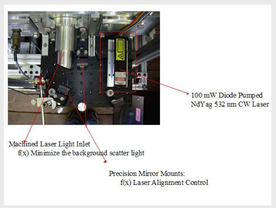

Figure 1b. Optical Setup for Laser

Figure 1a above shows the location of the light scattering module within the AMS. From this figure it is possible to see that for every non-refractory particle above our detection limit, we should obtain a light scattering signal and chemical ion signal each with its own associated time-of-flight.

Figure 1b (left) illustrates the arrangement of the laser optics.

Laser Geometry and Alignment

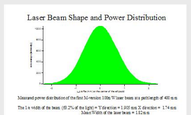

Figure 2a. Laser Power Profile

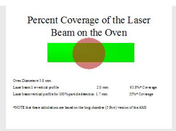

Figure 2b. Schematic of Laser Coverage on the Oven

Figure 2a (Left) shows the Gaussian distribution of the laser beam at a pathlength of 400 mm.

Figure 2b (Left) indicates the percent coverage of the oven by the laser beam.

Figure 2c (Below) shows the effective Vertical-profile measurement of the laser beam expanded onto the oven surface. The laser beam height is approximately 2.00 mm FWHM which corresponds with 64% coverage by area on the oven.

Figure 2d (Below) shows the effective horizontal-profile. Here we are measuring the oven diameter with the MS counts and the ellipsoid hole diameter (4.54 mm).

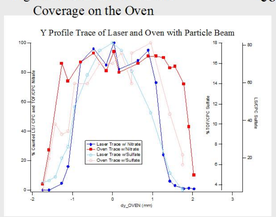

Figure 2c. Particle Walk in the Vertical

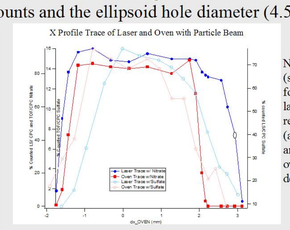

Figure 2d. Particle Walk in the Horizontal

Note that NH_4NO_3 particle beam (spherical particles) is well focused and that the edges of the laser and oven are highly resolved. The $(NH_4)_2SO_4$ beam (asphericaL,dry) is more divergent and thus the resolution of the oven and laser are not as sharply defined.

Laboratory Aerosol Light Scattering Signals

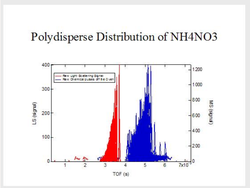

Figure 3a-1.

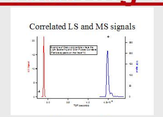

Figure 3b.

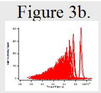

Figure 3a-2. Zoom-in on light scattering signal

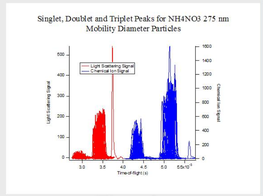

Figure 3c.

Figures 3a and 3c show the raw light scattering signals for an ammonium nitrate polydisperse and monodipserse aerosol. The monodisperse distrbution, sampled through a Differential Mobility Analyzer (DMA), shows the sensitivity of the light scattering system to detecting doubly and triply charged particles. The correlated nature of the light scattering and chemical ion signals is illustrated by the extracted single particle in figure 3b.

Calibration of the Light Scattering Signals

Size selecting aerosols from a differential mobility analyzer (DMA) allows one to calibrate the light scattering system.

The 50% detection limit of the light scattering system is approximately 280 nm mobility diameter (Figure 4a. Right).

Figure 4a, shows that the effect of refractive index on the magnitude of the light scattering signal is minimal. Particles of different compositions but with the same mobility size (Dmob), produce light scattering signals of the same intensity. This is shown for oleic acid and ammonium nitrate, and has also been tested with PSL. For a list of refractive indicies and densities see figure 4b on the right.

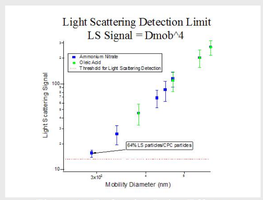

Figure 4a. Refractive Index Effect on Light Scattering Signal

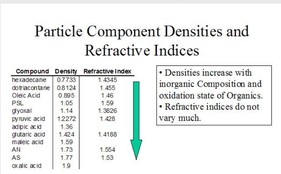

Figure 4b.

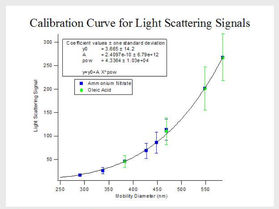

Figure 4c. Power Law Curve Fit Relation Between the Light Scattering Signal and Dmob

From figure 4c on the left, the light scattering signal dependence on mobility diameter can be described using a power law in which the scatter signal falls off as approximately Dmob^4 over the detected size range. This is due to the fact that our light scattering signals fall within a transition region between the geometric (Dmob^2) and Rayleigh (Dmob^6) scattering regimes.

Measuring Laboratory Aerosol Densities

Equation 5a. Dva = Dmob(Optical size) * ρ * Shape Factor (S)[1]

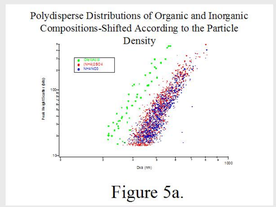

Figure 5a.

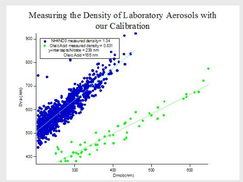

Figure 5b.

Using equation 5a we can solve for the Dmob (optical size) based on the light scattering signal and Dva for each individual particle. Plotting the Dva vs. the Dmob(optical size) in figure 5b, we extract particle densities for the calibration species.

[1]Jayne et al. Development of an Aerosol Mass Spectrometer for Size and Composition Analysis of Submicron Particles. *AS&T*, 33. 49-70. (2000)

S = 1 for spherical particles (oleic acid) and S = 0.8 for crystalline NH_4NO_3 particles

Chebogue Point, Nova Scotia NEAQS-ITCT 2004 Light Scattering Analysis of Two Different Ambient Aerosol Plumes

As part of the NEAQS (Northeast Air Quality Study) from July-August 2004, the combined LS-AMS system was deployed at Chebogue Point, Nova Scotia. The system recorded continuous data for six weeks without any major problems. The main objective of the study was to analyze processed pollution plumes advected off of the coastline of the northeastern United States and carried over the Bay of Fundy. In a preliminary analysis, the light scattering signals were analyzed for two plumes of different chemical composition with potential to discern a difference in density between the two plumes.

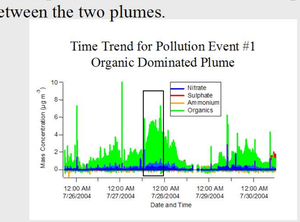

Figure 6a-1.

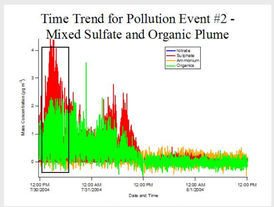

Figure 6a-2.

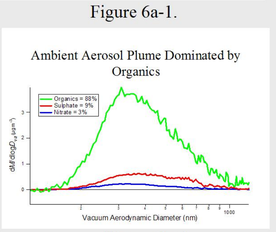

Figure 6b-1.

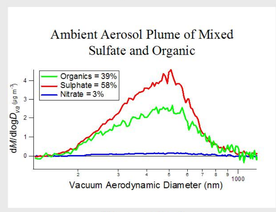

Figure 6b-2.

Figures 6a and 6b show the difference in the chemical composition for the two different pollution plumes. Plume #1 is dominated by organic species and plume #2 is a mixture of organic and sulfate aerosol. From 6b-2, one can see that the organic and sulfate size modes track one another indicating that the aerosols are most likely internally mixed within this plume. We explore this assumption further in figure 6d by plotting the distribution of light scattering signals from specific m/z's for sulfate and organic species. Plotting the light scattering signals for each of the plumes against Dva in figure 6c below shows that there is a slight shift due the difference in plume densities.

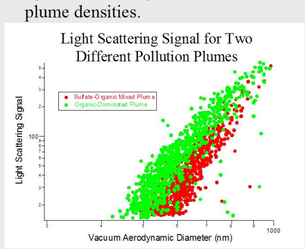

Figure 6c. Shift According to Difference in Density of the Two Plumes

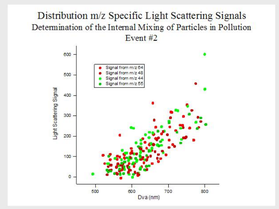

Figure 6d. Internal Mixing of Pollution Plume #2

Summary

The combined light scattering-AMS system is capable of distinguishing laboratory and ambient aerosols of different densities. The light scattering signals from laboratory distributions of oleic acid and ammonium nitrate show that the signal intensity is dependent upon the mobility diameter of the particles with differences in refractive index having little effect. Therefore, when the light scattering signals of two different aerosols plumes are plotted against vacuum aerodynamic diameter, they will be separated by their difference in density. Adding this density dimension to the quantitative information available from the AMS, will aid in our understanding of how the aerosols were formed and processed in the atmosphere.

Future Work

-Improve the detection efficiency of the light scattering system by optimizing our baffle arrangements within the inlet.

-Obtain maximum % coverage of the laser on the oven.

-Continue analysis of Chebogue Point data set and refine our calibration curve so that we can calculate estimates of the density for different pollution plumes. Use the light scattering signals to measure the extent of internal or external mixing within particles. Obtain information about the number of refractory vs. nonrefractory particles transmitted through the AMS.