APPLICATION NOTE

NIKIRA LABS – AN06 - OEA

App Note Series: Optical Extinction Analyzers

Measurement of the Complex Refractive Indices and Single Scattering Albedo of Post-Combustion Indoor Aerosol Samples

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Introduction

A major component of the aerosols' climatic effect is due to their scattering and absorption of solar radiation, which are governed by their optical and physical properties. However, the accurate assessment of this effect on global warming, climate change, and air quality is made difficult due to uncertainties in the calculation of single scattering albedo (SSA). Experimental complications arise in the determination of the single scattering albedo of an aerosol particle since it requires the simultaneous measurement of both the scattering and extinction. Despite substantial advances in optical property measurements over the years, there are still several challenges associated with measuring each of these components accurately, which propagate into large uncertainties in the reported SSA values that are incorporated into climate models. In fact, aerosol optical absorption, in particular, is a difficult measurement to perform, and it's often associated with large uncertainties when using filter methods or difference methods. This highlights the need for commercialization of novel instrumentation that is capable of accurate measurements with short instrument response times (~ 1 Hz) in order to monitor fast variations in the ambient aerosols' chemical and spatial makeup.

In this application note, we demonstrate the use of a new Optical Extinction Analyzer (OEA) in conjunction with a nephelometer and two particle sizers, emphasizing the benefits that co-employment of the OEA offers to derive the complex refractive index of aerosols and their single scattering albedo parameter.

Experimental Setup

For the purpose of our current study, we have staged a controlled combustion event in order to generate aerosols. We have then employed a novel Nikira Labs Optical Extinction Analyzer (OEA) to measure the aerosols' total optical extinction at 520nm and an Ecotech Aurora 3000 nephelometer to measure the scattering at 525 nm. To analyze the size distribution of the combustion-generated ambient aerosols, we have employed a scanning mobility particle sizer (SMPS) composed of a TSI model 3080 electrostatic classifier coupled to a TSI model 3782 water-based particle condensation counter to measure the small particle ($0.022 < d < 1.0$ um) size distributions, and a TSI 3321 aerodynamic particle sizer (APS) to measure the aerodynamic size distribution of larger particles ($1.0 < d < 20.0$ um).

Results & Discussions

The optical scattering and extinction measurements are shown in Figure 1. At time t = 2000 s, a stick of incense was lit inside the laboratory and was allowed to burn for ~ 13 minutes before it was put out at t = 2800 s. The optical scattering and extinction as well as the particle size distributions were measured before and after the combustion event, and the difference in aerosol populations at times t = 0 and t = 3600 s is assigned to the particles generated from burning the incense.

(See figures on the next page)

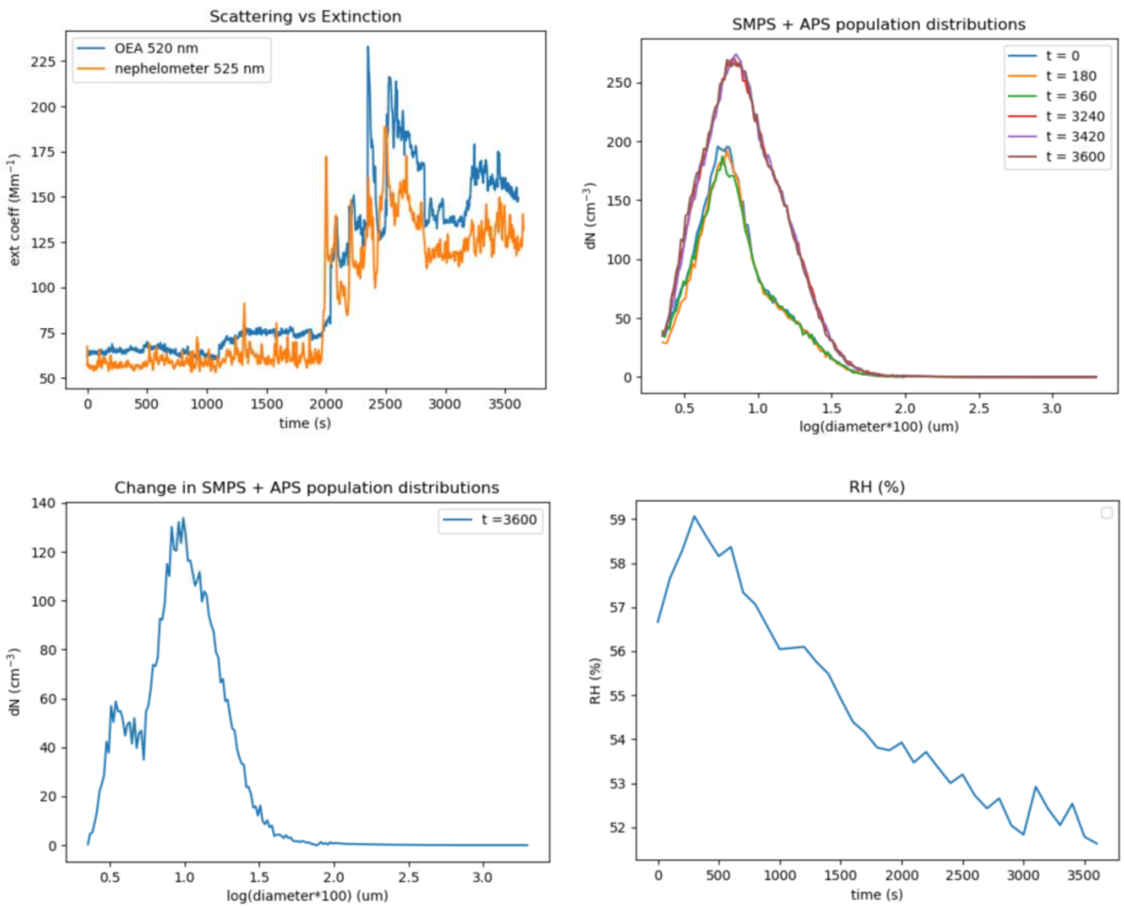


Figure 1. **Top left:** Optical scattering and extinction values measured indoors before and after lighting an incense stick. **Top right:** Aerosol size distributions measured by an SMPS and an APS. **Bottom left**: The difference in aerosol populations measured at t = 0 and t = 3600 s. **Bottom right:** The relative humidity in the laboratory measured at the OEA.

The values of the optical extinction and scattering before and after the combustion event and their differences are tabulated below in Table 1. These values are used together with the size distribution of the two populations to calculate the complex refractive index of the smoke particles.

Table 1: Optical Extinction & Scattering Measurements Pre- & Post-Combustion

t	Extinction (Mm^{-1})	Scattering (Mm^{-1})
0 s	65.3	57.3
3600 s	157.5	126.0
Difference (3600 s - 0 s)	92.2	68.7

Complex Refractive Index

The optical extinction coefficient associated with the particle size distribution is calculated using the Mie Theory-based Python library miepython [1]. Briefly, the optical extinction coefficient associated with each particle diameter bin is calculated, then the product of the calculated extinction coefficients and the measured populations is summed over the entire distribution, with the complex refractive index taken as an adjustable parameter that is varied to fit the measured values of extinction and scattering. The best fit to the measured scattering and extinction values is found using a value of the refractive index of n = 1.437 – 0.028i.

The complex refractive index is an intrinsic property of a material that depends on the sample composition. For aerosols generated from burning carbon-based fuel sources, the expected composition of the particles is a mixture of elemental carbon (EC), organic carbon (OC), sulfates, nitrates, and water. The amount of absorbed water depends on both the relative humidity (RH) and the volume fraction of water soluble species (f_s) present in the particles [2]. The most abundant water-soluble species in smoke aerosols are sulfates, with typical reported volume fractions of 0.05[3,4]. Using the thermodynamic relationship between RH, f_s, and the extent of particle hydration presented in reference 2, the calculated volume fraction of water in the aerosols measured here is ~ 1%. The particle composition is therefore mainly EC and OC, and the complex refractive index can be reasonably approximated as the volume fraction-weighted sum of the two materials, given in equation 2.

$$n = \sum_i f_i * n_i \qquad \text{Equation 1}$$

$$n \sim f_{OC} * n_{OC} + f_{EC} * n_{EC} \qquad \text{Equation 2}$$

Using n = 1.87 and k = 0.22 for EC and n = 1.40 and k = 0.0 for OC [3], the fractions of EC and OC present in the sampled aerosol are 0.1 and 0.9, respectively. This corresponds to an OC/EC ratio of 9 which is consistent with the findings of Wang *et al.* regarding the OC/EC ratio range of traditional and aromatic incenses [5]. It is a reasonable first approximation for the degree of graphitization achieved by the combustion conditions investigated in this experiment. A more accurate estimation of the degree of graphitization of the particles would be obtained by using filter-based methods that directly measure particle composition in conjuction with optical measurements.

Single Scattering Albedo

The Single Scattering Albedo (SSA) of the generated aerosols can be calculated from the ratio of the scattering efficiency to total extinction efficiency, as tabulated in Table 1. The aerosols generated by the incense combustion event investigated here yield an SSA of 0.75.

Further Work

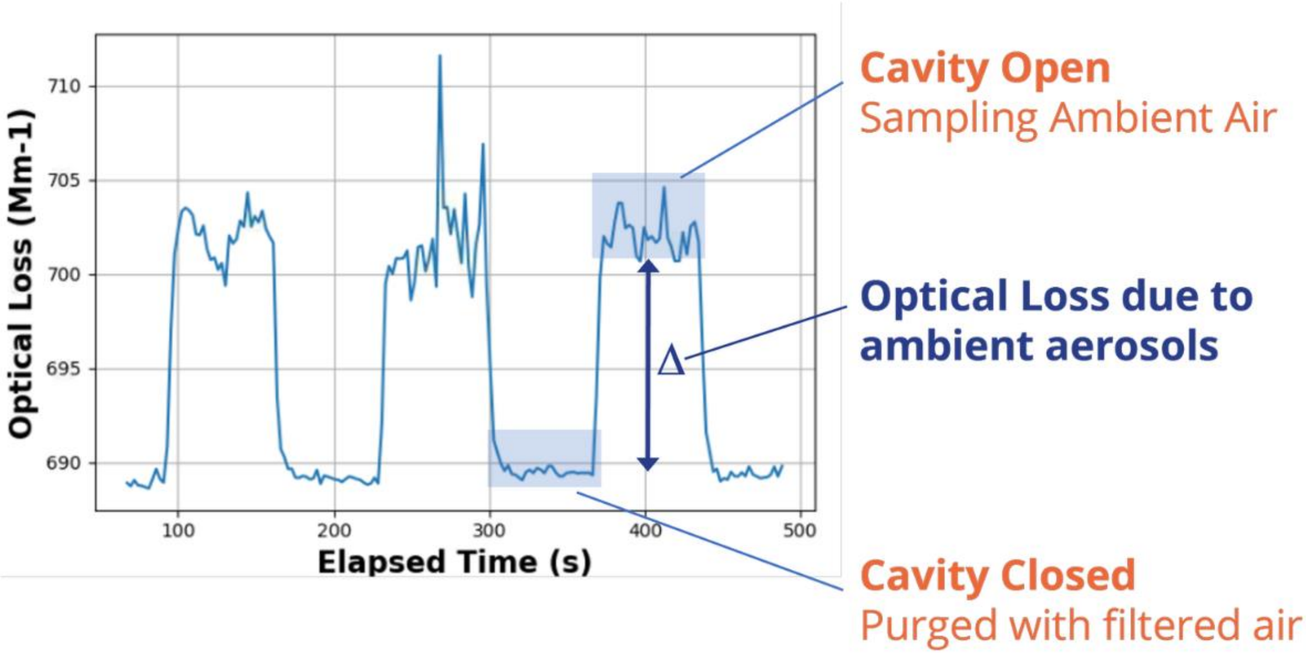
We envision that future experiments would focus on varying the combustion conditions (temperature, pressure, fuel source, and oxygen concentration) and measuring the change in complex refractive index and SSA of the generated aerosol.

References

1. https://miepython.readthedocs.io/enlatest/index.html
2. Hagen, D. E., Trueblood, M. B., White, D. R. Hydration Properties of Combustion Aerosols. *Aerosol Science and Technology,* **10**. (1989). 63-69.
3. Sloane, C.S. Effect of composition on aerosol light scattering efficiencies. *Atmos. Environ*. **20**, (1986). 1025-1037.
4. Schkolnik, G., Chand, D., Hoffer, A., Andreae, M.O., Erlick, C., Swietlicki, E., Rudich, Y. Constraining the density and complex refractive index of elemental and organic carbon in biomass burning aerosol using optical and chemical measurements. *Atmospheric Environment,* **41**. (2007). 1107-1118.
5. Wang, B., Lee, S.C. and Ho, K.F., 2006. Chemical composition of fine particles from incense burning in a large environmental chamber. Atmospheric environment, 40(40), pp.7858-7868.

How Does the OEA Work?

1. Ambient air is pulled through a duct by fans at a speed of ~1 m/s.
2. Open-path cavity ringdown spectroscopy is used to make a direct measurement of the optical extinction coefficient (beta) in the sample.
3. The duct is periodically closed, and the cell is purged with filtered air to perform a background measurement.
4. The difference between the open and closed duct values provides a direct, calibration-free measurement of the aerosol optical extinction.



The OEA Harnesses 3 Technologies: