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Estimation of aerosol optical properties considering hygroscopicity and light absorption

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HIGHLIGHTS

- Influence of the water solubility and light absorbing of organic aerosol investigated.
- Size resolved model developed by combining aerosol hygroscopic growth and dynamics.
- Mass absorption efficiency(MAE) of Water Soluble Organic Carbon explain 5–10% of EC.
- Water Soluble Organic Carbon enhances absorption by increasing imaginary refractivity.

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ABSTRACT

In this study, the influences of water solubility and light absorption on the optical properties of organic aerosols were investigated. A size-resolved model for calculating optical properties was developed by combining thermodynamic hygroscopic growth and aerosol dynamics models. The internal mixtures based on the homogeneous and core–shell mixing were compared. The results showed that the radiative forcing (RF) of Water Soluble Organic Carbon (WSOC) aerosol can be estimated to range from −0.07 to −0.49 W/m^2 for core–shell mixing and from −0.09 to −0.47 W/m^2 for homogeneous mixing under the simulation conditions (RH = 60%). The light absorption properties of WSOC showed the mass absorption efficiency (MAE) of WSOC can be estimated 0.43–0.5 m^2/g, which accounts for 5–10% of the MAE of elemental carbon (EC). The effect on MAE of increasing the imaginary refractive index of WSOC was also calculated, and it was found that increasing the imaginary refractive index by 0.001i enhanced WSOC aerosol absorption by approximately 0.02 m^2/g. Finally, the sensitivity test results revealed that changes in the fine mode fraction (FMF) and in the geometric mean diameter of the accumulation mode play important roles in estimating RF during hygroscopic growth.

1. Introduction

Aerosol optical properties are dependent on the size distribution, shape, chemical composition, and mixing state of the aerosol, which are strong functions of relative humidity (RH) (Pilinis et al., 1995). The main characteristics of an aerosol that influence its optical properties are scattering, light absorption, and hygroscopicity. The hygroscopic growth of an aerosol is closely related to the chemical composition and size distribution of its particles. For example, a water-soluble aerosol can absorb water, resulting in hygroscopic growth at a the degree f which being determined by RH and the composition of the aerosol as well as the size distribution of its particles, properties that change as the particles grow. These changes in the physical and chemical characteristics of the aerosol can subsequently change its optical properties. Usually, aerosol scattering and hygroscopicity are related to the real refractive index and negative radiative forcing (RF). On the other hand, aerosol light absorption is related to the imaginary refractive index and positive RF. Atmospheric aging processes can change the physical and chemical properties of these aerosol particles (such as their hygroscopicity) and influence their environmental effects. The focus of this study is the uncertainty in global aerosol RF, with a particular focus on organic aerosols.

An organic aerosol (OA) is either emitted as primary aerosol

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Table 1
Sampling information and target compounds analyzed in this study.

Sampling period	2006. 09.–2007. 08.
Sampling site	Yeongeon-dong, Jongno-gu, Seoul, Korea
Sampling frequency	Every 6th day
Particle size sampled	PM_{10} (Particle collected during 24 h)
Analyzed Species	Particle: Na^+, SO_4^{2-}, NH_4^+, Cl^-, NO_3^- Gas: NH_3, HNO_3, SO_2 WSOC (Water Soluble Organic Carbon) OC (Organic Carbon) EC (Elemental Carbon)
Total number of samples	62 samples
Total number of samples used in this study	34 samples after QA/QC

Table 2
Summary of seasonal average chemical species concentrations for the 34 analyzed samples.

Species ($\mu g/m^3$)	Fall	Winter	Spring	Summer
Number of samples	10	16	4	4
NaCl	2.33	3.23	3.16	1.97
$(NH_4)_2SO_4$	7.84	12.67	14.04	13.67
NH_4NO_3	3.11	10.31	7.21	6.16
Total ion	13.27	26.20	24.41	21.80
EC	3.22	3.48	1.63	2.45
WSOC	2.54	7.10	4.69	4.30
WISOC	7.50	7.13	5.34	2.85
OM[a]	14.05	19.93	14.04	10.01
Residue	30.40	29.27	38.94	13.25
PM10 mass	60.95	78.87	79.02	47.51

Fall: Sep.–Nov. 2006, Winter: Dec. 2006–Feb. 2007, Spring: Mar.–May 2007, Summer: June–Aug. 2007.
[a] The factor of 1.4 was applied for the calculation of OM from OC (Malm et al., 2011).

particles or produced as secondary aerosol particles from fossil fuel and biofuel burning. Of the organic carbon (OC) fraction of OA, a non-negligible portion is known to be water-soluble (Duarte et al., 2007). However, it is difficult to quantify the effect of Water Soluble Organic Carbon (WSOC), especially as a function of RH. Another important characteristic to be considered in the optical properties of WSOC is its light absorption. In this study, we focused primarily on the influence of water solubility on the optical properties of polydispersed aerosols and the role of WSOC on light-absorbing optical properties.

We developed a size-resolved model for calculating the optical properties of organic aerosols by combining thermodynamic hygroscopic growth and aerosol dynamics models (Jung et al., 2004). Then, we applied the model to the aerosol measurement data at a megacity in northeast Asia, Seoul. By doing so, we reported (1) how were the estimated seasonal optical properties based on the measurements, (2) what were the important variables that determined the optical properties of the aerosols over Seoul, and (3) how were the sensitivities of those variables to the water uptake or growth factor and optical properties.

Table 3
Refractive indices at a wavelength of 550 nm and the densities for the various aerosol components (Sloane, 1984; Arola et al., 2011).

Composition	Refractive index (550 nm)	Density (g/cm^3)
EC	1.7–0.48i	1.7
NaCl	1.51	2.165
$(NH_4)_2SO_4$	1.52	1.77
NH_4NO_3	1.55	1.72
WSOC	1.53–0.0136i	1.3
WISOC	1.53–0.1058i	1.3
Residue	1.62	2.3
Water	1.33	1

2. Measurement

Sampling of particulate matter with a nominal aerodynamic diameter of less than or equal to 10 μm (PM_{10}) was performed in Seoul, a representative urban site in Korea. The measurement site was located on the roof (~17 m above ground, 37.5 °N, 127.00 °E) of the former School of Public Health building at Seoul National University, a mixed commercial and residential area. A detailed description of the sampling and measurement methods has previously been reported (Heo et al., 2009). The sampling period was between August 2006 and August 2007.

Briefly, PM_{10} samples were collected for 24 h on every sixth day using a three-channel system consisting of one channel annular denuder and two channel filter packs (URG, USA), similar to the US EPA Compendium Method IO 4.2 (US EPA, 1999). The flow rate of sampling was 16.7 L/min and was monitored for each channel using independent dry gas meters. One channel was used to measure carbonaceous aerosols. A quartz filter prebaked at 550 °C for 10 h in a furnace was used to analyze the OC, elemental carbon (EC), and WSOC. OC and EC were analyzed using the NIOSH thermal/optical transmittance (TOT) method (NIOSH, 1999; Birch and Cary, 1996). Half of the 47-mm quartz filter was extracted by ultrasonification with 30 mL deionized water twice for 30 min to quantify WSOC. Ice water was used in the sonication bath to minimize the possible loss of volatile OC due to temperature increase during extraction. Filter debris and suspended insoluble particles were removed from the extracts using a syringe filter (0.2-μm PTFE membrane, PALL

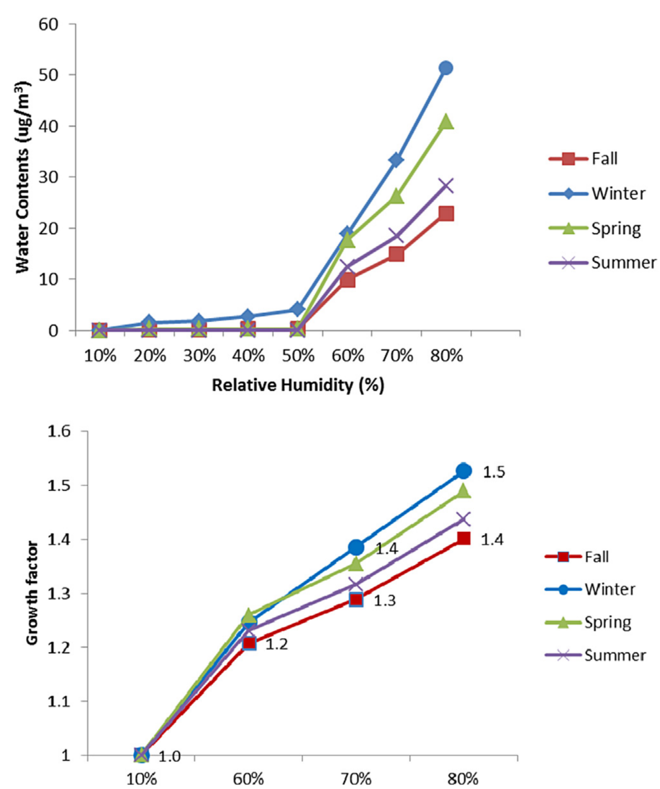

Fig. 1. Water content and growth factor results from the SCAPE2 thermodynamic equilibrium model as a function of relative humidity (d_{gv1} = 0.3 μm, d_{gv2} = 2.5 μm, σ_{g1} = 2.5, σ_{g2} = 1.7).