



Cloud Condensation Nuclei Activation of Internally Mixed Black Carbon Particles

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ABSTRACT

Cloud condensation nuclei (CCN) activation of black carbon (BC) particles coated with different amounts of inorganic (sulfuric acid) and organic (levoglucosan or succinic acid) compounds was investigated. An evaporation-condensation method was used to produce coated BC particles, and the volume fractions of coating species were measured using the tandem differential mobility analyzer method. Furthermore, the CCN activation of the coated BC particles was measured using a differential mobility analyzer-CCN counter method. Coating with both aforementioned compounds engendered the activation of the BC particles as CCN at a supersaturation of 0.5%. Sulfuric acid coating was associated with the highest CCN activation fraction (= CCN/condensation nuclei), followed by levoglucosan coating and then succinic acid coating; this indicates that the water solubility of the coating compounds played a crucial role in the CCN activation of the coated BC particles. In general, the CCN activation fraction of the coated BC particles increased with the volume fraction of the coating species, but the manner of increase differed with the coating compounds and generation method. A premixed solution method was also used to produce mixed BC particles, and among the coating compounds, sulfuric acid was associated with the highest CCN activation fraction. For a given volume fraction, sulfuric-acid-coated BC particles produced using the premixed solution method showed a higher CCN activation fraction compared with those obtained using the evaporation-condensation method, and this was due to the presence of residual water in the sulfuric-acid-coated BC particles produced using the premixed solution method.

Keywords: TDMA; Water solubility; CCN activation fraction; Coating.

INTRODUCTION

Atmospheric aerosols, depending on their size and chemical composition, can be activated into cloud condensation nuclei (CCN) that provide a surface for water vapor condensation and facilitate cloud droplet nucleation under atmospheric supersaturation conditions (Cruz and Pandis, 1997; Martin, 2000). These particles indirectly influence the earth's climate because they affect the formation, persistence, and optical properties of clouds in the atmosphere (Twomey, 1991; Lohmann and Fletcher, 2005; Denman *et al.*, 2007; Solomon, 2007). When the number concentration of CCN increases, the size of cloud droplets may decrease, leading to an increase in the lifetime of the cloud droplets (Albrecht, 1989). Leitch *et al.* (1992) reported that in the lower atmosphere, an increase in the CCN number under polluted conditions caused the size of cloud droplets to decrease. Furthermore, the cloud droplets can interfere with incoming solar radiation as well as reflected terrestrial

radiation. Therefore, understanding how atmospheric aerosols serve as CCN is imperative.

Black carbon (BC) particles in the atmosphere are considered to be the second largest contributor to anthropogenic radiative forcing (light absorption) after carbon dioxide because of their strong capability to absorb visible solar radiation (Jacobson, 2001). They are produced by a variety of anthropogenic and natural sources, including incomplete combustion of fossil fuels, biomass burning, forest fires, traffic exhaust, and industrial emissions (Jacobson, 2001; Schulz *et al.*, 2006; Ramanathan and Carmichael, 2008; Jacobson, 2010; Bond *et al.*, 2013; Seinfeld and Pandis, 2016). Newly generated BC particles typically exist in the form of agglomerates that comprise hydrophobic primary spherules, and therefore, they are less likely to become CCN under atmospheric supersaturation conditions (Zhang *et al.*, 2008). However, the BC particles can undergo several atmospheric aging processes; for example, they can adsorb gaseous species, allow gaseous species to condense on them, and coagulate with preexisting aerosol particles (Saathoff *et al.*, 2003; Zhang *et al.*, 2005; Andreae and Rosenfeld, 2008). BC particles with an open structure (i.e., an agglomerated shape) can be converted into a compact structure (more spherical), and they can appear as internally

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mixed particles, which normally have a core-shell structure (a solid core partially or entirely coated with various condensable compounds) (Novakov and Corrigan, 1996; Dusek *et al.*, 2006; Henning *et al.*, 2010). Therefore, the morphological and chemical properties of BC particles can change during aging processes. A study reported that internally mixed BC particles cause higher positive direct radiative forcing than externally mixed particles do (Li *et al.*, 2005). In addition, the hygroscopicity of aged BC particles may increase and considerably influence their cloud-forming potential (Dusek *et al.*, 2006; Zhang *et al.*, 2008). The cloud forming properties of internally mixed BC particles in the atmosphere are difficult to assess because of the complexity of the coating species, interaction between inorganic and organic aerosol components, and shape variation among the particles (Saxena *et al.*, 1995; Ji *et al.*, 1998; Zhang *et al.*, 2008).

Numerous studies have investigated the CCN activity of aged and coated BC particles generated from different emission sources. A previous study showed that although BC particles (i.e., thermal soot particles produced by pyrolysis of natural gas) did not show CCN activity, oxidized and compact BC particles exhibited CCN activity (Koehler *et al.*, 2009). Another study found that BC particles generated at the flaming stage of wood combustion were not activated to CCN at a supersaturation of 1% (Hallett *et al.*, 1989). Dusek *et al.* (2006) showed that the CCN activation of internally mixed BC particles varied with the type of coating compounds. For example, the coating of BC particles by sodium chloride and hexadecanol enhanced and suppressed the CCN activation of the BC particles, respectively. Other studies have revealed that fresh soot particles in diesel exhaust do not show CCN activity, but aging the BC particles by condensing secondary organic species on them resulted in an increase in their CCN activity (Tritscher *et al.*, 2011). Furutani *et al.* (2008) showed that a distinct relationship exists between the extent of anthropogenic aging of BC particles and their CCN activity, and that the most highly aged air masses were associated with BC particles showing the highest CCN activity. Lammel and Novakov (1995) observed that the soluble fraction of BC particles affected their nucleation capability (i.e., the CCN activity of BC particles increases with the soluble mass fraction). Coated BC particles are expected to have different amounts of coating compounds, which result in a wide range of CCN activity (Varutbangkul *et al.*, 2006; Zelenyuk *et al.*, 2007). Henning *et al.* (2010) determined the relationship between droplet activation and the total number of moles of coating material per particle, as derived from aerosol mass spectrometer measurements, and reported that more soluble materials on particles indicated easier activation of the particles as CCN. Khalizov *et al.* (2009) showed that agglomerated soot particles with a mobility size of approximately 150 nm and heavily coated with sulfuric acid had lower critical supersaturation compared with lightly coated particles of the same mobility size (0.18% versus 0.42%). Although previous studies have provided useful information on the CCN activity of mixed BC particles, it is necessary to acquire more data for expanding the CCN

activity database of internally mixed BC particles coated with various types and amounts of compounds.

This study investigated the CCN activation of laboratory-generated BC particles (as internal mixtures) coated with different amounts of inorganic and organic compounds. Synthesized core-shell structure particles were used to simulate the aging process of freshly emitted BC particles in the atmosphere. The BC particles were coated using a silicone oil bath vaporizer. BC particles of a certain size were mixed with vapor of the coating compounds evaporated from the vaporizer, and the vapor condensed onto the particles upon cooling to room temperature (i.e., the evaporation-condensation method). The amount of coating compounds was expressed as a volume fraction, and it was controlled and measured by using the tandem differential mobility analyzer (TDMA) method (Rader and McMurry 1986; Park *et al.*, 2008; Khalizov *et al.*, 2009; Shila *et al.*, 2014). Mixed BC particles with various weight fractions (2.5%–15.0%) of coating compounds were also produced through direct atomization of premixed solutions of BC. The differential mobility analyzer (DMA)-CCN counter method (Park *et al.*, 2011) was used to determine the CCN activity of size-resolved particles at a designated supersaturation. Three types of chemical compounds were used as coating compounds: sulfuric acid (as an inorganic compound), which is often involved in particle nucleation and growth (Bernd *et al.*, 2005; Kulmala *et al.*, 2006); levoglucosan (as an organic acid compound), which is known as a tracer for biomass combustion aerosols (Mochida and Kawamura, 2004; Dixon and Baltzell, 2006; Hoffmann *et al.*, 2009); and succinic acid (as an organic acid compound), which is often found in secondary organic aerosols (Chebbi and Carlier, 1996; Kerminen *et al.*, 2000; Vanhanen *et al.*, 2008). In addition, transmission electron microscopy (TEM)/energy dispersive spectroscopy (EDS) was employed to investigate the morphology and elemental composition of the coated BC particles.

EXPERIMENTAL

As shown in Fig. 1, coated BC particles were generated using a single-jet atomizer (Model 9302, TSI Inc., USA), DMA, and silicone oil bath vaporizer coating system. The atomizer generated pure BC particles from a liquid suspension of BC powder (carbon black N220, intermediate super abrasion furnace, Poland). The BC powder was heated at 400°C for several hours to remove any adsorbed semivolatile impurities, and approximately 0.1 g of the heated powder was added to 100 mL of a mixture of 20% reagent-grade 2-propanol (99.9% purity, Junsei Chemical, USA) and 80% ultrapure water (TKA GenPure, Thermo Scientific, Germany). The mixture was placed in an ultrasonic bath for 20 min and then placed over a magnetic stirrer to obtain a liquid suspension, which was dispersed by the atomizer (Dusek *et al.*, 2006). The polydisperse particles passed through two dilution chambers before being dried to approximately 6% relative humidity (RH) in two diffusion dryers with 53.5-cm-long outer tubes containing silica gel. The dried particles were introduced into a heated tube

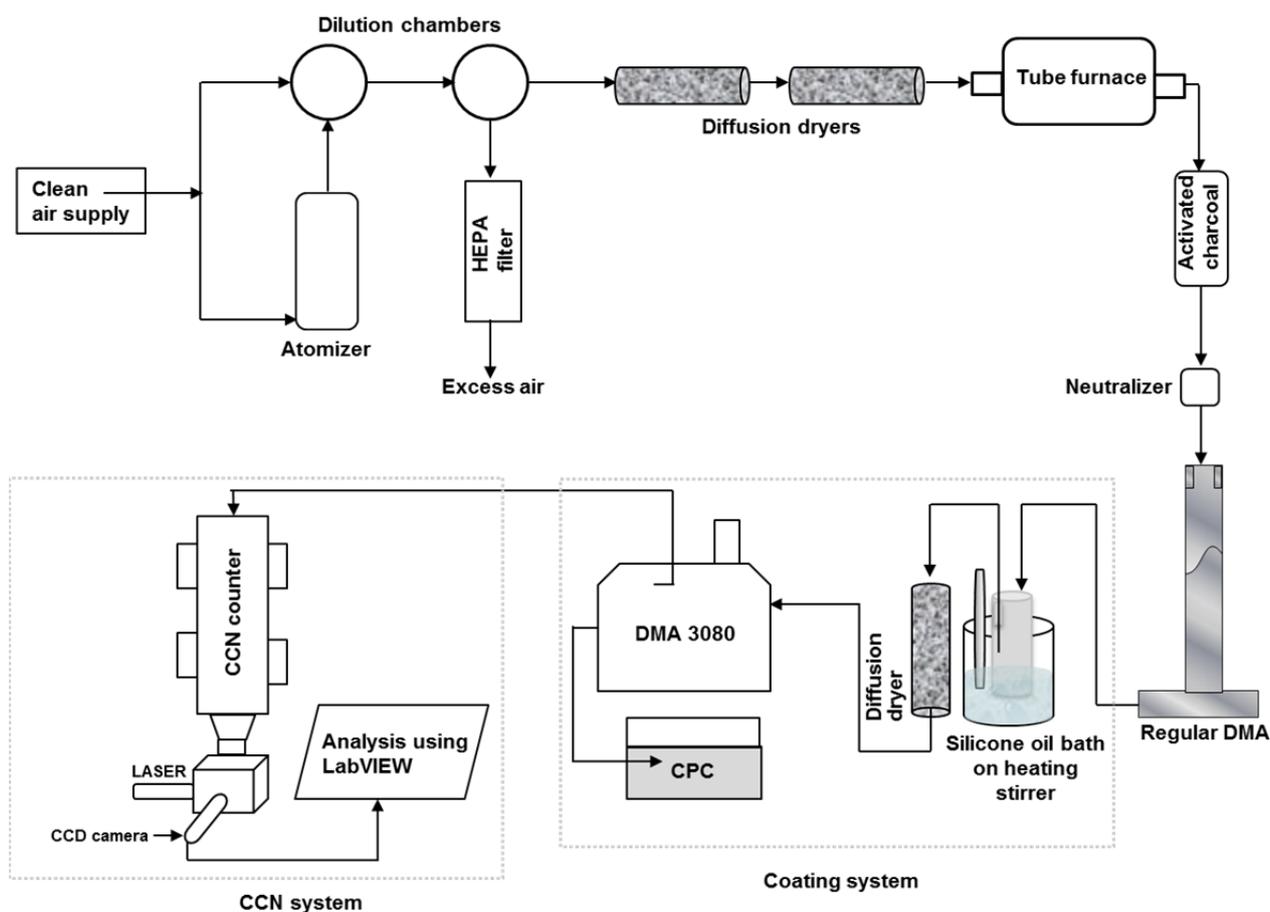


Fig. 1. Schematic of the experimental setup.

furnace ($\approx 400^\circ\text{C}$) (Linderberg, Thermo Electron Corp., USA) and passed through an activated charcoal dryer for removing semivolatile/volatile materials. Subsequently, the particles were neutralized using a bipolar aerosol neutralizer (^{241}Am) and then introduced into the DMA. The particle size distribution was measured using a scanning mobility particle sizer consisting of a DMA (Model 3081, TSI Inc., USA) and condensation particle counter (CPC) (Model 3022A, TSI Inc., USA). Particles with a mode mobility diameter were selected by the first DMA in the TDMA system (i.e., monodisperse aerosol) and introduced into the silicone oil bath vaporizer in which a hard glass test tube was partially filled with pure sulfuric acid (95%–98% purity, Sigma-Aldrich, USA), 0.1 wt% aqueous solution of an organic compounds (i.e., succinic acid, 99.0% purity, Sigma-Aldrich, USA), or 0.1 wt% aqueous solution of levoglucosan (99% purity, Sigma-Aldrich, USA) (Moteki and Kondo, 2007), depending on the required coating liquid. The coating liquid was evaporated by heating, and the vapor was entrained in the aerosol flow; subsequently, the vapor condensed onto the particles after it was cooled to room temperature, leading to the formation of coated particles (i.e., core-shell particles). The residence time for particles in the vaporizer is 4.8 sec. A diffusion dryer with silica gel was used to remove water vapor after the vaporization of the coating liquid. To ensure a uniform distribution of heat

in the silicone oil bath, a heating/stirring plate (StableTemp, Cole-Parmer, USA) was used. The oil bath temperature was controlled to coat size-selected BC particles with different amounts of condensable compounds.

The TDMA method can be used to estimate the amount of coating compounds by deriving the difference between the mobility size before and after coating. The volume fraction of the coating compounds in the internally mixed BC particles was calculated by assuming the BC particles to be spherical. Coated particles were selected using the second DMA in the TDMA system before being sent to the CCN counter for the measurement of their CCN activity. Residence time of coated particles before being counted was approximately 240 ms in the second DMA. T and RH after the second DMA was 25°C and 20%, respectively. BC particles examined by TEM showed compact agglomerates comprising many individual BC particles, which in turn contained primary spherules with diameters typically ranging from 10 to 50 nm. The BC particles became more spherical after being coated. Although the morphological change during coating was not large (uncoated BC particles are compact agglomerates), it could cause the values of the volume fraction of the coating species to differ from those calculated from the mobility size change under the assumption that the particles are spherical before and after being coated (Zhang *et al.*, 2008). Note that it is difficult to

measure the volume of non-spherical particles. The real volume fraction of the coating compounds obtained by assuming that the BC particles are in the form of a cluster before coating and that they assume a spherical shape after coating should be considerably higher than the value calculated by assuming the particles to always be spherical. Therefore, the real volume fractions should be higher than the values presented here.

In addition to the evaporation-condensation method, mixed BC particles were also produced from premixed solutions. In this method, mixed BC particles were generated through the direct atomization of premixed solutions containing BC powder and specific weight fractions (2.5%–15.0%) of the coating compounds. The aerosolized BC particles were assumed to be a homogeneous mixture with their mass fraction being identical to that in the premixed solution (Brooks *et al.*, 2002). The mixed BC particles were selected using the DMA, and they were then sent to the CCN counter for the measurement of their CCN activity.

Size-selected uncoated or coated BC particles were introduced into the CPC and CCN counter for determining the number concentrations of condensation nuclei (CN) and CCN, respectively. A detailed description of the CCN counter was provided by Park *et al.* (2011). Briefly, the CCN counter is a continuous-flow streamwise thermal-gradient-type CCN counter, and it mainly consists of a sheath air focusing system, supersaturator, laser, and charge-coupled device (CCD) camera detector. The CCN counter maintains a controlled and precise supersaturation within a column by sustaining a temperature difference between the top and bottom of the column. When particles passing through the centerline of the column are exposed to this supersaturation, they can absorb water vapor and become activated to form droplets, which depends on their size and chemical composition. The measurement of CCN activation was performed at a constant supersaturation. To measure the number concentration of activated particles, a 633-nm helium–neon laser (18 mW) (LGK 7654-8, LASOS, Germany) and CCD camera (1394 × 1040, 17 fps) (scA1390-17, Basler, Germany) were used. The laser beam was adjusted using an XY stage controller with respect to the focal point of the CCD camera and aerosol beam. In this study, the particles were activated in a temperature gradient of 5 K, which provides a supersaturation of approximately 0.5%. The supersaturation was determined by using standard NaCl and (NH₄)₂SO₄ particles in our previous study (Park *et al.*, 2011). Warner (1968) reported that the ambient supersaturation should typically be less than 1% and should not exceed 2%. The activation fraction was obtained by dividing the number of particles acting as CCN (measured by the CCN counter at a supersaturation of 0.5%) by the total number of CN counted by the CPC (*i.e.*, CCN/CN).

For morphological and elemental analysis of the BC particles, particles of a certain size were selected by the DMA, and they were subsequently collected on a TEM grid (Formvar/carbon-coated 200-mesh Cu grid, 30–50 nm thick, Ted Pella Inc., USA) using a nanometer aerosol sampler (Model 3089, TSI Inc., USA). Each TEM grid was placed in a polystyrene petri dish and stored in a desiccator

for equilibration at a constant temperature of 25°C and 20% RH. The morphology and elemental composition of the collected particles were analyzed using a 200-kV field emission analytical transmission electron microscope (JEM-2100, JEOL, Japan). The beam current and magnification were 102 μA and 25,000–50,000, respectively.

RESULTS AND DISCUSSION

The amount of coating compounds on the mixed BC particles at various silicone oil bath temperatures was measured using the TDMA method. Size distributions of the size-selected particles in the first DMA at oil bath temperatures of 20°C (before coating) and 40°C (after coating with sulfuric acid) are shown in Fig. 2(a). The mode diameter of the BC particles at 20°C was 163 nm, and it was identical to the size of BC particles before entering the coating system. The mode diameter of the BC particles increased to 175 nm at the 40°C. This indicates the evaporation of sulfuric acid as well as the condensation of the vapor onto the BC particles, resulting in the formation of coated BC particles. The number size distributions of the BC particles coated with levoglucosan and succinic acid at different oil bath temperatures are presented in Figs. 2(b) and 2(c), respectively; the size distributions of the uncoated BC particles are also displayed for comparison. The mode diameter of the coated BC particles increased with the oil bath temperature. Data suggest that the amount of coating compounds on the mixed BC particles can be adjusted by controlling the silicone oil bath temperature.

For CCN activation measurements, BC particles were first coated with the appropriate compounds—sulfuric acid, levoglucosan, or succinic acid—with a volume fraction of approximately 10% by maintaining the oil bath temperature at $28 \pm 5^\circ\text{C}$, $52 \pm 5^\circ\text{C}$, or $40 \pm 5^\circ\text{C}$, respectively. Since levoglucosan has a lower vapor pressure than those of succinic acid (48.3 μPa versus 92.0 μPa at 25°C and 30°C, respectively), a higher evaporation temperature (*i.e.*, oil bath temperature) would be required for coating levoglucosan (10% volume fraction) compared with that required for coating succinic acid. Coating sulfuric acid with a volume fraction of 10% required the lowest oil bath temperature (Lide, 2004). Unlike the coating of levoglucosan and succinic acid, for the aqueous solutions (0.1 wt%) which were used in the coating system, pure sulfuric acid was used. Therefore, sulfuric acid had a considerably higher partial vapor pressure and rate of evaporation compared with the other coating compounds.

TEM measurements were performed to determine the shape of the coated BC particles. Fig. 3(a) shows a TEM image of uncoated BC particles preclassified by the DMA (163 nm). The particles are in the form of compact agglomerates and consist of small primary particles, and 2-propanol, which was used as the dispersing agent for BC powder in the aerosol generation system, could be responsible for restructuring the agglomerates into the compact form (Dusek *et al.*, 2006; Henning *et al.*, 2010). Fig. 3(b) presents a TEM image of 180-nm BC particles coated with sulfuric acid. The TEM image was obtained within a few seconds

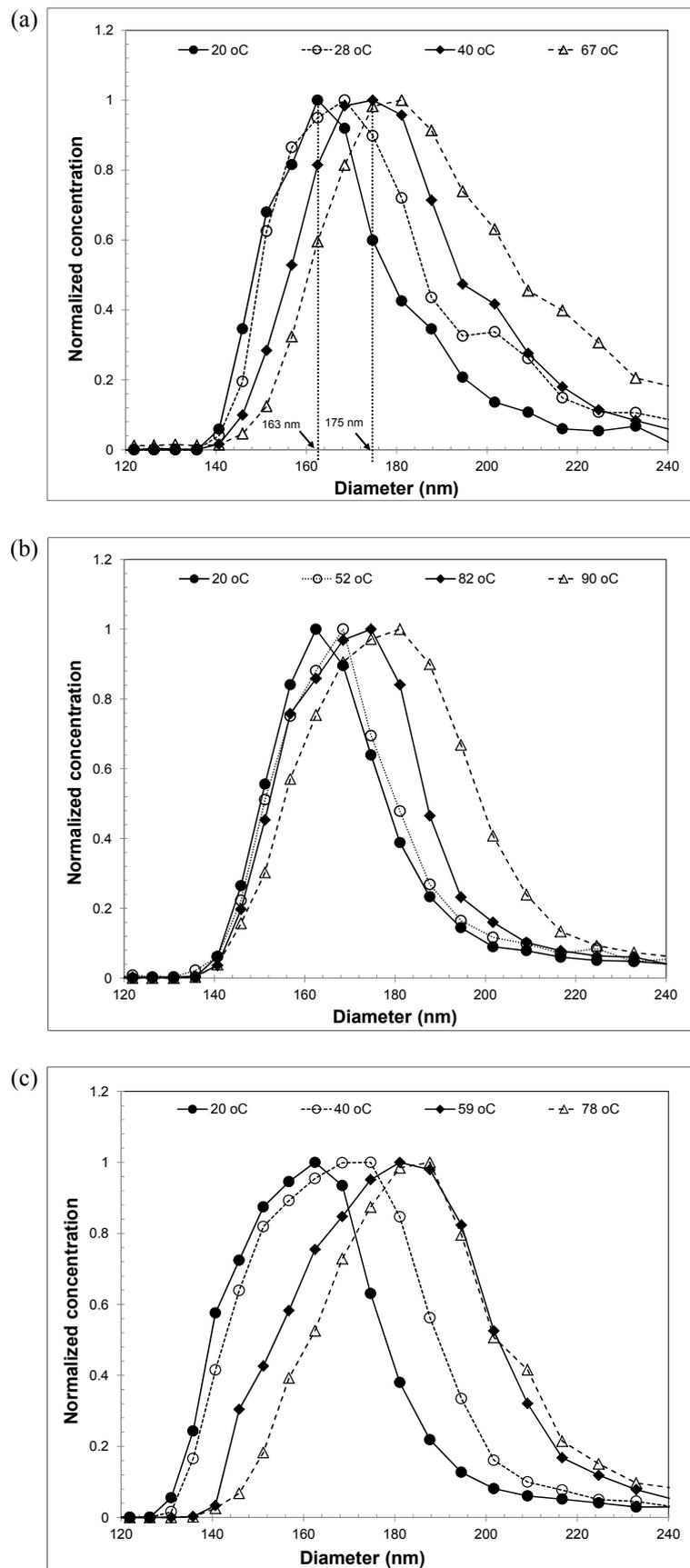


Fig. 2. Normalized number size distributions of BC particles coated with (a) sulfuric acid, (b) levoglucosan, and (c) succinic acid at different temperatures in the coating system.

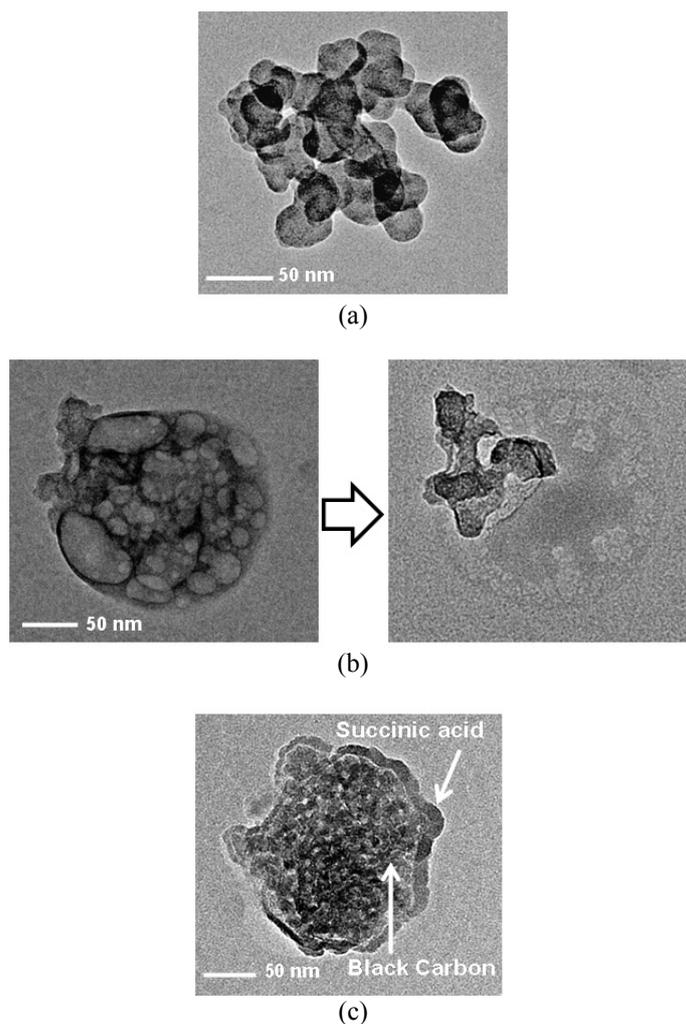


Fig. 3. TEM images of (a) uncoated BC particles, (b) BC/sulfuric acid particles with a core-shell structure and the same BC/sulfuric acid particles after their exposure to the electron beam for a long time (≈ 30 s), and (c) BC/succinic acid particles in the core-shell structure.

of exposure to the electron beam under vacuum. The BC particles became more spherical than the uncoated BC particles, which is consistent with the observations of Weingartner *et al.* (1997) and Kiselev *et al.* (2010). The agglomerated shape of the fresh BC particles (i.e., high surface-area-to-volume ratio) can provide more active sites for the condensation of the coating species compared with spherical particles (Crouzet and Marlow, 1995). A TEM image was also obtained after exposure of the sulfuric acid-coated BC particles to the electron beam for a known duration (≈ 30 s). The image shows many semitransparent areas in bright contrast inside the particles, which have internal voids, indicating that sulfuric acid evaporated rapidly upon exposing to the electron beam. A similar result has been obtained in previous studies (Weingartner *et al.*, 1997; Zuberi *et al.*, 2005; Khalizov *et al.*, 2009). Fig. 3(c) presents a TEM image of the BC particles coated with succinic acid. Although both the shell and core regions of the particles appear darker than the bare substrate, the boundary between the core and the shell is clear. The core region comprises many individual small spherules in a compact

form, suggesting that the BC particles form the core. Furthermore, the microstructure of the shell region is similar to that of amorphous carbon (Ramirez *et al.*, 1999), suggesting that succinic acid might be pyrolyzed by the strong electron beam to form amorphous carbon (Shila *et al.*, 2014). A similar structure was observed for the BC particles coated with levoglucosan.

Fig. 4 shows the CCN activation fraction (CCN/CN) of the size-selected particles (163 nm for uncoated BC particles, and 169 nm, 175 nm, 181 nm, and 188 nm for coated BC particles) at a supersaturation of 0.5% as a function of the volume fraction of the coating compounds. The CCN activation fractions of the pure BC particles (i.e., uncoated) and sulfuric acid, levoglucosan, and succinic acid particles were also measured. CCN activation was close to zero for the pure BC particles. Previous studies have reported that uncoated BC particles hardly show CCN activation (Dusek *et al.*, 2006; Henning *et al.*, 2012). However, the sulfuric acid, levoglucosan, and succinic acid particles showed CCN activation fractions of $0.995 (\pm 0.08)$, $0.995 (\pm 0.06)$, and $0.817 (\pm 0.05)$ at a supersaturation of 0.5%, respectively

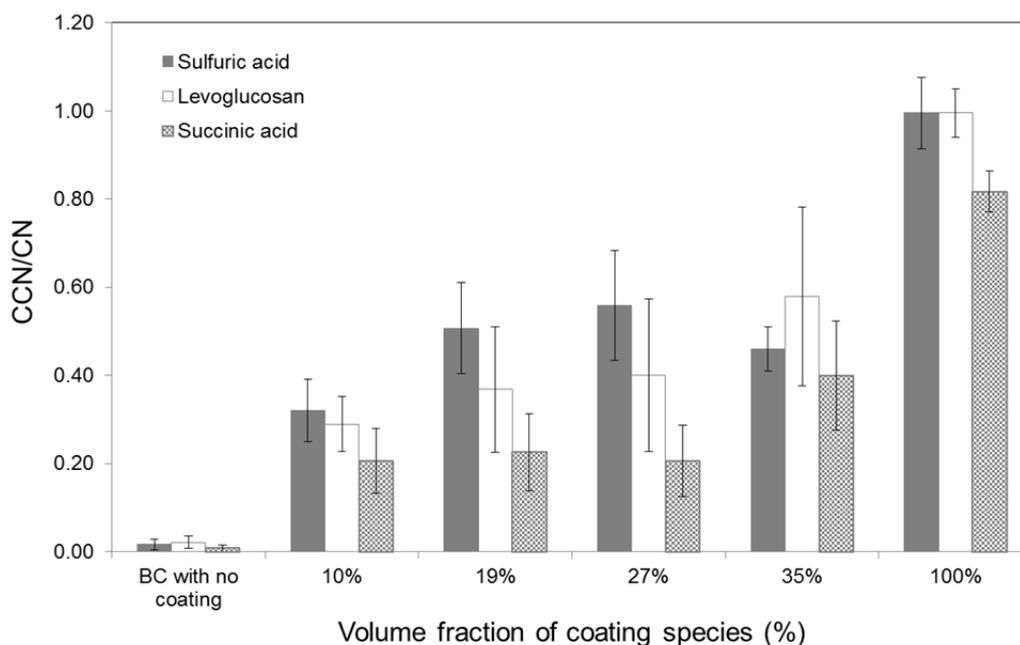


Fig. 4. CCN activation fractions of BC particles (no coating) (163 nm); BC particles coated with sulfuric acid, levoglucosan, or succinic acid (169 nm, 175 nm, 181 nm, and 188 nm); and sulfuric acid, levoglucosan, and succinic acid particles at a supersaturation of 0.5% as a function of the volume fraction of the coating species (the mixed particles were produced using the evaporation-condensation method, and the volume fraction was determined with the TDMA method).

(Fig. 4). These results are consistent with previous data for CCN activation fractions of levoglucosan particles (0.90 ± 0.10); Rosenørn *et al.*, 2006) and succinic acid particles (0.948 ; Corrigan and Vovakov, 1999).

To observe the effect of different coating compounds on the activation of BC particles, we examined the activation of the BC particles coated with sulfuric acid, levoglucosan, and succinic acid (Fig. 4). The CCN activation of the BC particles coated with sulfuric acid, levoglucosan, and succinic acid with a volume fraction of approximately 10% and at a supersaturation of 0.5% increased to approximately 0.32 ± 0.07 , 0.29 ± 0.06 , and 0.21 ± 0.07 , respectively. The data suggest that coating with a soluble species caused the BC particles to be activated as CCN. The BC particles coated with sulfuric acid showed the highest activation fraction, followed by those coated with levoglucosan and then those coated with succinic acid. Because of the higher efficiency of sulfuric acid in attracting water molecules compared with levoglucosan and succinic acid, the coated sulfuric acid could dissolve more easily to the BC/sulfuric acid particles relative to BC/levoglucosan and BC/succinic acid particles. Similarly, the higher activation fraction of the BC/levoglucosan particles relative to the BC/succinic acid particles could result from the higher water solubility of levoglucosan compared with that of succinic acid (Saxena and Hildemann, 1996; Svenningsson *et al.*, 2006; Park *et al.*, 2009; Zamora *et al.*, 2011).

The effect of the amount of coating compounds on the activation of BC particles was also examined. The activation fractions of the BC particles coated with levoglucosan with volume fractions of 10%, 19%, 27%, and 35% were 0.29 ± 0.06 , 0.37 ± 0.14 , 0.40 ± 0.17 , and 0.58 ± 0.2 , respectively

(Fig. 4). The activation fraction gradually increased with the volume fraction of levoglucosan. The activation fractions of the BC particles coated with sulfuric acid with volume fractions of 10%, 19%, 27%, and 35% were 0.32 ± 0.07 , 0.51 ± 0.10 , 0.56 ± 0.12 , and 0.46 ± 0.05 , respectively. The gradual increase in the activation fraction of the BC particles coated with sulfuric acid stopped at a volume fraction of 35% (the activation fraction somewhat decreased at the volume fraction of 35%). Trace amounts of impurities present in the BC particles coated with sulfuric acid could react with sulfuric acid to produce either an insoluble fraction or lower molecular weight products with different hygroscopic properties, and the fraction or products produced may influence the activation of the BC particles coated with a large amount of sulfuric acid (Henning *et al.*, 2012). The activation fractions of the BC particles coated with succinic acid with volume fractions of 10%, 19%, 27%, and 35% were 0.21 ± 0.07 , 0.23 ± 0.09 , 0.21 ± 0.08 , and 0.40 ± 0.12 , respectively. The CCN activation fraction of the BC particles remained fairly constant for volume fractions of 10%–27% and began to increase at the volume fraction of 35%. The reason for this is unclear. Because the calibration results of the CCN counter for sodium chloride and ammonium sulfate showed high reproducibility, instrumental bias was excluded. Henning *et al.* (2012) reported that soot (graphite soot) particles coated with succinic acid and having sizes similar to those of the succinic-acid-coated BC particles of the current study showed CCN activation fractions of 0.47–0.51 at a supersaturation of 0.5%. The difference in the CCN activation fractions between their study and the current study could be because of either the use of BC particles with different characteristics or the

condensation of different amounts of succinic acid on BC particles that was engendered by the use of different coating methods. We vaporized 0.1 wt% aqueous solution of succinic acid in an oil bath vaporizer, whereas Henning *et al.* (2012) employed a method involving the saturation of particle-free synthetic air with succinic acid; the air was heated to 100°C and then cooled to room temperature to facilitate the condensation of succinic acid on the soot particles.

A different method (premixed solution method, discussed in the Experimental section) was also used to produce mixed BC particles with varying fractions of sulfuric acid, levoglucosan, and succinic acid. The mass fractions of the coating species in aerosolized particles (2.5–15%) were assumed to be the same as their weight fractions in the premixed solutions (Brooks *et al.*, 2002). The results revealed that the BC particles mixed with sulfuric acid showed the highest activation fraction, followed by those mixed with levoglucosan and succinic acid (see Fig. 5), which is consistent with our data with volume fractions of coating species less than 27% in evaporation-condensation method. For the sulfuric-acid-coated BC particles, the volume fraction of sulfuric acid was calculated from the density and mass fraction of the sulfuric acid used for coating the particles. For similar volume fractions of sulfuric acid, the sulfuric-acid-coated BC particles produced using the premixed solution method showed higher CCN activation fractions compared with those produced with the evaporation-condensation method. The reason for this observation could be that the sulfuric-acid-coated BC particles produced with the premixed solution method contained more water (not having dried perfectly).

CONCLUSIONS

The CCN activation of BC particles coated with different amounts of inorganic (sulfuric acid) and organic (levoglucosan and succinic acid) compounds was investigated. Two methods (evaporation-condensation method and premixed solution method) were employed to produce coated BC particles, and the volume fraction of the coating species was determined using the TDMA method. The coating of a small amount of sulfuric acid, levoglucosan, or succinic acid species ($\approx 10\%$ volume fraction) caused the BC particles to be activated as CCN at a supersaturation of 0.5%. The BC particles coated with sulfuric acid showed the highest activation fraction, followed by those coated with levoglucosan and then those coated with succinic acid, regardless of the particle generation method used. The solubility of the coating species appeared to play a major role in determining the CCN activation of the BC particles. In general, a higher volume fraction of the soluble species engendered a higher CCN activation fraction; however, the manner of increase in the CCN activation fraction following an increase in the volume fraction of the coating species differed with the coating species. Furthermore, the sulfuric-acid-coated BC particles produced using the premixed solution method showed a higher CCN activation fraction compared with those generated by the evaporation-condensation method. We attribute this to the sulfuric-acid-coated BC particles produced by the premixed solution method containing residual water (the particles not having dried completely).

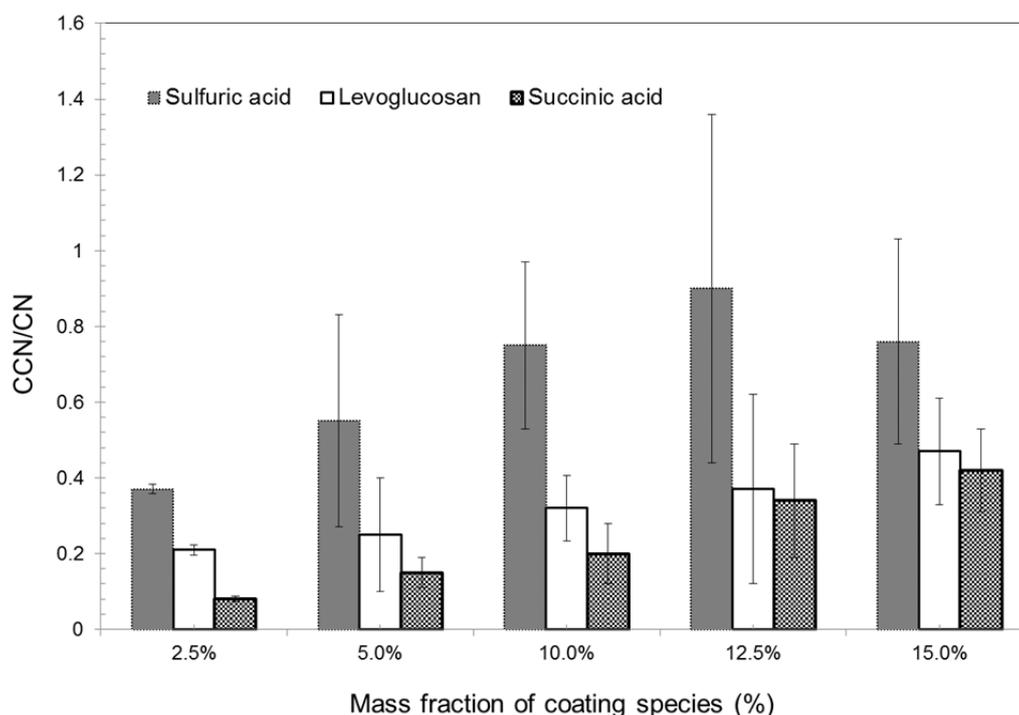


Fig. 5. CCN activation fractions of BC particles coated with sulfuric acid, levoglucosan, or succinic acid at a supersaturation of 0.5% as a function of the mass fraction (in percentage) of the coating species (the coated BC particles were produced using the premixed solution method).

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