Study of Cloud Condensation Nuclei Activities and Hygroscopic Properties Based on Core-Shell Model

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Abstract—Cloud condensation nuclei (CCN) activities and hygroscopic properties of aerosol particles were studied based on the core-shell model with extended Köhler equation. Three kinds of core-shell particles were mainly considered: (1) insoluble core and soluble inorganic shell; (2) insoluble core and organic shell with limited solubility; (3) organic core with limited solubility and soluble inorganic shell. Results of the model calculations showed that Köhler curves of aerosol particles were different from classical Köhler curves of inorganic particles. Particle size, chemical composition and mass fraction of components all influence the curve shape, and the curve might show one or more extreme points of supersaturation. Critical supersaturation decreased when the mass fraction of the shell increased. This implied that the heterogeneous chemical reaction that happened to suspended particles in air could increase shell mass fraction and make particles more CCN active and hygroscopic. When the three models were compared, particles in model 3 were most CCN active, while those in model 2 were least CCN active. If the shell mass fraction was 0.1–0.2 for model 3 and 0.3–0.4 for model 1, the calculated hygroscopic parameter $\kappa$ was closer to historical measuring results of 0.2–0.3 in polluted areas in China while the hygroscopic parameter would never be close to measuring results in model 2 no matter how high the shell mass fraction was. Based on discussion of two component particles of the core-shell model, more components in core-shell model could be considered, and the calculations were discussed in this article.

Keywords: Core-shell model, Aerosol particles, Köhler equation, Cloud condensation nuclei, Hygroscopic properties.

1. Introduction

Atmospheric aerosol particles serving as cloud condensation nuclei (CCN) are very important elements of the hydrological cycle and climate (Rose et al., 2010) because aerosol particles change cloud microphysical structures and radiative properties (Li et al., 2007; Rosenfeld et al., 2007; Xu, 2001; Zhao et al., 2006). Under increasing aerosol number concentrations, clouds tend to have more but smaller droplets and thus reflect more solar radiation (Twomey, 1977). Aerosol and cloud interactions directly influence the cloud microphysical process, thereby leading to changes of spatio-temporal distribution of precipitation rather than the globally averaged amount of precipitation (Rosenfeld et al., 2008).

Aerosol particles have complicated chemical compositions and many mixing states, and the spatio-temporal distribution is quite uneven (Mao, 2002). Different chemical features of aerosol particles influence their hygroscopic properties and CCN activities (Hanel et al., 1981). It is very important to set up a relationship between chemical features of particles and the CCN activities so their influence to clouds and precipitation process will be clearer.

Many observations and experimental studies have been carried out and many results gained. For example, observation showed spatio-temporal distribution of CCN and its influence by local chemical components and resources in mountainous areas of China (Li, 2014), Nevada in the USA (Hudson et al., 1991), plains areas in China (Duan, 2010; Liu et al., 2011; Lu & Guo, 2012; Shi & Duan, 2007), desert area in China (Sang, 2012), Amazon rain forest (Gunthe et al., 2009), the Mediterranean (Bougiatioti et al., 2009); Toronto (Broekhuizen et al., 2006); North Atlantic Ocean (Liu et al., 1996); Ireland (Reade et al., 2006) and so on. Many experimental studies showed the activation diameters and
hygroscopic parameters of specific particles, such as single component inorganic (Orr et al., 1958), organic (Cruz & Pandis, 1997; Corrigan & Novakov, 1999; Raymond & Pandis, 2002; Varutbangkul et al., 2006; Huff-Hartz et al., 2006; Awuku et al., 2009; Lambe and Onasch, 2011; Engelhart et al., 2008), and a mixture of inorganic and organic (Cruz & Pandis, 1998, 2000; Hansson et al., 1998; Abbatt et al., 2005; Trochkine, 2003). There are some studies related to heterogeneous chemical reactions (Zhu, 2010) or (BC), sand or dust, organic and so on through heterogeneous chemical reactions (Hudson & Clark, 1992; Jordanov & Zellner, 2006; Takahama et al., 2007; Pant et al., 2006).

2. Model Introduction

2.1. Extended Köhler Equation

The extended Köhler equation for multicomponent particles with components of limited solubility (Raymond & Pandis, 2002, 2003) assumes the particle is spherical and can be written as:

$$ S = \frac{\gamma_w}{\gamma_w + \frac{n_w}{n_w} \sum \frac{n_{w,li} \rho_w}{M_w}\exp\left(\frac{4M_w \sigma}{RTD_r \rho_{sol}}\right)} $$

(1)

In which $n_w = \frac{\rho_w \pi (D_p^3 - D_s^3)}{6M_w}$

(2)

$$ \varepsilon_i = \frac{C_{sat,i} \rho_w (D_p^3 - D_s^3)}{D_s \sum \rho_{si} x_i} $$

(3)

$$ \rho_{sol} = \frac{\rho_w D_p^3 - D_s^3}{D_p^3 - D_s^3} - \sum \rho_{si} \varepsilon_i $$

(4)

where $S$ is the water vapor saturation relative to a flat pure water surface, $\gamma_w$ is the water activity coefficient in the aqueous phase, $n_w$ is the number of moles of water in the droplet, $M_w$ is the molecular weight of water, $\sigma$ is the surface tension of the solution, 0.0761 J/m2 (Raymond & Pandis, 2002, 2003), $R$ is the gas constant, $T$ is the temperature, $D_p$ is the droplet diameter, $\rho_{sol}$ is the density of the solution, $D_s$ is the diameter of the dry particle, $\varepsilon_i$ is the dissolved mass fraction of component $i$, $x_i$ is the mass fraction of component $i$ in the dry particle, $\eta_i$ is the average number of ions into which a solute molecule dissociates, $\rho_{si}$ is the density of component $i$, $M_{si}$ is the molecular weight of component $i$, $\rho_w$ is the density of water, and $C_{sat,i}$ is the solubility of component $i$.

2.2. Hygroscopic Parameter $\kappa$

Petters and Kreidenweis (2007, 2008) used a hygroscopic parameterization, $\kappa$, to parameterize the hygroscopic properties of particles, as in Eq. 5. Higher values of $\kappa$ indicate being more hygroscopic,
while lower values of $\kappa$ indicate being less hygroscopic.

$$S = \frac{D_p^3 - D_s^3}{D_p^3 - D_s^3(1 - \kappa)} \exp\left(\frac{4\sigma M_w}{RT \rho_w D_p} \right)$$  \hspace{1cm} (5)

$\kappa$ can be derived from Eq. 5, and if $\kappa > 0.2$, it can be calculated by Eqs. 6 and 7.

$$\kappa = \frac{4A^3}{27D^3 \ln^2 S_c}$$  \hspace{1cm} (6)

$$A = \frac{4M_w \sigma}{RT \rho_w}$$  \hspace{1cm} (7)

Here, $S_c$ is the critical supersaturation and $D_{sc}$ is the activation diameter.

### 2.3. Core-Shell Model

Based on the main pollutants and heterogeneous reactions (Zhu, 2010; Takahama et al., 2007), three core-shell models were mainly considered: (1) insoluble core and soluble inorganic shell; (2) insoluble core and organic shell with limited solubility; (3) organic core with limited solubility and inorganic shell.

In the calculation with the extended Köhler equation, the core and shell components and the mass fraction of the shell are given in each case. When dry particles absorb water vapor, some of the core or shell can be dissolved, and the dissolved mass fraction for each component can be calculated by the solubility of component $C_{sat,i}$.

Based on the dissolved mass fraction of each component and the original dry particle diameter, we calculated the water vapor saturation with the extended Köhler equation.

To simplify the calculation, ammonium sulfate represented a soluble inorganic compound and CaCO$_3$ represented an insoluble substance. There are many kinds of organics with different solubilities. Hygroscopic properties are very weak for organics with low solubility, so we only included ones with higher solubility, for example, 27.5 g l$^{-1}$. The parameters of the main components are shown in Table 1. The organic density is an estimate average of density values of some common organics (Kostenidou et al., 2007).

### 3. Results

#### 3.1. Köhler Curves

From the calculation results, the Köhler curves of aerosol particles were different from the classical Köhler curve of inorganic particles. Sizes, chemical composition and mass fraction of different components influence the shapes of curves. Some curves might have one or more extreme points. The curve and extreme points reflect how a particle grows when saturation increases. When supersaturation increases, usually the droplet particle diameter increases. After the extreme point, the droplet can grow with less supersaturation. In this study, Köhler curves were drawn for 0.1-μm particles based on three core-shell models. The mass fraction of the shell is 0.1, 0.5 and 0.9, as shown in Fig. 1.

If the shell mass fraction is 0.1, the Köhler curve in model 1 is simple with only one extreme point showing that the critical supersaturation was 0.43%. The Köhler curve in model 2 showed a monotonic declining trend with a very high extreme point at 2.1% supersaturation, which showed that the dry particles began to absorb water vapor at 2.1% supersaturation and the supersaturation decreased when the particles grew bigger. Therefore, the critical supersaturation was 2.1%. The Köhler curve in model 3 had two extreme points with supersaturations of 0.18% and 0.26%. In Fig. 1a, the first point meant that the particle grew to 0.33 μm when supersaturation increased to 0.18%. After that, the particle could grow even if the supersaturation was a little bit lower than 0.18% until it grew to 0.42 μm, and then it needed higher supersaturation to grow. After the second extreme points of 0.26%, it could grow with

| Parameters of different components | Molecular weight $M_{si}$ (kg/mol) | Density $\rho_{si}$ (kg/m$^3$) | Solubility $C_{sat}$ (g/g) | Average number of ions $c_i$ |
|-----------------------------------|-----------------------------------|-----------------------------|---------------------------|-----------------------------|
| (NH$_4$)$_2$SO$_4$                 | 0.132                             | 1770                        | 0.754                     | 3                           |
| Organics (OC)                     | 0.15                              | 1500                        | 0.0275                    | 1                           |
| CaCO$_3$                          | 0.1                               | 2710                        | 1.5 × 10$^{-4}$           | 1                           |

Table 1

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supersaturation < 0.26%, which meant that the particles could be activated into water droplets at supersaturation of 0.26% (Fig. 1a).

If the shell mass fraction was 0.5, Köhler curves in model 1 and 3 were very similar, with one extreme point individually. The critical supersaturation of particles in model 1 was 0.2% and that in model 3 was 0.18%. Model 2 still showed a declining Köhler curve with an extreme point at 2% supersaturation (Fig. 1b). The critical supersaturation for model 1, 2 and 3 was 0.2%, 2% and 0.18%, respectively.

Figure 1
Köhler curves for particles of 0.1-μm at core:shell mass ratio of a 0.9:0.1; b 0.5:0.5; c 0.1:0.9
If the shell mass fraction was 0.9, the Köhler curves in models 1 and 3 nearly overlapped because the hygroscopicity was mainly determined by the mass fraction of the soluble shell. In this mass fraction, the critical supersaturation was 0.15%. The Köhler curve in model 2 changed significantly; the particles began to absorb water vapor at a very high supersaturation (1.8%) and then the supersaturation decreased when the particles grew until the supersaturation was 0.27%. After that, the supersaturation continued to increase and achieved an extreme point (0.3%) and dropped down again, which meant the solution droplet was activated at 0.3% supersaturation. The critical supersaturation for models 1, 2 and 3 was 0.15%, 1.8% and 0.15%, respectively.

At the same size and shell mass fraction, the critical supersaturation in model 3 was lowest, followed by that in model 1, while that in model 2 was the highest. The mass fraction of the soluble component greatly influenced the critical supersaturation.

3.2. Particle Size and Critical Supersaturation

Critical supersaturation from each Köhler curve for each dry particle was extracted and a figure of critical supersaturations as a function of dry particle sizes was produced for the three core-shell models, as shown in Fig. 2. Shell mass fractions of 0.01, 0.1, 0.5 and 1 were mainly considered (Fig. 2). Shell mass fraction 0.01 meant fresh particles without evident heterogeneous chemical reaction; fraction 0.1 and 0.5 meant particles at different aging levels through heterogeneous chemical reactions; those with fraction 1 represented particles composed totally by pure inorganic or organic components.

As Fig. 2 shows, critical supersaturation in model 3 was the lowest at the same size and shell mass fraction followed by that in model 1, while that in model 1 was the highest. The mass fraction of the soluble component greatly influenced the critical supersaturation.

3.3. Mass Fraction and Supersaturation

Critical supersaturation as a function of shell mass fraction is shown in Fig. 3. As Fig. 3 shows, the higher the shell mass fraction, the lower the critical supersaturation was for a certain size particle. For models 1 and 3, critical supersaturation decreased very fast when shell the mass fraction increased, especially when the shell mass fraction changed from 0 to 0.1 (Fig. 3a–c). For example, when the shell mass fraction increased from 0 to 0.1, the critical supersaturation of 100 nm particles in model 1 decreased from 2% to 0.5%. When the shell mass fraction increased from 0.1 to 0.2, the critical supersaturation decreased from 0.5% to 0.35% (Fig. 3b). Similar changes also took place in the particles in model 3. This meant that the inorganic shell could make particles more hygroscopic and CCN active.

For model 2 (Fig. 2b), particles were quite hard to activate. At shell mass fractions of 0.01 and 0.1, only particles > 1 μm could be activated at 0.1% supersaturation, while bigger particles could not be suspended in air for a long time, so they seldom influenced clouds or precipitation. At shell mass fractions of 0.5 and 1, the activation diameters were 860 and 530 nm at 0.1% supersaturation.

Comparing the three models, more particles in model 1 and 3 could be activated into water droplets than those in model 2.
3.4. Comparison of Three Models

At the same size and shell mass fraction, the critical supersaturation for particles in model 2 was the highest (Fig. 4b) and least hygroscopic because the chemical components were less soluble. The increase of shell mass fraction could help activation, while still not being efficient enough to decrease critical supersaturation. Particles in model 1 and 3 were more hygroscopic, and the increase of shell mass fraction could decrease the critical supersaturation quickly (Fig. 4a, c).

There were hardly any differences when the particles were bigger or mass fraction of the shell was higher between model 1 and 3 (Fig. 4a, c)
because they are both hygroscopic. The difference between these two models occurs when the diameter and mass fraction are lower, as shown in the red to yellow zone in Fig. 4a, c. The particle in model 3 is more hygroscopic than in model 1.

3.5. Hygroscopic Parameter $\kappa$

Figure 5 shows the hygroscopic parameter $\kappa$ calculated by Eqs. 5–7. As Fig. 5 shows, the higher the shell mass fraction is, the higher the $\kappa$. The $\kappa$ was
highest for model 3 followed by model 1. The lowest was model 2.

According to historical observation results in heavily polluted areas in China (Liu et al., 2011), κ mainly was 0.2–0.3. The shadowed part in Fig. 5 shows the calculation results, which fit the observation results. From Fig. 5, the shell mass fraction of 0.3–0.4 for particles in model 1 and 0.1–0.2 in model 3 could reflect the true hygroscopic properties of ambient particles. For the particles in model 2, even if the shell mass fraction was 1, the
calculation results could not match the observation results.

4. Extended Core-Shell Model Calculation

The discussion in Sect. 3 was based on the hypothesis that the core or shell contained only one component. In fact, the ambient particles are very complicated, and the core or shell might contain more components. The core-shell model can be extended based on Eqs. 1–4.

Take an example of particles with three components: the core contains one component with limited solubility of 0.01 g l⁻¹, the molecular weight is 1500 kg mol⁻¹, and density is 0.012 kg m⁻³. The shell contains two components. One component is organic with solubility 5 g l⁻¹, the molecular weight is 1360 kg mol⁻¹, and density is 0.015 kg m⁻³. The other component is inorganic with 500 g l⁻¹ solubility, the molecular weight is 1770 kg mol⁻¹, and density is 0.013 kg m⁻³. In the calculation, the molecular weight in Table 2 shows the activation diameters at supersaturation of 0.2 and 0.4% and hygroscopic parameter κ.

Based upon long-term atmospheric observation, main chemical composition and mass fraction, the average activation diameter and hygroscopic parameter can be calculated in a local area. According to the results in Table 2, the more mass fraction of soluble component a particle contains, the more hygroscopic the particle is.

5. Conclusions

According to the main heterogeneous chemical reaction, three kinds of core-shell models were studied based on an extended Köhler equation, and CCN activities and hygroscopic properties were then analyzed. The following conclusions were reached in our study:

(1) The Köhler curves of particles with core-shell structures were different from classical Köhler curves of inorganics. Particle size, chemical composition and mass fraction of different components influenced the shape of the Köhler curve, which may contain one or more extreme points.

(2) Three core-shell models were mainly considered:
   (a) insoluble core and soluble inorganic shell;
   (b) insoluble core and organic shell with limited solubility;
   (c) organic core with limited solubility and inorganic shell. Comparing these three models, particles in model 3 were the most hygroscopic and CCN active, followed by model 1; those in model 2 were the least hygroscopic.

(3) Shell mass fraction represented the aging level of particles through heterogeneous chemical reaction. The increase of shell mass fraction caused the decrease of critical supersaturation. The
higher the shell mass fraction, the more hygroscopic and CCN active the particles were. 

(4) According to historical observation results in heavily polluted areas in China, the hygroscopic parameter \( \kappa \) mainly was 0.2–0.3. At shell mass fraction of 0.3–0.4 for particles in model 1 and 0.1–0.2 for model 3, the calculated results of hygroscopicity could reflect the realistic hygroscopic properties of ambient particles. For the particles in model 2, the calculation results were difficult to match to the observation results no matter how high the shell mass fraction was.

(5) The core-shell model can be extended considering more kinds of components. If the main chemical composition and mass fraction can be gained through long-term atmospheric observation, the average activation diameter and hygroscopic parameter can be calculated in a local area.

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Declarations

Conflict of interest The authors declare no conflict of interest.

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