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The Effect of Hygroscopic Growth on Continental Aerosols

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ABSTRACT

In this paper, the authors investigated some microphysical and optical properties of continental clean aerosols from OPAC to determine the effect of hygroscopic growth at the spectral range of 0.25 µm to 2.5 µm and eight relative humidities (RHs) (0%, 50%, 70%, 80%, 90%, 95%, 98% and 99%). The microphysical properties extracted were radii, volume mix ratio, number mix ratio and mass mix ratio as a function of RH while the optical properties are scattering and absorption coefficients and asymmetric parameters. Using the microphysical properties, growth factors of the mixtures were determined while using optical properties the enhancement parameters were determined and then parameterized using some models. We observed that the data fitted the models very well. The angstrom coefficients show that the mixture has bimodal type of distribution with the dominance of fine mode particles.

Keywords: Microphysical Properties; Optical Properties; Hygroscopic Growth; Parametrization; Enhancement Parameters; Angstrom Coefficients

1. Introduction

Aerosol in the atmosphere is comprised of numerous and diverse components originating from both natural and anthropogenic activities.

An important factor affecting the role aerosols play in climate change is their hygroscopicity and is currently modeled in global climate models (GCMs), mostly to better predict the scattering properties and size distribution under varying humidity conditions [1]. The swelling of aerosols due to water vapor uptake will enhance their ability to scatter radiation. Numerous studies have investigated the relationship between aerosol scattering and relative humidity RH in terms of the hygroscopic growth factor *gf*(RH) using humidified nephelometers. These have been used for airborne or ground-based determination of the growth factor considering a "dry" RH over the range from 20% - 40% and a ''wet'' RH up to 90% [2-5].

The characterization of particle hygroscopicity has primary importance for climate monitoring and prediction. Model studies have demonstrated that relative humidity (RH) has a critical influence on aerosol climate forcing. Hygroscopic properties of aerosols influence particle size distribution and refractive index and hence their radiative effects. Aerosol particles tend to grow at large relative humidity values as a result of their hygroscopicity.

Some aerosol particles, such as ammonium sulphate $(NH_4)_2SO_4$, sea salt and ammonium nitrate NH_4NO_3 are hygroscopic. Changes in relative humidity modify their size distribution and refractive index and hence the optical properties of the aerosol, including the scattering coefficient [6-9]. Jeong *et al*. [10] demonstrated an exponential dependence of the aerosol optical thickness on relative humidity. A strong correlation of spectral aerosol optical thickness with precipitable water, especially for continental air masses, was shown by Rapti [11].

Water-soluble organic carbon (WSOC) species are emitted as primary particles, especially during biomass combustion, and produced as a result of reactions in the gas and aqueous phases [12-18]. Moreover, WSOC has been suggested as a marker for secondary organic aerosol (SOA) in the absence of biomass burning (e.g., Docherty *et al*. [19]).

In a study of aged continental aerosols, Swietlicki *et al*. [7] observed 2 modes, a less hygroscopic mode with a *gf*(RH) of 1.12 and a more hygroscopic mode with a *gf*(RH) between 1.44 and 1.65. They postulated that the hygroscopic growth could be attributed entirely to the inorganic content of the aerosol: sulfate, nitrate and ammonium ions. Particle hygroscopicity may vary as a function of time, place and particle size [20-22]. The size and the solubility of a particle determine the response of an ambient particle to changes in RH. The water vapor pressure above a water droplet containing dissolved material is lowered by the Raoult effect. The equilibrium size of a droplet was first described by Kohler [23], who considered the Kelvin (curvature) and Raoult (solute) effect. Using optical properties, several previous studies (e.g. Sheridan *et al*. [24]) have measured and modeled enhancement factors for continental aerosols.

The aim of this study is to determine the aerosols' hygroscopic growth and enhancement factors for continenttal clean aerosols from the data extracted from OPAC. One variable and two variables parameterizations models will be performed to determine the relationship of the particles' hygroscopic growth and enhancement parameters with the RH. Angstrom coefficients are used to determine the particles' type and the type mode size distributions.

2. Methodology

The models extracted from OPAC are given in **Table 1**.

The main parameter used to characterize the hygroscopicity of the aerosol particles is the aerosol hygroscopic growth factor *gf*(RH), which indicates the relative increase in mobility diameter of particles due to water absorption at a certain RH and is defined as the ratio of the particle diameter at any RH to the particle diameter at RH = 0 and RH is taken for seven values 50%, 70%, 80%, 90%, 95%, 98% and 99% [22,26]:

$$gf(\mathrm{RH}) = \frac{D(\mathrm{RH})}{D(\mathrm{RH}=0)} \tag{1}$$

The *gf*(RH) can be subdivided into different classes with respect hygroscopicity. One classification is based on diameter growth factor by Liu *et al.* [27] and Swietlicki *et al*., [22] as barely hygroscopic (*gf*(RH) = 1.0 – 1.11), less Hygroscopic (*gf*(RH) = 1.11 − 1.33), more Hygroscopic (*gf*(RH) = 1.33 − 1.85) and most hygroscopic growth (*gf*(RH) > 1.85).

Atmospheric particles of a defined dry size typically exhibit different growth factors. This is due to either external mixing of particles in an air sample or variable relative fractions of different compounds in individual particles (the latter here in after referred to as quasi-internally mixed). A mono-modal growth distribution without spread can only be expected in very clean and homogeneous air parcels. For further details on mixing states see e.g. Buzorius *et al*. [28].

Most atmospheric aerosols are externally mixed with respect to hygroscopicity, and consist of more and less hygroscopic sub-fractions [22]. The ratio between these fractions as well as their content of soluble material determine the hygroscopic growth of the overall aerosol.

Prediction of hygroscopic growth factors with Kohler theory requires detailed knowledge of particle composition as well as a thermodynamic model, which describes the concentration dependence of the water activity for such a mixture. The hygroscopic growth factor of a mixture, gf_{mix}(RH), can be estimated from the growth factors of the individual components of the aerosol and their respective volume fractions, V_k, using the Zdanovskii-Stokes-Robinson relation and other researchers [29-32]:

$$gf_{mix}(\mathrm{RH}) = \left(\sum_k V_k gf_k^3\right)^{1/3} \tag{2}$$

where the summation is performed over all compounds present in the particles. Solute-solute interactions are neglected in this model and volume additivity is also assumed. The model assumes spherical particles, ideal mixing (*i.e.* no volume change upon mixing) and independent water uptake of the organic and inorganic components.

It can also be computed using the corresponding number fractions n_k as [33,34];

$$gf_{mix}(\mathrm{RH}) = \left(\sum_k n_k gf_k^3\right)^{1/3} \tag{3}$$

where n_k is the number fraction of particles having the growth factor gf_k .

We now proposed the gf_{mix}(RH) to be a function of mass mix ratio as

$$gf_{mix}(\mathrm{RH}) = \left(\sum_k m_k gf_k^3\right)^{1/3} \tag{4}$$

where m_k represents the mass mix ratio of particles having the growth factor gf_k.

The RH dependence of gf_{mix}(RH) were parameterized in a good approximation by a one-parameter equation, proposed e.g. by Petters and Kreidenweis [35]:

$$gf_{mix}(a_w) = \left(1 + \kappa \frac{a_w}{1 - a_w}\right)^{\frac{1}{3}} \tag{5}$$

Here, a_w is the water activity, which can be replaced by the relative humidity RH, if the Kelvin effect is negligible, as for particles with sizes more relevant for light scattering and absorption. At equilibrium, it can be shown that, over a flat surface, the water activity equals the ambient relative humidity in the sub-saturated humid

Table 1. Compositions of aerosol type [25].

Aerosol model types	Components	Concentration N_i (cm^{-3})
Continental clean	WASO	2600.0
	INSO	0.15
	Total	26000.15

Note: N_i is the mass concentration of the component, water soluble components (WASO, consists of scattering aerosols, that are hygroscopic in nature, such as sulfates and nitrates present in anthropogenic pollution) and water insoluble (INSO).

environment [36,37]. The coefficient κ is a simple measure of the particle's hygroscopicity and captures all solute properties (Raoult effect).

Humidograms of the ambient aerosols obtained in various atmospheric conditions showed that gf_{mix}(RH) could as well be fitted well with a γ-law [38-42] as

$$gf_{mix}\left(\mathrm{RH}\right)=\left(1-\frac{\mathrm{RH}}{100}\right)^{\gamma} \tag{6}$$

Particle hygroscopicity is a measure that scales the volume of water associated with a unit volume of dry particle [35] and depends on the molar volume and the activity coefficients of the dissolved compounds [43].

The bulk hygroscopicity factor B under subsaturation RH conditions was determined using the relation:

$$B=\left(1-gf_{mix}^{3}\right)\ln a_{w} \tag{7}$$

where a_w is the water activity, which can be replaced by the RH as explained earlier.

The impact of hygroscopic growth on the aerosol optical properties is usually described by the enhancement factor $f_\chi(\mathrm{RH},\lambda)$:

$$f_{\chi}\left(\mathrm{RH},\lambda\right)=\frac{\chi\left(\mathrm{RH},\lambda\right)}{\chi\left(\mathrm{RH}=0,\lambda\right)} \tag{8}$$

where $\chi(\mathrm{RH},\lambda)$ can be denoting the aerosol scattering and absorption coefficients, and asymmetry parameters. RH corresponds to any condition, and can cover the entire RH spectrum. In this paper we will only use scattering, absorption and asymmetric parameter. The reason for using asymmetric parameter is to determine the effect of hygroscopic growth on forward scattering. This method was initially introduced by Covert *et al*. [2].

In general the relationship between $f_{\chi}\left(\mathrm{RH},\lambda\right)$ and RH is nonlinear (e.g. Jeong *et al*. [10]). In this paper we determine the empirical relations between the enhancement parameter and RH [44] as:

$$f_{\chi}\left(\mathrm{RH},\lambda\right)=\frac{\chi\left(\mathrm{RH},\lambda\right)}{\chi\left(\mathrm{RH}=0,\lambda\right)}=\left(\frac{100-\mathrm{RH}_{ref}}{100-\mathrm{RH}_{high}}\right)^{\gamma} \tag{9}$$

where in our study RH_{ref} is 0%. The γ known as the humidification factor represents the dependence of aerosol optical properties on RH, which results from changes in the particle size and refractive index upon humidification. The parameter in our case was obtained by combining the eight $\chi\left(\mathrm{RH},\lambda\right)$ parameters at 0%, 50%, 70%, 80%, 90%, 95%, 98% and 99% RH. The use of γ has the advantage of describing the hygroscopic behavior of aerosols in a nonlinear manner over a broad range of RH values; it also implies that particles are deliquesced [45], a reasonable assumption for this data set due to the high ambient relative humidity during the field study. The γ parameter is dimensionless, and it increases with increasing particle water uptake. From previous studies, typical values of γ for ambient aerosol ranged between 0.1 and 1.5 [45-47].

Two parameters empirical relation is also used [10,48] as:

$$f_{\chi}\left(\mathrm{RH},\lambda\right)=a\left(1-\frac{\mathrm{RH}(\%)}{100}\right)^{b} \tag{10}$$

The model assumes equilibrium (metastable) growth of the aerosol scattering with RH such that the humidigraph profile does not display a deliquescent growth profile. For aerosol in a humid environment, this behavior will hold true. Most aerosols are a mixture of metastable and deliquescent particles and will exhibit some deliquescent behavior. To verify the non-linearity of the relation between $f_{\chi}\left(\mathrm{RH},\lambda\right)$ and RH, the Equations (9) and (10) were modeled at λ = 0.25, 1.25 and 2.50 μm.

The Angstrom exponent being an indicator of the aerosol spectral behaviour of aerosols [49], the spectral behavior of the aerosol optical parameter (X, say), with the wavelength of light (λ) is expressed as inverse power law [50]:

$$X\left(\lambda\right)=\beta\lambda^{-\alpha} \tag{11}$$

where $X(\lambda)$ can represent scattering and absorption coefficients. The variable $X(\lambda)$ can be characterized by the Angstron parameter, which is a coefficient of the following regression,

$$\ln\mathrm{X}\left(\lambda\right)=-\alpha\ln\left(\lambda\right)+\ln\beta \tag{12}$$

however the Angstrom exponent itself varies with wavelength, and a more precise empirical relationship between aerosol extinction and wavelength is obtained with a 2nd-order polynomial [51-61], as:

$$\ln X\left(\lambda\right)=\alpha_{2}\left(\ln\lambda\right)^{2}+\alpha_{1}\ln\lambda+\ln\beta \tag{13}$$

and then we proposed the cubic

$$\ln X\left(\lambda\right)=\ln\beta+\alpha_{1}\ln\lambda+\alpha_{2}\left(\ln\lambda\right)^{2}+\alpha_{3}\left(\ln\lambda\right)^{3} \tag{14}$$

where $X\left(\lambda\right)$ can be any of the optical parameter, β, α, α_1, α_2, α_3 are constants that are determined using regression analysis with SPSS16.0.

We also determine the effect of hygroscopic growth on the effective refractive indices of the two mixed aerosols using the following formula [62]:

$$\frac{\varepsilon_{eff}-\varepsilon_{0}}{\varepsilon_{eff}+2\varepsilon_{0}}=\sum_{i=1}^{2}f_{i}\frac{\varepsilon_{i}-\varepsilon_{0}}{\varepsilon_{i}+2\varepsilon_{0}} \tag{15}$$

where f_i and ε_i are the volume fraction and dielectric constant of the i^{th} component and ε_0 is the dielectric constant of the host material. For the case of Lorentz-Lorentz [63,64], the host material is taken to be vacuum, $\varepsilon_0 = 1$.

3. Results and Observations

Figure 1 is the plot from the data of **Table 2**, and it shows non-linear relation gf_{mix} with RH, (a steep curve) with deliquescence observed at relative humidities as from 90% to 99% RH.

The results of the parameterizations by one and two parameters of Equations (5) and (6) are:

$$C = 1.433571, k = 0.012412, R^2 = 0.8731$$

from Equation (5)

$$\gamma = -0.069589,\ R^2 = 0.9962 \quad \text{from Equation (6)}$$

The fitted curve can be represented by one and two empirical parameters fit of the form of Equations (5) and (6), though Equation (6) has higher coefficient of determination.

Figure 2 is a plot from the data of **Table 2**, and shows non-linear relation B with RH, (a steep curve) with deliquescence observed at relative humidities as from 90% to 99% RH.

Figure 3 is a plot from the data of **Table 3**, and it shows an increase in particle diameter with increasing RH and shows a steep curve with deliquescence observed at relative humidities as from 90% to 99% RH.

The results of the parameterizations by a one and two parameters of Equations (5) and (6) are:

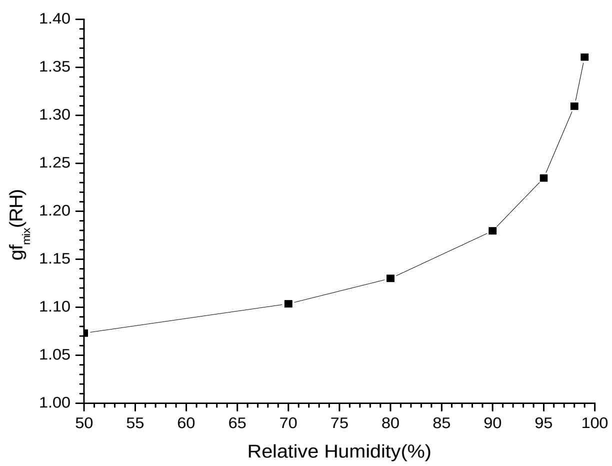

Figure 1. A graph of growth factor of the mixture using number mix ratio (Equation (3)) against RH.

Table 2. The growth factor of the aerosols using number mix ratio (Equation (3)) and bulk hygroscopicity factor (Equation (7)).

RH(%)	50	70	80	90	95	98	99
gf_{mix}(RH)	1.0731	1.1036	1.1301	1.1796	1.2346	1.3094	1.3606
Bulk Hygroscopicity factor (B)	0.1634	0.1228	0.0989	0.0676	0.0452	0.0252	0.0153

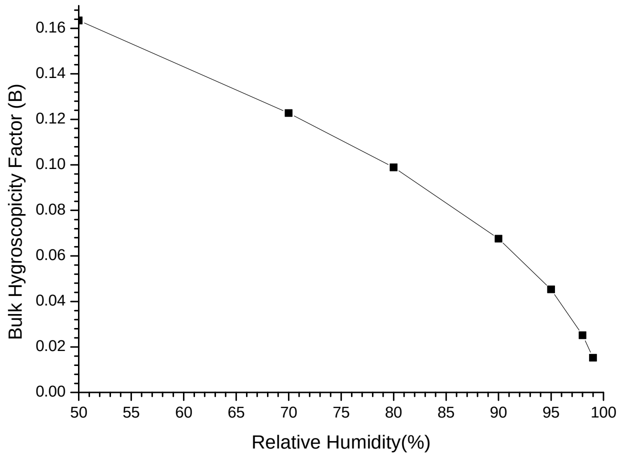

Figure 2. Bulk hygroscopcity factor of the mixture using number mix ratio (Equation (7)).

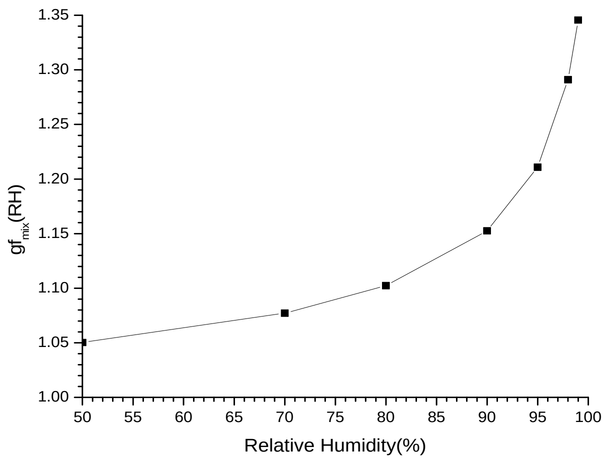

Figure 3. A graph of growth factor of the mixture using volume mix ratio (Equation (2)).

Table 3. The growth factor of the aerosols using volume mix ratio (Equation (2)) and bulk hygroscopicity factor (Equation (7)).

RH(%)	50	70	80	90	95	98	99
gf_{mix}(RH)	1.0503	1.0772	1.1024	1.1526	1.2108	1.2910	1.3456
Bulk Hygroscopicity factor (B)	0.1100	0.0891	0.0758	0.0560	0.0398	0.0233	0.0144

$$C = 1.334733,\ k = 0.012542,\ R^2 = 0.8860$$

from Equation (5)

$$\gamma = -0.064125,\ R^2 = 0.9995 \quad \text{from Equation (6)}$$

The fitted curve can be represented by one and two empirical parameters fit of the form of Equations (5) and (6), though Equation (6) has higher coefficient of determination.

Figure 4 is a plot from the data of **Table 3**, is almost the same as **Figure 2**.

Figure 5 is a plot from the data of **Table 4**, it shows an increase in particle diameter with increasing RH and shows a steep curve with deliquescence observed at relative humidities as from 90% to 99% RH.

The results of the parameterizations by a one and two parameters of Equations (5) and (6) are:

$$C = 1.294418,\ k = 0.012166,\ R^2 = 0.8983$$

from Equation (5)

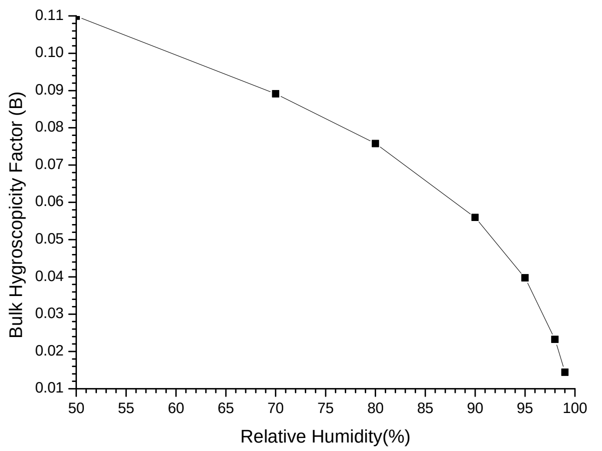

Figure 4. Bulk Hygroscopcity factor of the mixture using volume mix ratio (Equation (7)).

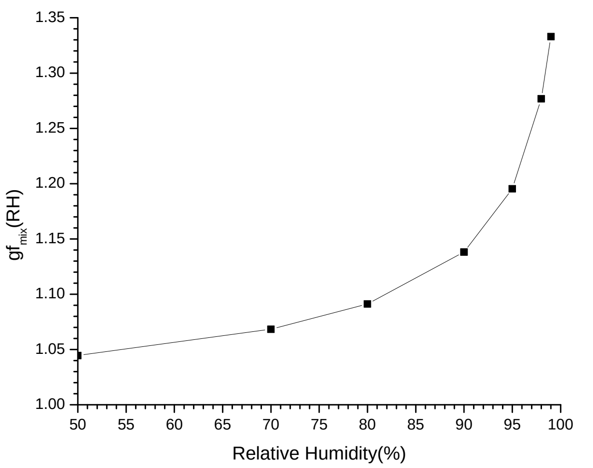

Figure 5. A graph of growth factor of the mixture using mass mix ratio (Equation (4)).

Table 4. The growth factor of the aerosols using mass mix ratio (Equation (4)) and bulk hygroscopicity factor (Equation (7)).

RH(%)	50	70	80	90	95	98	99
gf_{mix}(RH)	1.0446	1.0684	1.0912	1.1382	1.1953	1.2768	1.3331
Bulk Hygroscopicity factor (B)	0.0970	0.0783	0.0668	0.0500	0.0363	0.0218	0.0138

$$\gamma = -0.060798,\ R^2 = 0.9981 \quad \text{from Equation (6)}$$

The fitted curve can be represented by one and two empirical parameters fit of the form of Equations (5) and (6), though Equation (6) has higher coefficient of determination.

Figure 6 is a plot from the data of **Table 4**, is the same as **Figures 2** and **4**.

Figure 7 shows that scattering increases substantially as a result of the increase in hygroscopic growth most especially at smaller wavelength. This shows the high concentration of smaller particles, and that hygroscopic growth has more effect on small particles. This increase is due to the growth of smaller particles to sizes at which they scatter more light being more pronounced than that for larger particles.

Table 5 shows that the linear part reflects the dominance of fine mode particles because $\alpha > 1$ and has been verified by the sign of α_2 as reported by [52,56,68-71] for the existence of negative curvatures for fine-mode aerosols and positive curvatures for coarse mode. As from

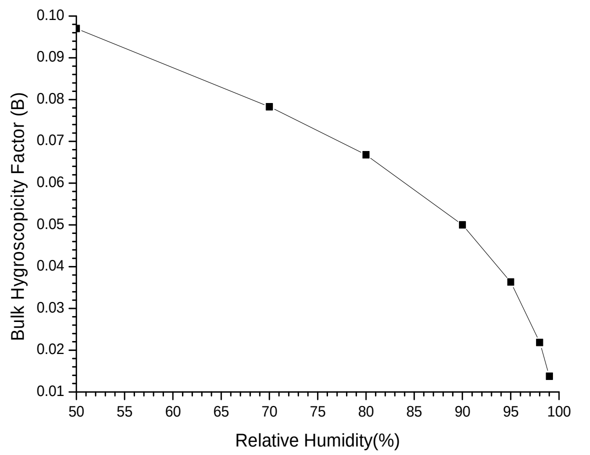

Figure 6. Figure 2; Bulk Hygroscopcity factor of the mixture using mass mix ratio (Equation (7)).

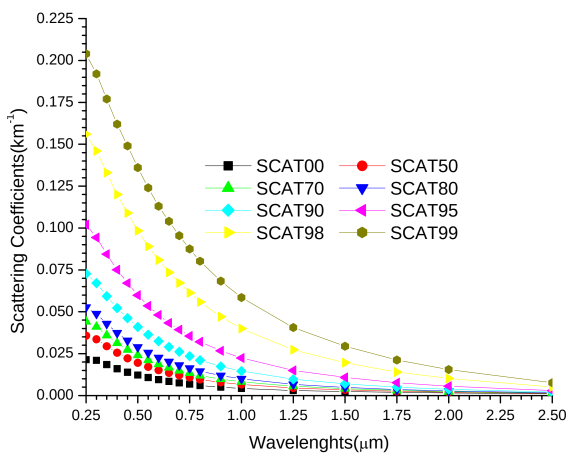

Figure 7. A plot of scattering coefficients against wavelength.

Table 5. The results of the Angstrom coefficients of scattering coefficients using Equations (12)-(14) for continental clean model at the respective relative humidities using regression analysis with SPSS16.0.

RH	Linear		Quadratic			Cubic			
(%)	R2	A	R^2	α_1	α_2	R^2	α_1	α_2	α_3
0	0.9887	1.3858	0.9956	−1.4647	−0.1718	0.9991	−1.6016	−0.0156	0.2048
50	0.9839	1.5029	0.9985	−1.6276	−0.2715	0.9997	−1.7142	−0.1727	0.1295
70	0.9802	1.5327	0.9992	−1.6781	−0.3164	0.9998	−1.7403	−0.2454	0.0931
80	0.9763	1.5483	0.9995	−1.7108	−0.3537	0.9998	−1.7522	−0.3065	0.0619
90	0.9682	1.5522	0.9998	−1.7429	−0.4151	0.9998	−1.7469	−0.4106	0.0060
95	0.9586	1.5227	0.9995	−1.7371	−0.4666	0.9997	−1.7067	−0.5013	−0.0455
98	0.9451	1.4423	0.9988	−1.6764	−0.5096	0.9995	−1.6141	−0.5807	−0.0933
99	0.9356	1.3710	0.9983	−1.6127	−0.5260	0.9993	−1.5366	−0.6129	−0.1138

0% to 90% the hygroscopic growth has caused increase in α and increase in the curvature from the quadratic part. As from 95%, α started decreasing and continued to decrease with the increase in RH despite the fact that α_2 continued to increase. This shows that as the deliquescence point increase the values of α continued to decrease. It also shows that hygroscopic growth enhances mode size growth. The increase in α_2 signifies the increase in the domination of fine particles. The cubic part shows that the mixture has bimodal type of distribution with the dominance of fine mode particles because the magnitude of $\alpha_1 > 1$.

Figure 8 shows that refractive indicies decrease with the increase in RH. It also shows that the non-sphericity increases with the increase in RH. This shows that increase in hygroscopic growth causes the particles to be more non-spherical with wavelengths.

Figure 9 shows that enhancement increases with the increase in hygroscopic growth and is also a function of wavelengths. Enhancement factor as a function of RH shows a nonlinear relation.

The results of the fitted curves of Equations (9) and (10) are presented as follows:

For one parameter (Equation (9))

At $\lambda = 0.25\mu$, $\gamma = 0.512704$, $R^2 = 0.9961$

At $\lambda = 1.25\mu$, $\gamma = 0.552129$, $R^2 = 0.9984$

At $\lambda = 2.50\mu$, $\gamma = 0.367149$, $R^2 = 0.9722$

For two parameters (Equation (10))

At $\lambda = 0.25\mu$, a = 1.204648, b = −0.454391, R^2 = 0.9992

At $\lambda = 1.25\mu$, a = 0.925221, b = −0.576471, R^2 = 0.9957

At $\lambda = 2.50\mu$, a = 0.737846, b = −0.462366, R^2 = 0.9753

Because of the very good correlations, they verify the non-linearity relation between the enhancements parameters and RH.

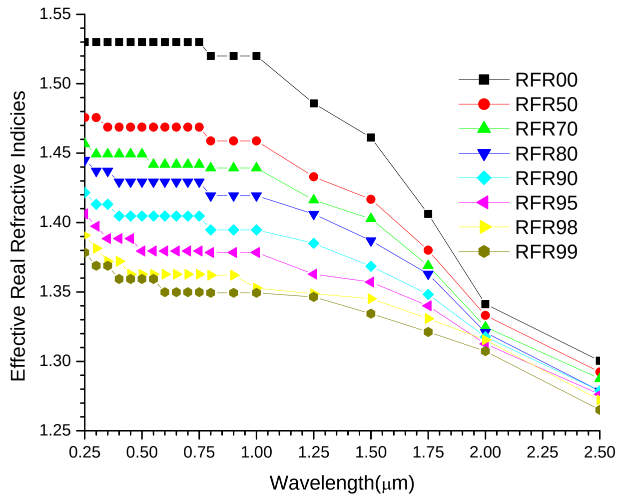

Figure 8. A plot of effective real refractive indices against wavelength.

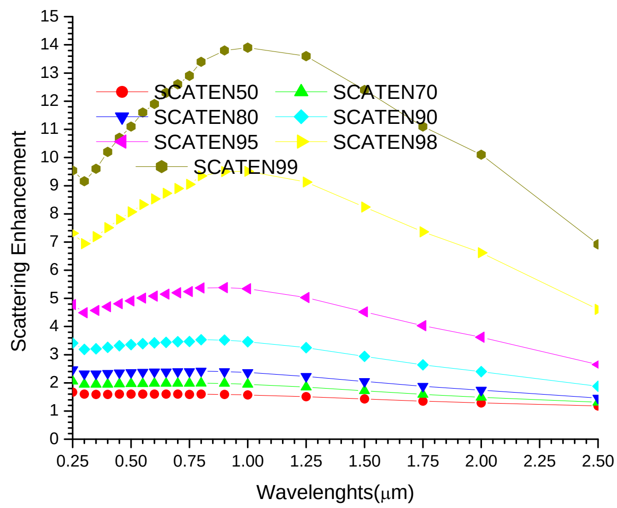

Figure 9. A plot of scattering enhancement parameters against wavelengths.

Figure 10 shows that absorption is barely dependent of hygroscopic growth at smaller wavelengths but increases as the wavelengths increases. This shows that larger particles absorbs more than smaller as the hygroscopic growth increases. The plots can be approximated by power law.

From **Table 6**, the values of α shows the dominance of coarse particles because it is less than 1. As the RH increases it also continued to decrease and the values of α_2 continued to increase and positive throughout. This shows that increase in hygroscopic growth increases the particle size and shows the dominance of the coarse particles. This also verifies bi-modal type of particle size distribution.

From **Figure 11**, the behavior of the effective imaginary refractive indicies with wavelengths shows the dominance of non-spherical particles. It also shows decrease in refractive indicies as a result of the increase in hygroscopic growth. Comparing **Figures 10** and **11** shows that particle has more dominance in absorption than the imaginary effective refractive indicies. This is because as a result of the decrease in the effective imaginery refractive indicies, we expect decrease in absorption instead of the increase as shown in **Figure 10**.

Figure 12 shows that the enhancement parameter increases with the increase in wavelengths and this implies increase with the increases of the particle size. This shows that it increases with the increase in particle size as observed in **Figure 10**.

Enhancement factor as a function of RH shows a nonlinear relation.

The results of the fitted curves of Equations (9) and (10) are presented as follows:

For one parameter (Equation (9))

At $\lambda = 0.25\mu$, $\gamma = 0.033611$, $R^2 = 0.9799$

At $\lambda = 1.25\mu$, $\gamma = 0.032435$, $R^2 = 0.9931$

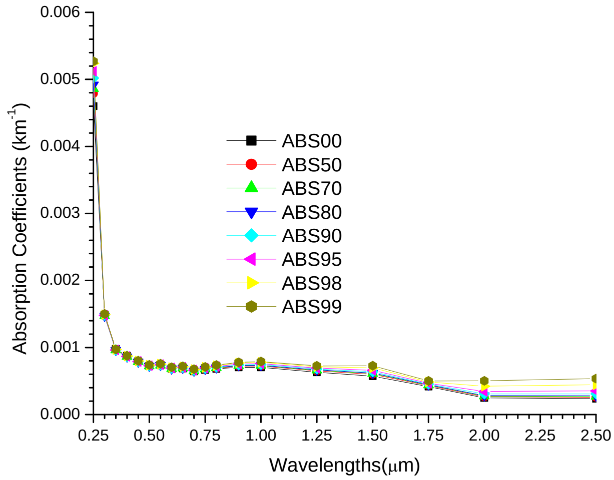

Figure 10. A plot of absorption coefficients against wavelength.

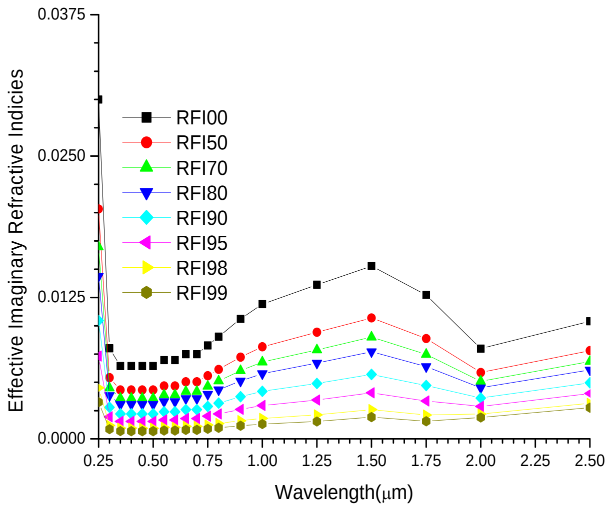

Figure 11. A plot of effective imaginary refractive indices against wavelength.

Table 6. The results of the Angstrom coefficients of absorption coefficients using Equations (12)-(14) for continental clean model at the respective relative humidities using regression analysis with SPSS16.0.

RH	Linear		Quadratic			Cubic			
(%)	R^2	α	R^2	α_1	α_2	R^2	α_1	α_2	α_3
0	0.7321	0.8145	0.7574	−0.7114	0.2243	0.9472	−0.0227	−0.5615	−1.0300
50	0.6988	0.7801	0.7326	−0.6633	0.2542	0.9397	0.0419	−0.5504	−1.0547
70	0.6866	0.7655	0.7252	−0.6419	0.2690	0.9368	0.0638	−0.5362	−1.0555
80	0.6757	0.7518	0.7195	−0.6214	0.2838	0.9343	0.0824	−0.5193	−1.0527
90	0.6537	0.7228	0.7108	−0.5773	0.3168	0.9293	0.1166	−0.4749	−1.0378
95	0.6226	0.6824	0.7026	−0.5158	0.3626	0.9219	0.1567	−0.4047	−1.0058
98	0.5596	0.6126	0.6940	−0.4082	0.4450	0.9083	0.2214	−0.2733	−0.9416
99	0.4973	0.5532	0.6912	−0.3180	0.5120	0.8954	0.2707	−0.1597	−0.8805

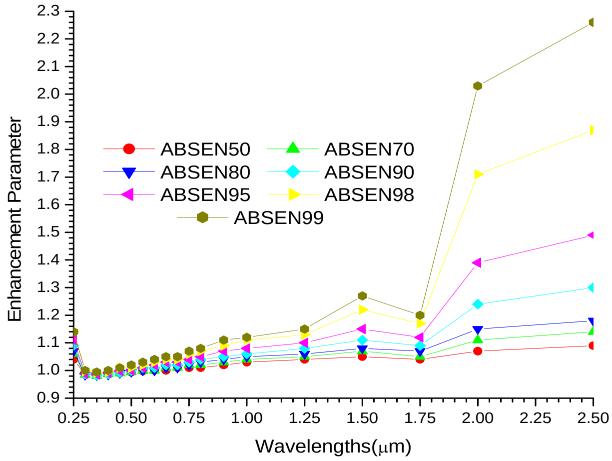

Figure 12. A plot of Absorption enhancement parameters against wavelengths.

At $\lambda = 2.50\mu$, $\gamma = 0.153640$, $R^2 = 0.9740$

For two parameters (Equation (10))

At $\lambda = 0.25\mu$, a = 1.027460, b = −0.025126, R^2 = 0.9845

At $\lambda = 1.25\mu$, a = 1.016064, b = −0.027444, R^2 = 0.9995

At $\lambda = 2.50\mu$, a = 0.894238, b = −0.188649, R^2 = 0.9669

Because of the very good correlations, they verify the non-linearity relation between the enhancements parameters and RH.

Figure 13 shows that hygroscopic growth causes smaller particles to scatter more in the forward and forward scattering decreases with the increase in particle size.

Figure 14 shows that hygroscopic growth causes enhancement in the forward direction to decrease with the increase in particle size.

The results of the fitted curves of Equations (9) and (10) are presented as follows:

For one parameter (Equation (9))

At $\lambda = 0.25\mu$, $\gamma = 0.029089$, $R^2 = 0.9291$

At $\lambda = 1.25\mu$, $\gamma = 0.027159$, $R^2 = 0.9826$

At $\lambda = 2.55\mu$, $\gamma = -0.060856$, $R^2 = 0.9717$

For two parameters (Equation (10))

At $\lambda = 0.25\mu$, a = 1.014671, b = −0.014565, R^2 = 0.9348

At $\lambda = 1.25\mu$, a = 0.980473, b = −0.033335, R^2 = 0.9920

At $\lambda = 2.50\mu$, a = 0.954266, b = 0.046195, R^2 = 0.9136

Because of the very good correlations, they verify the non-linearity relation between the enhancements parameters and RH.

4. Conclusions

From the gf_{mix}(RH) determined, it can be observed that despite the higher fractions of more strongly absorbing particles, very low values of gf_{mix}(RH) were observed, and this is in line with what Sheridan *et al*. [24] determined.

It shows that increase in RH increases forward scattering because particle growth enhances forward.

These hygroscopic growth behaviors also reveal an immense potential of light scattering enhancement in the forward scattering [65] for smaller particles while in larger particles it causes increase in the backward scattering at high humidities and the potential for being highly effective cloud condensation nuclei.

It also shows that the mixture is internally mixed for smaller particles because of the increase in forward scattering as a result of the hygroscopic growth [66].

Field measurements have noted a k value of 0.01 for fresh soot rich biomass [67]. The overall, modeled k ranges from 0.012 to 0.163 depending on the RH and the

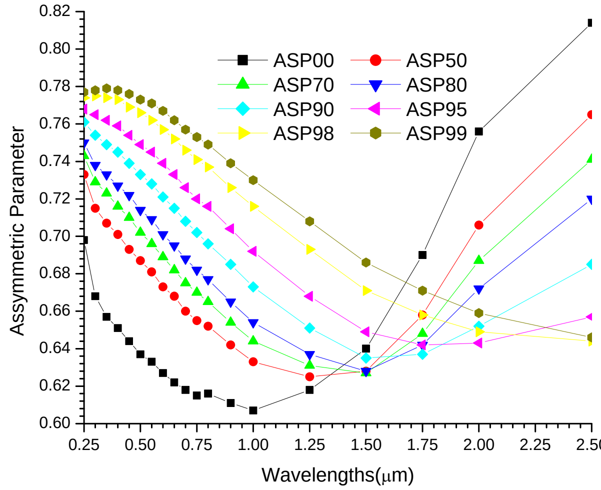

Figure 13. A plot of Asymmetric parameter against wavelength.

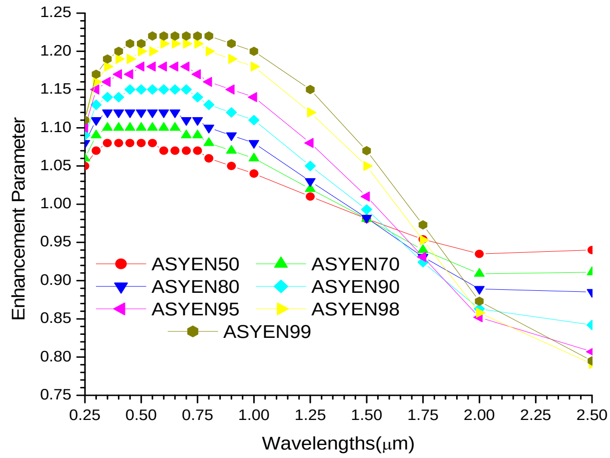

Figure 14. A plot of Asymmetric parameter enhancement parameters against wavelengths.

type of the mixing ratio used.

Finally, it can be observed that the absorption and scattering coefficients together with their enhancement parameters have exponential dependence with RH.

The modeling shows that hygroscopic growth at higher relative humidity increases the effective radii, scattering coefficients, scattering enhancement parameters, absorption coeffeicnts, absorption enhancement parameters, but decreases effective real refractive indices, effective imaginary refractive indices. However, the asymmetric and enhancement asymmetric parameters increase with the increase in RH but decreases with the increase in wavelength.

Jeong *et al*. [10] demonstrated an exponential dependence of the aerosol optical thickness on relative humidity.

Finally, the data fit our models very well and can be used to extrapolate the hygroscopic growth at any RH and enhancement parameters at any RH and wavelengths. The importance of determining $gf_{\mathrm{mix}}(\mathrm{RH})$ as a function of RH and volume fractions, mass fractions and number fractions, and enhancement parameters as a function of RH and wavelengths can be potentially important because it can be used for efficiently representing aerosols-water interactions in global models.

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