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ORIGINAL RESEARCH ARTICLE

Microphysical Analysis of Maritime and Continental Aerosols: Determination of Hygroscopic Growth Factors Using OPAC Data and Modeling Approaches

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ABSTRACT
The atmospheric aerosols over coastal areas are governed by the interplay of marine and continental sources. The transport of aerosols from continental sources into sea surfaces through deposition or diffusion is what causes the fast reduction of continental aerosols. A mass based hygroscopicity models were applied to the data extracted from the Optical Properties of Aerosols and Clouds (OPAC). The microphysical properties obtained were radii, density, refractive index, mass, volume, and sphericity of the atmospheric aerosols of continental and maritime aerosols at eight different relative humidity of 0%, 50%, 70%, 80%, 90%, 95%, 98%, and 99%. Using the microphysical properties, hygroscopic growth factors, and effective radii of the mixtures, mass growth factor G_m and diameter growth factor D_G were determined, and also the parameter K_m for mass based of the aerosols were determined using multiple regression analysis with SPSS 16.0 at each relative humidity. The results show that G_m for maritime clean is higher than other aerosols, with a value of 34.46 at 99% RH, while the lowest value is for continental average, with a value of 5.03 at 99% RH. Also, R^2 for the model is greater than 90%. The significance and P-values are less than 0.05; therefore, the model is good for atmospheric modeling and remote sensing.

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Hygroscopicity, maritime, continental, atmospheric aerosols, OPAC.

INTRODUCTION
Aerosols are suspensions of small liquid or solid particles in air or gas with aerodynamic diameters ranging from 10^{-3} µm to 100µm. Aerosols play a crucial role in climate and environment at both regional and global scales. The sources of aerosols include vehicle emissions (soot/black carbon, organics), industrial activities (soot/black carbon, sulphate, organics, metals, and nitrate), construction and agriculture emission (soot/black carbon, nitrate, and soil), sea-spray (salt), biomass and fires (soot/black carbon, nitrate and organics) (Sun *et al.,* 2013). By dispersing and absorbing solar radiation, aerosols in the atmosphere can directly affect climate. Alternatively, they can indirectly affect climate by acting as cloud condensation nuclei (CCN), which aid in the formation and reflection of clouds (Twomey, 1977). By reflecting, absorbing, and facilitating the development of clouds, atmospheric aerosol particles have an impact on Earth's overall radiation budget (Boucher et al., 2013).1 Accurate knowledge of the physical characteristics of atmospheric aerosol particles is necessary for the quantification of this influence within the context of theoretical climate models (Hess et al., 1998). They include their optical characteristics, their capacity to serve as cloud condensation nuclei, as well as their hygroscopicity, or capacity to absorb water from their surroundings. Hygroscopic particles' mass and size can change dramatically depending on the relative humidity (RH) of their surroundings, which in some cases, such as with some inorganic salt aerosol particles, results in a change in their scattering cross-section by an order of magnitude (Zieger et al., 2013). Understanding the hygroscopic properties of aerosols and the processes that govern cloud droplet activation is important. Koehler's theory has been used to predict the CCN-activity of inorganic compounds for many years, and the hygroscopicity also plays an important role in the activation of cloud condensation nuclei, as described by Köhler theory, which facilitates cloud formation (Koehler, 1936).

New measurements were made of the hygroscopic mass growth of sea salt and single NaCl particles, together with values for their densities as a function of relative humidity (Oliver et al., 2023). The earth's radiation balance, local radiative forcing, and climate are all impacted by the optical characteristics of particulate atmospheric constituents, such as water and ice clouds and aerosol particles (Hess et al., 1998). The mass growth factor was

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determined for Antarctic, arctic, desert, and urban aerosols. It found that there is an increment in the mass growth factors as relative humidity increases, but there is much increment for the Antarctic and Arctic at 98%and 99% relative humidity (Sa'adu et al., 2020)

The study aims to determine the mass growth and diameter growth factors of the atmospheric aerosols in the maritime and continental regions, and the objective is to analyze the microphysical features of the atmosphere, such as radii, refractive index, volume, mass, density, and mass-based, using data from the optical properties of aerosols and clouds (OPAC).

METHODOLOGY

Koehler theory

The saturation ratio, S, over an aqueous solution droplet, can be calculated from

$$S = a_w exp\left(\frac{4\,\sigma_{s/a}\,M_w}{RT\rho_w D}\right) \quad (1a)$$

$$s_w \approx a_w exp\left(\frac{4\sigma_w M_w}{RT\rho_w D}\right) \quad (1b)$$

where a_w is the activity of water in the solution, ϱ_w is the density of water, M_w is the molecular weight of water, $\sigma_{s/a}$ is the surface tension of the solution/air interface, R is the universal gas constant, T is temperature, and D is the diameter of the droplet. In our proposed parameterization, the mass-based hygroscopicity parameter k_m is defined through its effect on the water activity of the solution:

$$k_m = \left(\frac{m_w}{m_d}\right)\left[\frac{1-a_w}{a_w}\right] \quad (2)$$

Where m_d is the mass of the dry particle material, m_w is the mass of water in the wet particle (aqueous droplet), and a_w is the water activity.

However, since atmospheric aerosols are typically mixtures of soluble components, the data on hygroscopicity modes was combined to create $g_{eff}(S)$, a representative particle population for all aerosols, which is the "overall," "bulk," or "effective" hygroscopic growth factor of the mixture.

$$g_{eff}(S) = \left(\sum_k x_k g_k^3\,(S)\right)^{1/3} \quad (3)$$

The effective or volume-equivalent radius of the mixture was determined using the relation.

$$r_{eff}(S) = \left(\sum_k x_k r_k^3\right)^{1/3} \quad (4)$$

where the Zdanovskii-Stokes-Robisnson relation (ZSR relation) is used to accomplish the summing across all compounds contained in the particles, with Xk representing each compound's corresponding volume percentage; Sjogren et al., 2007; Stokes and Robinson 1966; Meyer et al., 2009; Stock et al., 2011).

The aerosol's hygroscopicity growth factor g(S) (Swietlicki et al.,2008; Randles et al.,2004) is defined as:

$$g(S) = \frac{r(S)}{r(S=0)} \quad (5)$$

Where S is taken for eight values 0%, 50%,70%,80%,90%,95%,98% and 99% RH

Since mass m is proportional to r^3, this implies that g_m is proportional to g_{eff}^3, therefore:

$$lnS = \frac{A}{(g_m)^{1/3}} + \frac{B}{1-g_m} \quad (6)$$

Using multiple regression analysis with SPSS 16.0 for Windows, the constants A and B were determined.

The first term on the right-hand side of equation (6) can be written as

$$lnk_e = \frac{A_1}{(g_m)^{1/3}} \quad (7a)$$

This implies that

$$k_e = exp\left(\frac{A}{(g_m)^{1/3}}\right) \quad (7b)$$

The second term on the right-hand side of equation (6) is

$$lna_w = \frac{B}{1-g_m} \quad (8a)$$

This implies

$$a_w = exp\left(\frac{B}{1-g_m}\right) \quad (8b)$$

The diameter growth factor G_d is defined as the ratio between the volume equivalent diameters of the aqueous droplet and of the dry aerosol particle.

$$G_D = \frac{D}{D_d} \quad (9)$$

By defining the mass growth factor G_m as

$$G_m = \frac{m_w + m_d}{m_d} \quad (10)$$

And combining Eq. (2) and Eq. (10), we obtain

$$\frac{1}{a_w} = \left(\frac{k_m}{G_m - 1} + 1\right) \quad (11)$$

According to Mikhailov et al. (2013), the volume-based formulation is identical to the mass-based relationship between particle hygroscopicity and water activity seen in Eq. (11). However, because of mass conservation, deviations from spherical geometry and volume additivity, which typically restrict the applicability and precision of volume-based parameters determined in mobility diameter-based HTDMA and CCN experiments, do not affect the practical applicability of Eq. (11) and the precision of related parameters determined in mass-based experiments. (Kramer et al., 2000; Gysel et al., 2002, 2004; Rose et al., 2008; Mikhailov et al., 2004, 2009; Wang et al., 2010). The hygroscopicity parameter km (Eq. 11) is

defined as mass-based, avoiding a problem that arises in the volume-based definition of κv (Mikhailov et al., 2013). It is simple to convert the masses of dry particles and aqueous droplets into volume-equivalent diameters if the densities of the dry solute and the solution are known:

$$D_d^3 = {}^{6m_d}/_{(\pi\rho_d)}, D^3 = {}^{6G_m m_d}/_{(\pi\rho)}$$

$$D = D_d \left({}^{G_m\rho_d}/_{\rho}\right)^{1/3} \text{or}$$

$$\frac{D}{D_d} = \left({}^{G_m\rho_d}/_{\rho}\right)^{1/3} = G_d \quad (12)$$

Under the assumption of volume additivity, Eq. (9) and Eq. (11) can be combined to

$$\frac{G_d^3}{G_m} = \frac{\rho_d}{\rho} = \frac{\rho_d}{\rho_w} \quad (13)$$

Equation (13) is the relation between mass growth and diameter growth factor, which is equivalent to the ratio of the density of the dry particle and the wet particle.

Or

$$r_{eff} = r_d \left({}^{G_m\rho_d}/_{\rho}\right)^{1/3} \quad (14)$$

Where ϱ is the density of the aqueous solution droplet. For dilute aqueous solution droplets with $\varrho \approx \varrho_w$ follows

$$D = D_d \left({}^{\rho_d}/_{\rho_w}\right)^{1/3} \quad (16)$$

By inserting Eq. (12) and Eq. (13) in Eq. (1b), we obtain an approximate mass- based κ_m -K¨ohler equation:

$$\frac{RH}{100\%} = S_W \approx \left(\frac{k_m}{G_{m-1}} + 1\right)^{-1} exp\left(\frac{4\sigma_w M_w}{RT\rho_w D_d}\left[\frac{\rho_w}{\rho_d G_m}\right]^{1/3}\right) \quad (17)$$

Table 1: Some microphysical properties and composition of atmospheric aerosols extracted from OPAC at 0 RHs (Hess et al., 1998)

Model Number	Aerosols Model Types	Aerosols Components	Number Concentrations (cm^{-3})	Rmin (μm)	Rmax (μm)	Sigma σ	Rmod (μm)
1	Continental Average	Inso	0.4000	0.0050	20.0000	2.5100	0.4710
		Waso	7,000.0000	0.0050	20.0000	2.2400	0.0212
		Soot	8,300.0000	0.0050	20.0000	2.0000	0.0118
2	Continental Clean	Waso	2,600.0000	0.0050	20.0000	2.2400	0.0212
		Inso	0.1500	0.0050	20.0000	2.5100	0.4710
3	Continental Polluted	Inso	0.6000	0.0050	20.0000	2.5100	0.4710
		Waso	15,700.0000	0.0050	20.0000	2.2400	0.0212
		Soot	34,300.0000	0.0050	20.0000	2.0000	0.0118
4	Maritime Clean	Waso	1,500.0000	0.0050	20.0000	2.2400	.0212
		Ssam	20.0000	0.0050	20.0000	2.0300	0.2090
		Sscm	0.0032	0.0050	60.0000	2.0300	1.7500
5	Maritime Polluted	Waso	3,800.0000	0.0050	20.0000	2.2400	0.0212
		Soot	5,180.0000	0.0050	20.0000	2.0000	0.0118
		Ssam	20.0000	0.0050	20.0000	2.0300	0.2090
		Sscm	0.0032	0.0050	60.0000	2.0300	1.7500
6	Maritime Tropical	Waso	590.0000	0.0050	20.0000	2.2400	0.0212
		Ssam	10.0000	0.0050	20.0000	2.0300	0.2090
		Sscm	0.0013	0.0050	60.0000	2.0300	1.7500

Aerosols may be shown to be soluble or insoluble by the sol and ns values; the inso indicates the portion of aerosol particles that are insoluble in water and is primarily composed of soil particles that include a small amount of organic material. The waso, which is made up of different types of sulfates, nitrates, and other organic water-soluble compounds, is the water-soluble portion of aerosol particles that are produced during the gas-to-particle conversion process. Thus, it comprises more than only the anthropogenic aerosol commonly referred to as sulfate aerosol. Black carbon that has been absorbed is shown by the soot component. The particles are thought to not develop with increased relative humidity because carbon is not soluble in water. The coarse modes and sea-salt accumulation particles known as the ssam and sscm are made from with less big particles present. It is expected that the size of mineral aerosol particles does not increase as relative humidity rises. The suso describes the quantity of sulfate present in Antarctic aerosol as a sulfate component (75% H_2SO_4). Mineral aerosols, also known as desert dust, are formed in dry environments and are identified as mineral (nucleation mode) MINM, mineral (accumulation mode) MIAM, and mineral (coarse mode) MICM (Tijjani B.I. et al., 2015).

RESULT AND DISCUSSION

As can be seen from Table 2, the data match the equation quite well, with the mass-based hygroscopicity parameter increasing in the ocean zone and decreasing in the continental region.

Table 3 indicates that, based on the values of R^2, the data fitted the equation very well.

According to Table 4, mass growth increases marginally for both continental and marine aerosols as relative humidity rise, while for maritime aerosols, mass growth increases significantly between 98% and 99% of relative humidity.

The result of Table 5 above indicates that the diameter growth factor slightly increases as the relative humidity increases, but there are high increments for maritime clean at 80% relative humidity.

From Table 6, it can be seen that there is are good linear relationship between the ratio of densities and the ratio of diameter growth and mass growth for only Maritime polluted aerosols and are non-linear for continental average, continental clean, continental polluted, maritime clean and maritime tropical aerosols, their non-linearity is due to the compositions of the aerosols.

Figure 1 demonstrates that mass increase is less significant at high relative humidity and exhibits a sharp curve at the 90% to 99% deliquescence stage for all aerosols. Although there is a minor rise in the mass growth of the particle at low relative humidity, indicating that the particle is more hygroscopic, they are more sensitive at the deliquescence threshold.

Figure 2 displays the aerosol diameter growth factor. There are notable increases for both tropical and marine dirty waters, and it rises when relative humidity rises as well. Regardless of relative humidity, the continental aerosols follow a linear pattern. The maritime clean has indicated that it is independent between 0 and 70% RH and dependent between 70 and 99% RH. It has diverged at 70 RH.

The Kelvin effect of the aerosols in Figure 3 shows its effect over the entire aerosol, but the effect is high in the maritime region by considering the curve of the maritime clean, maritime polluted, and maritime tropical due to the composition of the aerosols.

Figure 4 shows the water activity of the aerosols, which increase as the relative humidity increases. The maritime aerosol shows a curve at 70 relative humidity, while the continental aerosols start at 90 relative humidity.

Table 2: Result of mass-based hygroscopicity using equation (11)

Aerosol models	k_m	p-value	R^2	Significance
Continental average	0.2147	3.69E-10	0.99894	7.82E-09
Continental clean	0.21995	3.75E-10	0.99894	7.93E-09
Continental Polluted	0.25542	4.16E-10	0.9989	8.65E-09
Maritime clean	1.77645	9.53E-10	0.99855	1.73E-08
Maritime polluted	1.52779	8.13E-10	0.99863	1.51E-08
Maritime tropical	1.79002	8.81E-10	0.99859	1.62E-08

Table 3: Results of Kelvin radii and bulk hygroscopicity for mass based using equation (6)

Aerosol models	Kelvin radius A	p-value	Bulk hygroscopicity B	p-value	R^2	Significance
Continental Average	0.69666	4.02E-05	0.206747	1.04E-07	0.9994	2.22E-07
Continental Clean	0.708339	3.77E-05	0.21178	1.01E--07	0.9994	2.12E-07
Continental Polluted	0.7843	1.92E-05	0.245763	6.12E-08	0.9996	1.24E-07
Maritime clean	4.480338	1.69E-05	1.710696	8.65E-07	0.9995	1.55E-07
Maritime polluted	3.854419	1.69E-05	1.472411	7.10E-07	0.9995	1.50E-07
Maritime tropical	4.468648	9.99E-06	1.724686	4.92E-07	0.9996	9.38E-08

Table 4: Result of mass growth G_m using equation (10)

Aerosols model	Rh (50%)	Rh(70%)	Rh(80%)	Rh(90%)	Rh(95%)	Rh(98%)	Rh(99%)
Continental Average	1.236829	1.385432	1.543329	1.920525	2.525279	3.77555	5.032013
Continental Clean	1.241913	1.393445	1.555051	1.939557	2.558173	3.835131	5.117764
Continental polluted	1.275721	1.448371	1.632292	2.070811	2.774904	4.229674	5.691404
Maritime clean	2.287786	2.997474	3.774385	5.921243	9.852812	20.29097	34.46068
Maritime polluted	2.144613	2.7688	3.463892	5.337581	8.787509	17.74187	29.85679
Maritime tropical	2.27232	2.959099	3.72752	5.811085	9.685498	19.83565	33.72816

Table 5: Result of diameter growth factor using equation (9)

Aerosols model	Rh(50%)	Rh(70%)	Rh(80%)	Rh(90%)	Rh(95%)	Rh(98%)	Rh(99%)
Continental Average	1.009921	1.014484	1.018651	1.026984	1.037103	1.052381	1.063889
Continental Clean	1.010158	1.014831	1.019098	1.027631	1.037993	1.053637	1.065421
Continental Polluted	1.009921	1.014484	1.018651	1.026984	1.037103	1.052381	1.063889
Maritime Clean	0.013231	0.014392	1.987981	2.379457	2.906222	3.882739	4.867387
Maritime Polluted	1.603414	1.801355	1.982129	2.371285	2.894930	3.865663	4.844478
Maritime Tropical	1.607009	1.806131	1.987981	2.379457	2.906222	3.882739	4.867387

Table 6: Results of the ratio of densities and growth factors using equation (13)

Aerosols model	ϱ_d/ϱ_w	G_d^3/G_m
Continental Average	1.85275	0.44158
Continental Clean	1.89576	0.43699
Continental polluted	1.78945	0.40223
Maritime tropical	2.15708	1.06050
Maritime polluted	2.06046	2.11356
Maritime clean	2.14968	1.92081

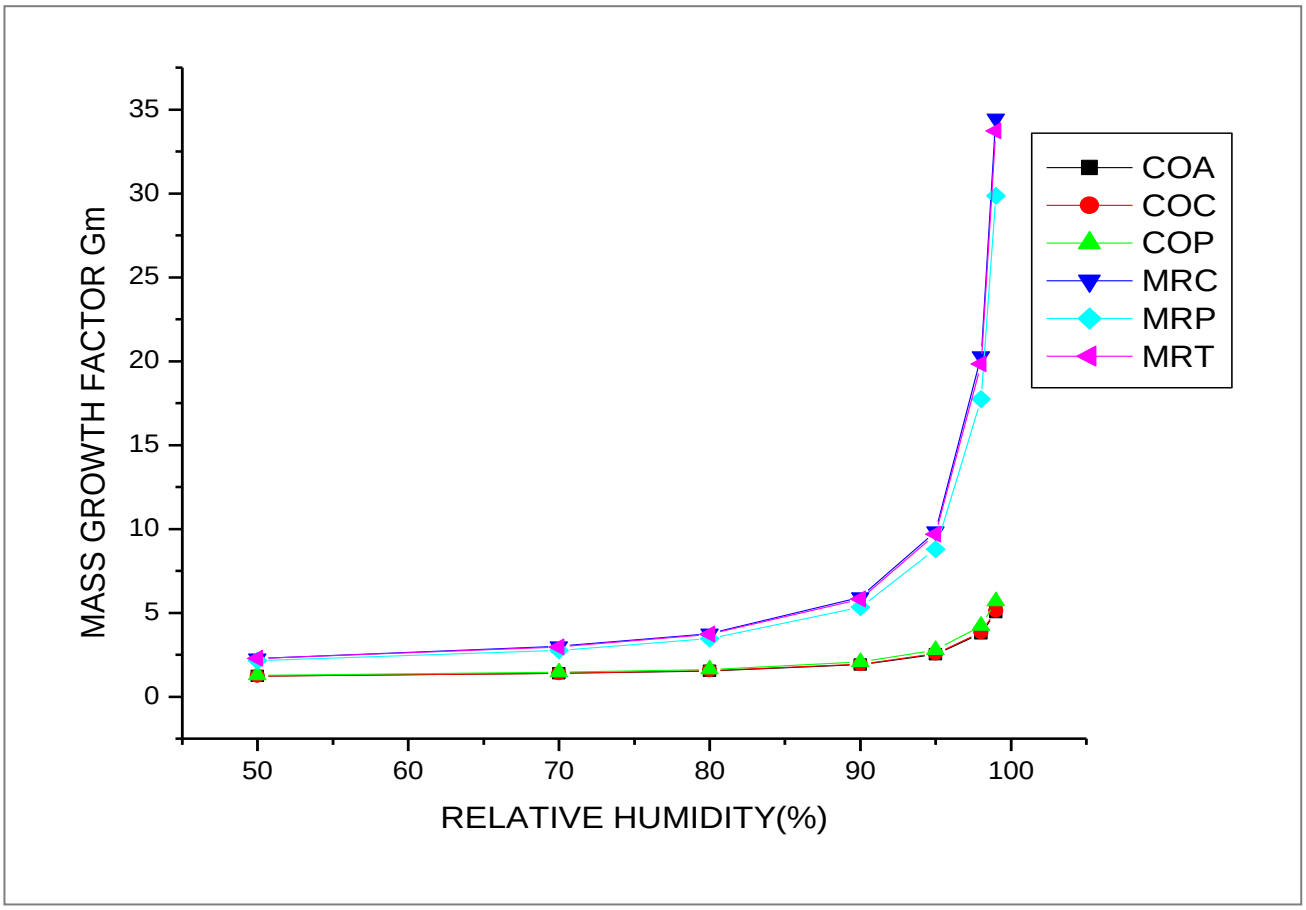

Figure 1: Graph of mass growth factor against relative humidity

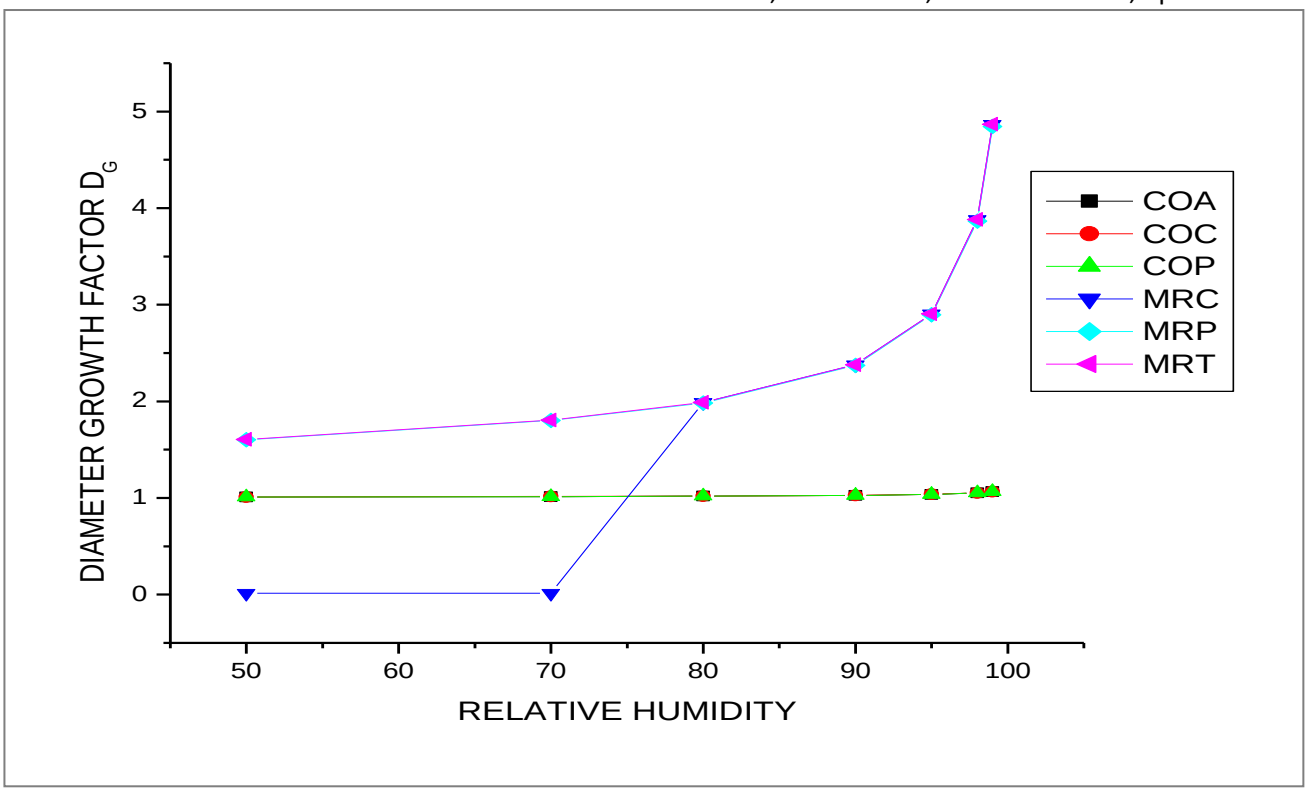

Figure 2: Graph of diameter growth factor against relative humidity

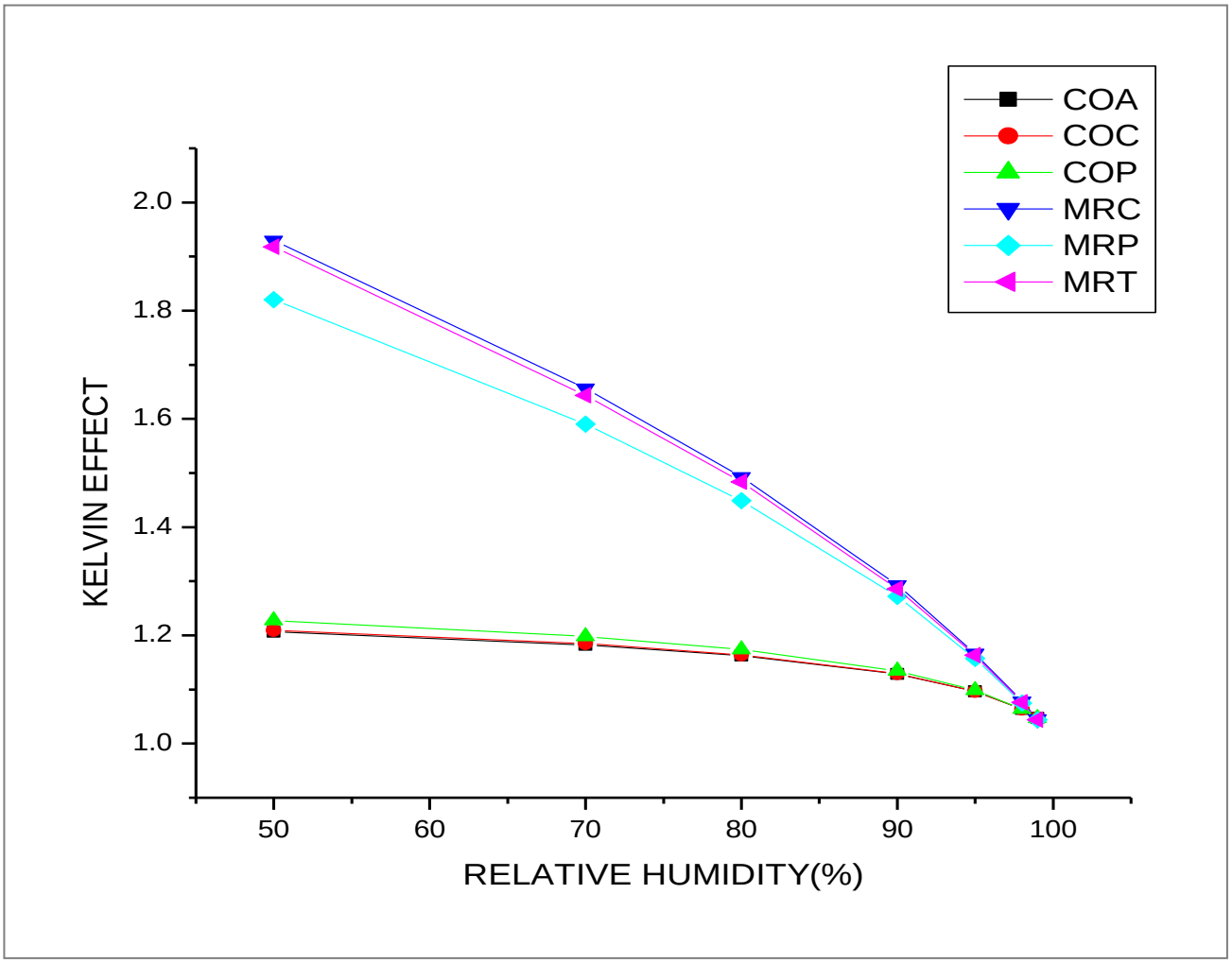

Figure 3: Graph of Kelvin effect against relative humidity

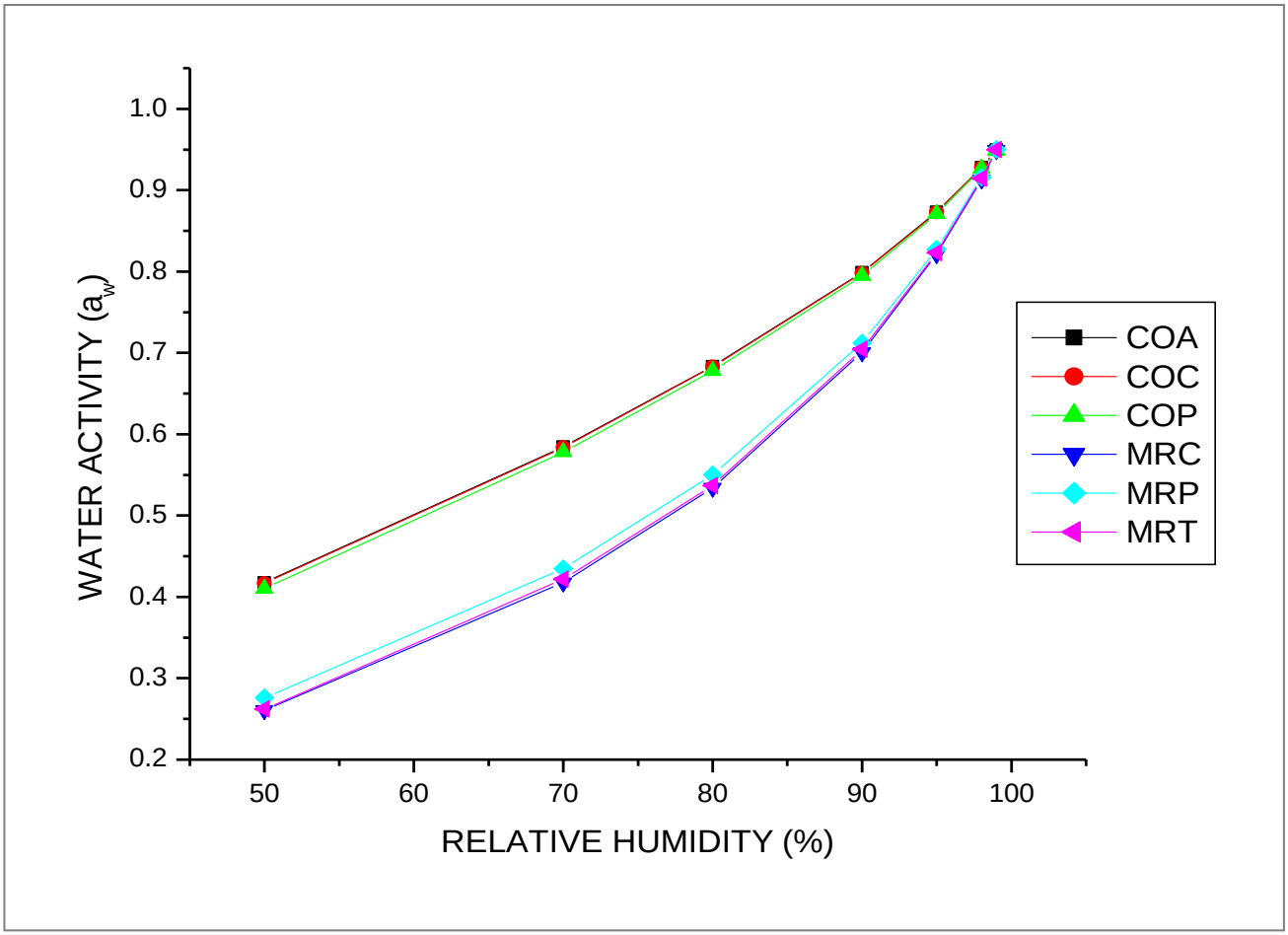

Figure 4: Graph of water activity against relative humidity

CONCLUSION

The mass growth and diameter growth of the atmospheric aerosols over the maritime and continental regions were determined. The result indicates that there is are increase of both mass growth and diameter growth as the relative humidity increases in the region. The mass-based hygroscopicity parameter k_m fitted the equation obtained using Kholer theory and the data extracted from OPAC by considering the value of coefficient relation $R^2 > 90$. The significance and p-value are less than 0.05. There is are good linear relationship between the ratio of densities and the ratio of diameter growth and mass growth for only Maritime polluted aerosols, and are non-linear for continental average, continental clean, continental polluted, maritime clean, and maritime tropical aerosols. Their non-linearity is due to the compositions of the aerosols.

Figure 1 shows that the mass growth has less effect at high relative humidity and shows a steep curve at the deliquescence point (90% to 99%) of all aerosols. They are more sensitive at the deliquescence point; however, there is a slight increase in the mass growth of the particle at low relative humidity, meaning the particles at this region are more hygroscopic. The regression analysis result shows that p-values and significance are less than 0.05; the R-square is greater than 90% for all the models, which means they can be used for atmospheric modeling.

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