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INVESTIGATION OF THE DENSITY OF AEROSOLS AND THE CORRELATION BETWEEN MASS-VOLUME BASED HYGROSCOPICITY PARAMETERS FOR ATMOSPHERIC AEROSOLS

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ABSTRACT

In this paper, the Optical Properties of Aerosols and Clouds (OPAC) data was retrieved and applied to the mass-based and volume-based hygroscopicity parameters models. The atmospheric aerosols of the continent-polluted (COP), maritime-polluted (MRP), Antarctic-polluted (ANT), urban-polluted (URB), and desert-polluted (DST) at eight different relative humidity levels of 0%, 50%, 70%, 80%, 90%, 95%, 98%, and 99% were measured for microphysical properties, including radii, density, refractive index, mass and volume. Effective radii of the mixtures were calculated using the microphysical characteristics, hygroscopic growth factors, and multiple regression analysis with SPSS 16.0 to determine the parameter Km for mass-based and Kv for volume-based aerosols at each relative humidity. For COP, MRT, ANT, URB, and DST, the corresponding Kv/Km ratios are 1.78, 1.33, 1.81, 1.86, and 4.41, In contrast, the density ρd /ρw values are 1.79, 2.06, 1.76, 1.74, and 2.59, in that order. The results indicates that, based on the correlation coefficient R^2>90, significance<0.05 and p-value, the two models are good for atmospheric modeling.

Keywords: Atmospheric aerosols, desert, hygroscopicity, OPAC, maritime polluted

INTRODUCTION

Aerosols in the atmosphere have the ability to directly impact climate change through their absorption and dispersion of solar energy, or indirectly through their function as cloud condensation nuclei (CCN), which promotes cloud formation and reflection (Twomey, 1977). By reflecting, absorbing, and encouraging cloud formation, airborne aerosol particles have an impact on Earth's overall radiation budget (Boucher *et al.*, 2013).Precise knowledge of the physical characteristics of atmospheric aerosol particles is necessary for measuring this influence within theoretical climate models (Hess *et al.*, 1998).These include their hygroscopicity, or capacity to absorb water from their surroundings, their optical characteristics, and their capacity to function as cloud condensation nuclei. Hygroscopic particles can differ greatly in size and mass. Aerosols in the atmosphere can directly or indirectly affect the climate by dispersing and absorbing solar radiation through their function as cloud condensation nuclei (CCN), which aid in the creation and reflection of clouds (Twomey, 1977). Because atmospheric aerosol particles reflect, absorb, and encourage the formation of clouds, they have an

impact on Earth's overall radiation budget (Boucher *et al.*, 2013). These include their hygroscopicity, or capacity to absorb water from their surroundings, their optical characteristics, and their capacity to function as cloud condensation nuclei. Particles that are hygroscopic can display significant variations in size and mass. Depending on their surroundings' relative humidity (RH), some inorganic salt aerosol particles' scattering cross-sections can occasionally vary by an order of magnitude (Zieger *et al.*, 2013). Understanding the hygroscopic properties of aerosols and the mechanisms governing cloud droplet activation are critical. For many years, Koehler theory has been used to predict the CCN-activity of inorganic substances. According to Koehler (1936), hygroscopicity is also essential for the activation of cloud condensation nuclei, which increases cloud production.

The hygroscopic mass increase of individual NaCl and sea salt particles, along with their densities as a function of relative humidity, were measured for the first time (Oliver *et al.*, 2023).

The optical characteristics of particulate air constituents, such as water and ice clouds and aerosol particles, have an impact on climate, local radiative forcing, and the earth's radiation balance (Hess *et al*, 1998).

The aim of the study is to explore the atmospheric aerosols composition from the software OPAC and the objective of the study is to analyze the aerosol density in relation to mass and volume-based hygroscopicity parameters. Data from the optical properties of aerosols and clouds (OPAC) was used to analyze the microphysical features of the atmosphere, including radii, refractive index, volume, mass, density, and mass-based information.

METHODOLOGY

Kohler theory is the one of the theories used to determine the saturation ratio S, over an aqueous solution droplet and is given by equation (1)

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

$$s_w \approx a_w exp\left(\frac{4\sigma_w M_w}{RT\rho_w D}\right) \quad (2)$$

where a_w is the activity of water in 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 by equation (3)

$$k_m = \left(\frac{m_w}{m_d}\right)\left[\frac{1-a_w}{a_w}\right] \quad (3)$$

where m_d and m_w represent the mass of the dry particle material and mass of the water in the wet particle (aqueous droplet) respectively. However, since atmospheric aerosols typically consisted of mixtures of soluble components, the data on the hygroscopicity modes was combined to create $g_{eff}(S)$, a representative value for the total population of aerosol particles, which is the "over-all," "bulk," or "effective" hygroscopic growth factor of the mixture.

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

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

$$r_{eff}(S) = \left(\sum_k x_k r_k^3\right)^{1/3} \quad (5)$$

Meyer et al. (2009), Stock *et al.* (2011), Sjogren *et al.* (2007), Stokes and Robinson (1966), and Zdanovskii-Stokes-Robinson connection (ZSR relation) are used to do the summation across all chemicals contained in the particles, with Xk representing their respective volume fractions.

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

$$g(S) = \frac{r(S)}{r(S=0)} \quad (6)$$

Where S is taken for different values of relative humidity 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:

$$lnS = \frac{A}{(g_m)^{1/3}} + \frac{B}{1-g_m} \quad (7)$$

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 (7) can be written as

$$lnk_e = \frac{A_1}{(g_m)^{1/3}} \quad (8a)$$

This implies that

$$k_e = exp\left(\frac{A}{(g_m)^{1/3}}\right) \quad (8b)$$

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

$$lna_w = \frac{B}{1-g_m} \quad (9a)$$

This implies

$$a_w = exp\left(\frac{B}{1-g_m}\right) \quad (9b)$$

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

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

Mikhailov *et al*. (2013) found that the volume-based formulation is similar to the mass-based relationship between particle hygroscopicity and water activity in Eq. (11). Deviations from spherical geometry and volume additivity which typically limit the applicability and precision of volume-based parameters do not determine in mobility diameter-based HTDMA and CCN experiments. However, it affect the practical applicability of Eq. (11) and the precision of related parameters determined in mass-based experiments due to mass conservation (Kramer *et al.*, 2000; Gysel *et al.*, 2002, 2004; Rose *et al.*, 2008; Mikhailov *et al.*, 2004, 2009; Wang *et al.*, 2010). A problem that arises in the volume-based determination of κv (Mikhailov *et al.*, 2013) is avoided by using the mass-based definition of the hygroscopicity parameter κm (Eq. 11).

Volume-based hygroscopicity parameter Kv

The volume-based hygroscopicity parameter κ_v relates the volume of the dry aerosol particle (V_d) to the volume of water (V_w) and the activity of water (a_w) in the aqueous droplet can be calculated according to equation (12):

$$k_v = \left(\frac{V_w}{V_d}\right)\left[\frac{1-a_w}{a_w}\right] \quad (12)$$

Or

$$\frac{1}{a_w} = \left[\frac{k_v}{g_{eff}^3-1} + 1\right] \quad (13)$$

Equations (3) and (12) can be merged, assuming volume additivity, to

$$k_m = k_v \frac{\rho_w}{\rho_d} \text{ Or } \frac{k_v}{k_m} = \frac{\rho_d}{\rho_w} \quad (14)$$

Equation (14) is the relation between mass and volume-based hygroscopicity parameter which is equivalent to the ratio of the density of dry particle and the wet particle. If the densities of the dry solute and of the solution are known, the masses of dry particles and aqueous droplets can be easily converted into volume equivalent diameters:

$$D_d^3 = {6m_d}/{(\pi\rho_d)}, D^3 = {6G_m m_d}/{(\pi\rho)} \text{ and } D = D_d\left({G_m\rho_d}/{\rho}\right)^{1/3}, \quad (15)$$

$$r_{eff} = r_d\left({G_m\rho_d}/{\rho}\right)^{1/3} \quad (16)$$

ρ represent the density of the aqueous solution droplet. For dilute aqueous solution droplets with $\rho \approx \rho_w$ follows

$$D = D_d\left({\rho_d}/{\rho_w}\right)^{1/3} \quad (17)$$

By inserting Eq. (12) and Eq. (13) in Eq. (2), we obtain an approximate mass- based κ_m -K¨ohler equation:

$$\frac{RH}{100\%} = S_W \approx \left(\frac{k_m}{G_{m-1}} + 1\right)^{-1} exp\left(\frac{4\sigma_w M_w}{RT\rho_w D_d}\left[\frac{\rho_w}{\rho_d G_m}\right]^{1/3}\right) \quad (18)$$

RESULTANDDISCUSSION

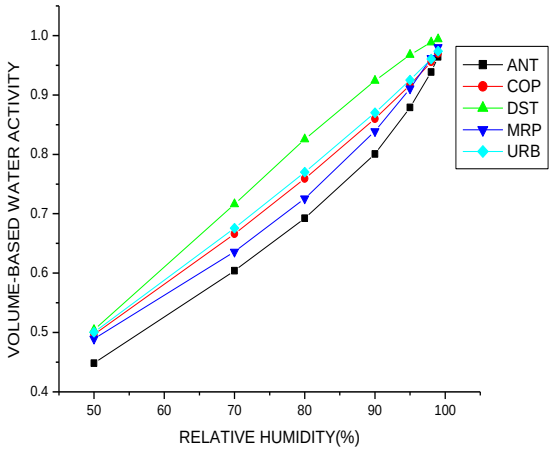

Figure 1: Graph of volume-based water activity against relative humidity.

The ambient RH in figure 1 was used to examine how the aerosols' ambient RH is impacted by the volume-based water activity. It is evident that the water activity reduces the ambient relative humidity in the Antarctic, the maritime polluted regions, and urban while the continental polluted regions' ambient relative humidity is linear and constant. Furthermore, Sa'adu (2020) reported that the water activity of desert aerosols is increased dependent on volume.

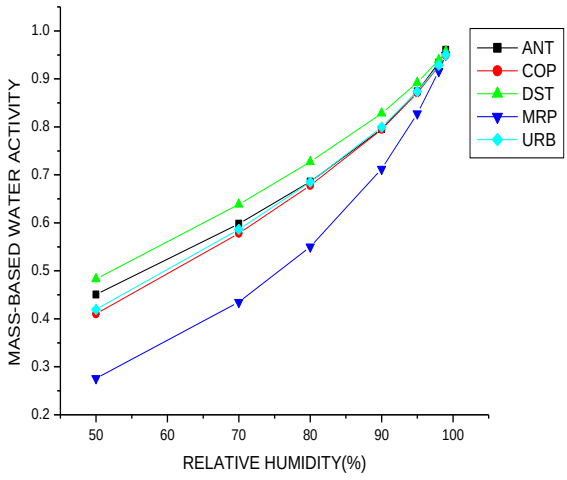

Figure 2.A graph of mass-based water activity against relative humidity

The ambient relative humidity (ARH) for atmospheric aerosols is compared with the impact of mass-based water activity using the data as shown in Figure 2. The aforementioned figure illustrates how water activity decreases ambient relative humidity in maritime and continental polluted areas, which are more sensitive to changes in relative humidity at higher levels (80–99). Conversely, it raises relative humidity in desert, urban, and Antarctic areas (Sa'adu 2020). With a steep curve at the deliquescence point, almost all aerosol particles are non-linear, and the smoothness of the curve indicates that the mixes are internally mixed.

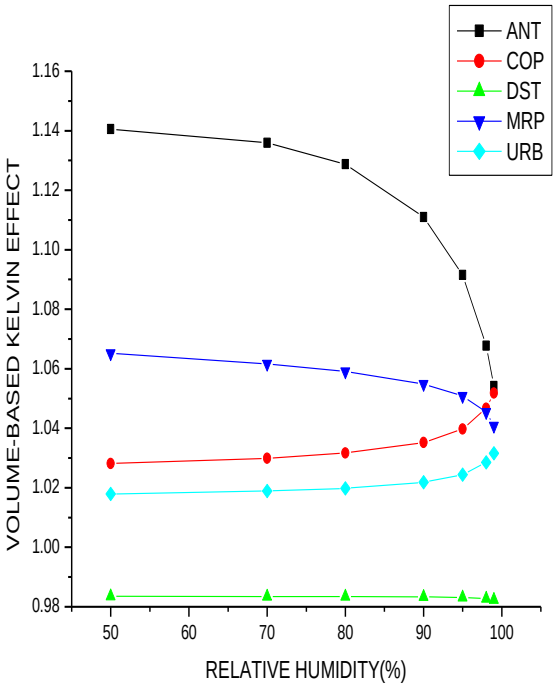

Figure 3: graph of volume-based Kelvin effect against relative humidity.

From figure 3, It is evident that the volume-based Kelvin effect in the desert is smaller than one and independent of RH, but the continental polluted and urban aerosols are both independent of RH but greater than one and occur at 90% RH. Meanwhile both the Antarctic and the maritime polluted exhibit similar behaviors, the Antarctic has more curve than the maritime polluted, which causes it to rise slightly with increasing relative humidity and become marginally more sensitive at higher relative humidity levels (90 to 99).

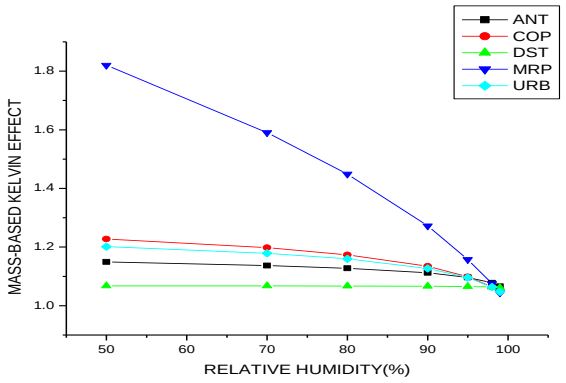

Figure 4: A graph of mass-based Kelvin effect against relative humidity

Figure 4 demonstrates how the mass-based Kelvin effect impacted on the desert aerosols which are linear, greater than one, and independent of RH. Subsequently, the characteristics of the Antarctic, continental polluted, desert, and urban aerosols are similar in their behavior, and they experience a minor increase in relative humidity (RH), with deliquescence occurring at RH levels between 90 and 99%. The remaining aerosols, which range in relative humidity from 70 to 99%, are non-linear and more sensitive at higher relative humidity levels.

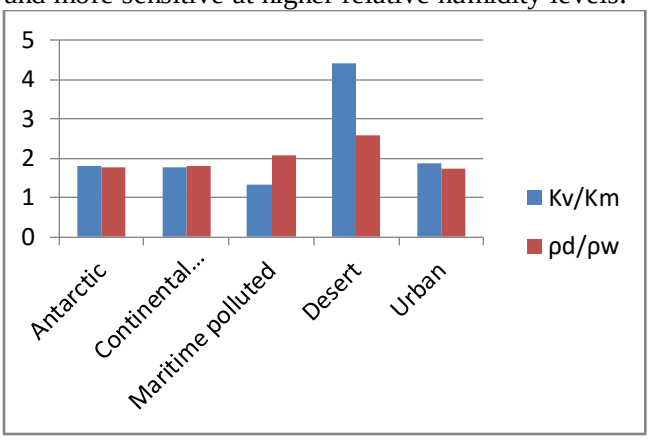

Figure 5: Relationship between the ratio of mass and volume-based hygroscopicity parameter and density.

Figure 5 shows that there are good agreements in Antarctic, continental polluted and urban while is non-linear for maritime polluted and desert by considering equation (14)

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

S/n	Aerosols Types	Aerosol Components	Number of Concentrations (cm^{-3})	R_{min} (µm)	R_{max} (µm)	Sigma (σ)	R_{mod} (µm)
1	Antarctic	Ssam	0.0470	0.005	20.00	2.0300	0.2090
		Mitr	0.0053	0.020	5.00	2.2000	0.5000
		Suso	42.9000	0.005	20.00	2.03	0.0695
2	Continental Polluted	Inso	0.6000	0.005	20.00	2.51	0.4710
		Waso	15,700.0000	0.005	20.00	2.24	0.0212
		Soot	34,300.0000	0.005	20.00	2.00	0.0118
3	Desert	Waso	2,000.0000	2.240	0.02	0.02	0.0050
		Minm	269.5000	1.950	0.07	0.07	0.0050
		Miam	30.5000	2.000	0.39	0.39	0.0050
		Micm	0.1420	2.150	1.90	1.90	0.0050
4	Maritime Polluted	Waso	3,800.0000	0.005	20.00	2.24	0.0212
		Soot	5,180.0000	0.005	20.00	2.00	0.0118
		Ssam	20.0000	0.005	20.00	2.03	0.2090
		Sscm	0.0032	0.005	60.00	2.03	1.7500
5	Urban	Waso	28,000.0000	0.0050	20.0000	2.2400	0.0212
		Inso	1.5000	0.0050	20.0000	2.5100	0.4710

		Soot	130,000.0000	0.0050	20.0000	2.0000	0.0118

The *sol* and *ns* show that the aerosols are soluble and insoluble respectively; the *inso* represents the water-insoluble part of aerosol particles and consists mostly of soil particles with a certain amount of organic material. The *waso* represents the water-soluble part of aerosol particles that originates from gas to particle conversion and consists of various kinds of sulfates, nitrates, and other, also organic, water-soluble substances. Thus it contains more than only the sulfate aerosol that is often used to describe anthropogenic aerosol. The soot component is used to represent absorbing black carbon. Carbon is not soluble in water and therefore the particles are assumed not to grow with increasing relative humidity. The *ssam* and *sscm* are Sea-salt accumulation and coarse mode particles that consist of various kind of salt contained in seawater. The *mitr* are Mineral transported, is used to describe desert dust that is transported over long distances with a reduced amount of large particles. Mineral aerosol particles are assumed not to enlarge with increasing relative humidity. The *suso*represent the sulfate component (75% H_2SO_4) which is used to describe the amount of sulfate found in the Antarctic aerosol. Mineral (nucleation mode) *MINM*, Mineral (accumulation mode) *MIAM*, and Mineral (coarse mode) *MICM*, are mineral aerosols or desert dusts that are produced in arid regions (Tijjani *et al.*, 2015).

Table 2: The Result of volume based hygroscopicity k_v

Aerosol models	k_v	p-value	R^2	Significance
Antarctic	1.51	1.20E-10	0.99	3.1E-09
Continental Polluted	0.45	8.10E-09	0.99	1.9E-07
Maritime polluted	2.03	6.31E-12	0.99	2.6E-10
Desert	0.02	6.27E-15	0.99	8.3E-13
Urban	0.38	1.25E-11	0.99	4.7E-10

Table 2 shows that; the data fitted the equation very well based on the correlation coefficient values. By looking at the results, it is evident that the Antarctic and maritime polluted regions have high k_v values because sea salt predominates the region, but the continental polluted regions have moderate k_v values and the desert has lower k_v values because there are enough minerals in the area (Sa'adu *et al.*, 2023).

Table 3: The Result of mass based hygroscopicity k_m

Aerosol models	k_m	p-value	R^2	Significance
Antarctic	0.82995	1.90E-10	0.99915	4.49E-09
Continental Polluted	0.25542	4.16E-10	0.9989	8.65E-09
Maritime polluted	1.52779	8.13E-10	0.99863	1.51E-08
Desert	0.0044	1.36E-10	0.99924	3.40E-09
Urban	0.20456	3.59E-10	0.99895	7.65E-09

Table 3 shows that the data fit the equation fairly well based on the correlation coefficient values. Tables 2 and 3 can be compared to observe that, as a result of the aerosol compositions, the values of k_v are higher than the values of k_m.

Table 4: Results of the ratio of densities and hygroscopicity parameters

Table 4 shows that, there is a good linear relationship between the ratio of densities and the ratio of hygroscopicities for Antarctic, continental polluted, and urban aerosols. However, it is non-linear formaritime polluted and desert aerosols, which is caused by the aerosol compositions (Sa'adu, 2020).

Aerosols model	Kv/Km	ρd/ρw
Antarctic	1.81357	1.76036
Continental polluted	1.77856	1.78945
Maritime polluted	1.32571	2.06046
Desert	4.41499	2.58684
Urban	1.85804	1.74264

Table 4 shows that, there is a good linear relationship between the ratio of densities and the ratio of hygroscopicities for Antarctic, continental polluted, and urban aerosols. However, it is non-linear formaritime polluted and desert aerosols, which is caused by the aerosol compositions (Sa'adu, 2020).

CONCLUSION

The volume-based hygroscopicity parameter (k_v) was found to be greater than the mass-based hygroscopicity parameter (k_m) for all aerosols, according to the obtained result. The data also fitted the equations very well, with a correlation coefficient > 90, significance, and p-values less than 0.05. We can determine that there is no linear relationship between mass and volume-based hygroscopicity parameters for desert and maritime polluted, and the nonlinearity depends on the composition of the aerosols. This is demonstrated by comparing the relationship between mass and volume-based hygroscopicity parameters and densities with figure 5. Therefore, the two models are good for atmospheric modeling and air quality observation.

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