



COMPARISON BETWEEN KELVIN RADII AND BULK HYGROSCOPICITY OF MASS AND VOLUME BASED HYGROSCOPICITY PARAMETER FOR ATMOSPHERIC AEROSOLS

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ABSTRACT

A mass based and volume based hygroscopicities models were applied to the data extracted from Optical Properties of Aerosols and Clouds (OPAC). The microphysical properties obtained were radii, density, refractive index, mass and volume of the atmospheric aerosols of continental average, continental clean, continental polluted, maritime tropical, maritime polluted, maritime clean, 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 were determined while the parameters A_v and B_v for volume based and A_m and B_m for mass based of the aerosols were determined using multiple regression analysis with SPSS 16.0 at each relative humidity. Although it was discovered that B_v is more dominant than A_v , the R^2 for all the models are greater than 90%. The significances are less than 0.05, $A_m > A_v$ and $B_m < B_v$, therefore the two models are good for atmospheric modeling.

Keywords: Hygroscopicity, volume based, mass based, atmospheric aerosols, OPAC.

INTRODUCTION

Hygroscopicity is the ability of aerosol particles to absorb water as a major property, which can change the particle size, shape, and optical properties (Ogren and Charlson, 1992; Tang, 1996), sometimes cause visibility deterioration (Liu et al., 2013), and thus control direct and indirect climate effects of the atmospheric aerosols. For a given size of the particles, the hygroscopic behaviors can also reflect information on their ageing process, mixing state (Maßling et al., 2003; Wang et al., 2017) and even the ability of the particles to be activated as cloud condensation nuclei, CCN (Kawana et al., 2014; Zhang et al., 2014, 2017).

Atmospheric aerosols can affect climate directly by scattering and absorbing incoming solar radiation, or indirectly by acting as cloud condensation nuclei (CCN), which form clouds and in turn can reflect light (Twomey, 1977).

The efficiency of particles as CCN also affects both aerosol particle and cloud droplet lifetimes. It is well recognized that these effects represent one of the largest uncertainties in assessing the changes in radiative forcing from pre-industrial times to the present (Solomon et al., 2007). As such, understanding the hygroscopic properties of aerosols and the processes that govern cloud droplet activation are important.

K"ohler theory has been used to predict the CCN-activity of inorganic compounds for many years (K"ohler, 1936). In the last decade, the focus has turned to the prediction of the CCNactivity of organic compounds in atmospheric particles. Laboratory studies have shown that the CCN- activity of organic, inorganic, or mixed aerosol systems can be predicted if the composition of the particles is well- characterized and their properties are known (Bilde and Svenningsson, 2004; Broekhuizen et al., 2004a; Raymond and Pandis, 2002, 2003). However, ambient aerosols are composed of numerous organic compounds that are difficult to identify and quantify (Jacobson et al., 2000; Saxena and Hildemann, 1996), therefore complicating the prediction of the CCN-activity of those ambient particles. The organic fraction comprises a significant fraction of the aerosol at many locations in the Northern Hemisphere (Zhang et al., 2007), highlighting the need to study organic aerosol hygroscopicity.

A mass-based hygroscopicity parameter km was introduced building on the single hygroscopicity parameter of Petters and Kreidenweis (2007), and developed a km-interaction model (KIM) to describe non-ideal solution behavior and concentration dependencies of single- and multi component systems. The KIM was demonstrated to capture and reproduce the characteristics of water uptake by pure reference substances as well as atmospheric aerosol samples under sub- and super saturated conditions (Mikhailov et al, 2011).

METHODOLOGY

The data extracted from Optical Properties of Aerosols and clouds are presented below where the *insoluble* component describes soil particles that also contain organic compounds, but do not experience hygroscopic growth while *water-soluble* component combines all aerosol particles that originate in gas to

particle conversion (including various kinds of sulfates, nitrates, and also organic substances). The *soot* component describes the absorbing black carbon. The *sea salt* components are given for the accumulation (100 nm< $D < 1 \mu m$) and the coarse ($D > 1 \mu m$) mode separately and both are dependent on RH. (Zieger et al,

2013).Water soluble components (WASO, consists of scattering aerosols), water insoluble (INSO), soot (SOOT) not soluble in water, *ssam* and *sscm* are Sea-salt accumulation and coarse modes particles that consist of the various kinds of salt contained in seawater.(Tijjani et al,2015)

Table1: the composition and some microphysical properties of atmospheric aerosols extracted from OPAC at 0RHs (Hess et al, 1998)

Model No.	Aerosols Model Types	Aerosols Components	Number of Conc. (cm ⁻³)	Rmin (µm)	Rmax (µm)	Sigma σ	Rmod (µm)
		Inso	0.4000	0.0050	20.0000	2.5100	0.4710
1	Continental Average	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
2	Continental Clean	Inso	0.1500	0.0050	20.0000	2.5100	0.4710
		Inso	0.6000	0.0050	20.0000	2.5100	0.4710
3	Continental Polluted	Waso	15,700.0000	0.0050	20.0000	2.2400	0.0212
		Soot	34,300.0000	0.0050	20.0000	2.0000	0.0118
		Waso	1,500.0000	0.0050	20.0000	2.2400	.0212
4	Maritime Clean	Ssam	20.0000	0.0050	20.0000	2.0300	0.2090
		Sscm	0.0032	0.0050	60.0000	2.0300	1.7500
		Waso	3,800.0000	0.0050	20.0000	2.2400	0.0212
5	Maritime Polluted	Soot	5,180.0000	0.0050	20.0000	2.0000	0.0118
5	Maritime i onuted	Ssam	20.0000	0.0050	20.0000	2.0300	0.2090
		Sscm	0.0032	0.0050	60.0000	2.0300	1.7500
		Waso	590.0000	0.0050	20.0000	2.2400	0.0212
6	Maritime Tropical	Ssam	10.0000	0.0050	20.0000	2.0300	0.2090
		Sscm	0.0013	0.0050	60.0000	2.0300	1.7500

Volume based Kelvin radii and bulk hygroscopicity factor

The kohler theory is given by

Where, a_w is water activity, σ_w is the surface tension, V_w is the molar volume R is the universal gas constant, T is the temperature and D is the diameter(Pruppacher and Klett, 2000; Seinfeld and Pandis, 2006).

According to petters and kreidenweis (2007), the hygroscopic growth of aerosol particle can be efficiently approximated by a simplified version of eq.(1), Since there is a good relation between density of water, mass and its molar volume. Eq (1) can be written as the so-called k_v -kohler equation:

Where aw is the volume based water activity and is define as

The aerosol's hygroscopicity growth factor g(s),(swietlicki et al.,2008;randles, et al.,2004) is define as:

Where S can be set for eight values, at 0%, 50%, 70%, 80%, 90%, 95%, 98% and 99% relative humidity RH substituting eqn (5) in to eqn (3) we get

$$\ln S = \frac{A}{r(S)} + \frac{B}{1 - (g(S))^3} - \dots - \dots - \dots - \dots - (6)$$

Where $r_K = \frac{2\sigma v_w}{RT} = A$ and $B = v \phi \frac{v_w}{v_s}$ are assumed to be constant, r(s) and r(s=0) are the radii of the mixture or the volume

equivalent radii of the wet and dry mixtures respectively, σ is the surface tension, v_w and v_s are the coefficient. The product of v and ϕ is equivalent to the so called van't Hoff factor (Tijjani et al,2015).

Where a characteristic length for the effect of surface tension (or Kelvin radius) $r_K = \frac{2\sigma \vartheta_W}{RT} = A$. The Kelvin effect is small for large droplet radii and It becomes very large for radii r(s) smaller than the Kelvin radius r_k

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

But generally atmospheric aerosols usually comprised mixtures of soluble components, therefore the information on the hygroscopicity modes was merged into an "over-all" or "bulk" or "effective" hygroscopic growth factor of the mixture, $g_{eff}(s)$ representative for the entire aerosols particle population as:

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

Where the summation is performed over all compounds present in the particles and X_k represent their respective volume fractions, using the Zdanovskii-Stokes-Robisnson relation (ZSR relation; Sjogren et al., 2007;Stokes and Robinson, 1966;Meyer et al., 2009;Stock et al., 2011).

Equations (6) can be written in terms of volume based as

The Kelvin effect and water activity are the major parameters that made hygroscopic growth of aerosols to be size and composition dependent and are given by

Where A_v is kelvin radii and B_v is bulk hygroscopicity of volume based hygroscopicity. and can be determine using regression analysis with SPSS 16.0.

Mass Based Hygroscopicity kelvin radii and bulk hygroscopicity factor

In analogy to the volume based hygroscopicity we define a mass based hygroscopicity as

Where A_m is the kelvin radii and B_m is the bulk hygroscopicity of mass based. Using multiple regression analysis with SPSS 16.0 for windows, the constants A_m and B_m were determined.

The first term on the right hand side of equation (14) can be written as

This implies that

The second term on the right hand side of equation (14) is

$$a_w = exp\left(\frac{B_1}{1-g_m}\right) - - - - - - - - - - - - - - - - - (16b)$$

By defining the mass growth factor G_m (Mikhailov et al, 2011) as

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

Results and Discussion

From the result below, it can be seen that, based on the values of \mathbb{R}^2 , that the data fitted the equation very excellent. By analysing the table, it can be seen that B is more dominant than A for maritime clean, maritime polluted and maritime tropical due to their p-values and from the values of Kelvin radii, it observed that, the values of Kelvin radii are less than the values of bulk hygroscopicity and Kelvin radii have higher values in maritime clean and maritime tropical than maritime polluted, continental average, continental clean and continental polluted.

		Kelvin					
s/n	Aerosols M	radii(A _v)	p-value	bulk hygroscopicity (Bv)	p-value	\mathbb{R}^2	significance
	Continental						
1	average	0.00623	0.03362	0.4005	3.09E-07	0.99729	4.72E-06
	continental						
2	clean	0.00655	0.03504	0.41251	4.02E-07	0.99698	5.87E-06
	continental						
3	polluted	0.00618	0.03703	0.43287	6.07E-07	0.99642	8.22E-06
4	Maritime Clean	0.05123	0.08021	2.14803	3.37E-05	0.98928	7.39E-05
	Maritime						
5	Polluted	0.04812	0.07925	1.99641	2.81E-05	0.99016	6.23E-05
	maritime						
6	tropical	0.0499	0.07798	2.17642	3.55E-05	0.9892	7.51E-05

Table 2: The results of Kelvin radii and bulk hygroscopicity for volume based using equation(11)

From the result in table below, it can be seen that maritime clean, maritime polluted and maritime tropical has high values of kelvin radii due to the dominant of sea salt.

s/n	Aerosols model	Kelvin radii(A _m)	p-values	Bulk hygroscopicity(Bm)	p-values	R ²	Significance
1	continental average	0.6556	2.60E-05	0.1302	1.14E-07	0.9991	5.01E-07
2	continental clean	0.6696	2.68E-05	0.1327	1.25E-07	0.9991	5.41E-07
3	continental polluted	0.7615	3.36E-05	0.1489	2.32E-07	0.9988	8.87E-07
4	maritime clean	1.3054	0.0001	0.2474	4.06E-06	0.9963	8.86E-06
5	maritime polluted	3.2413	0.0014	0.7551	0.0002	0.9936	0.0003
6	maritime tropical	3.3776	0.0005	0.8058	6.48E-05	0.9910	5.20E-05

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1 able 5: the results o	i keivin radii	and bulk hygro	scopicity for mas	s based using	eqn(14)

Table 4 shows that the mass growth factor increases as the relative humidity increases but there is much increments for maritime clean, maritime polluted and maritime tropical at the liquescence point (98% -99%).

Aerosols model	RH (0.5)	RH(0.7)	RH(0.8)	RH(0.9)	RH(0.95)	RH(0.98)	RH(0.99)
continental average	1.237	1.385	1.543	1.921	2.525	3.776	5.032
continental clean	1.242	1.393	1.555	1.940	2.558	3.835	5.118
continental polluted	1.276	1.448	1.632	2.071	2.775	4.230	5.691
maritime tropical	2.288	2.997	3.774	5.921	9.853	20.291	34.461
maritime polluted	2.145	2.769	3.464	5.338	8.788	17.742	29.857
maritime clean	2.272	2.959	3.728	5.811	9.685	19.836	33.728

Table 4.The mass growth factor G_{m using} eqn (17)

Table 5 shows that, the effective hygroscopic growth of the aerosols don't depend on the initial values of relative humidity. By comparing table 1 with the effective hygroscopic growth of the aerosols at 0%, it can be observe that they depend on the nature of the compositions.

	aerosols								
s/n	model	RH(0)	RH(0.5)	RH(0.7)	RH(0.8)	RH(0.9)	RH(0.95)	RH(0.98)	RH(0.99)
	continental								
1	average	1	1.164	1.262	1.358	1.559	1.809	2.188	2.471
	continental								
2	clean	1	1.169	1.268	1.364	1.565	1.815	2.193	2.475
	continental								
3	polluted	1	1.176	1.277	1.374	1.575	1.824	2.200	2.481
	maritime								
4	clean	1	1.589	1.788	1.967	2.351	2.864	3.797	4.722
	Maritime								
5	Polluted	1	1.560	1.755	1.933	2.312	2.819	3.744	4.665
	Maritime								
6	Tropical	1	1.593	1.792	1.972	2.357	2.871	3.804	4.730

 Table 5.the hygroscopic growth factor geff using equation (9)

From table 6 below, it can be seen that the effective radii of the aerosols decreases as the relative humidity increase for continental average, continental clean and continental polluted while it increases as relative humidity increases for maritime clean, maritime polluted and maritime tropical due to their compositions.

	aerosols								
s/n	model	RH(0)	RH(0.5)	RH(0.7)	RH(0.8)	RH(0.9)	RH(0.95)	RH(0.98)	RH(0.99)
	continental								
1	average	0.362	0.320	0.303	0.287	0.260	0.232	0.198	0.179
	continental								
2	clean	0.368	0.325	0.306	0.290	0.262	0.233	0.200	0.180
	continental								
3	polluted	0.329	0.288	0.271	0.256	0.231	0.205	0.175	0.158
4	maritime clean	0.593	0.788	0.830	0.862	0.924	0.991	1.103	1.220
	maritime								
5	polluted	0.552	0.762	0.804	0.838	0.901	0.970	1.084	1.204
	maritime								
6	tropical	0.561	0.744	0.783	0.816	0.875	0.944	1.061	1.187

Table 6. The effective radius r_{eff} of the aerosols using equation (10)



Figure 1: a graph of bulk hygroscopicity (B) and kelvin radii (A) for volume based

From the graph above, it can be seen that bulk hygroscopicity and kelvin radii shows good linear relationship in continental average, continental clean and continental polluted while continental clean, continental average and continental polluted lies between 0-0.5 and maritime clean, maritime polluted and maritime tropical also lies between 2-2.5.



Figure 2: A graph of kelvin radii (A) and bulk hygroscopicity (B) for mass based

According to the graph above, it shows that; there are good agreement between continental clean, continental average and continental polluted while maritime polluted and maritime tropical has deviated. By observing maritime tropical, it can be seen that it rises very high in the graph than the remaining aerosols component and also continental clean and continental average lies at the same level.

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Figure 3: A graph of volume based water activity against relative humidity using eqn(13)

From figure 3, the relative humidity is used to analyze how the water activity of the volume based affects the ambient RH for the aerosols. It is clearly seen that, the water activity lowers the ambient relative humidity of Maritime pollution and Maritime tropical while the Continental pollution, Continental clean and Continental average raises. The maritime clean show linearity except at one point.



Figure 4: A graph of volume based kelvin effect against Relative Humidity using eqn (12)

From figure 4, it can be seen that volume based Kelvin effect on Maritime clean is more sensitive to RH although the sensitivity increase with increase in RH in a non-linear form and is less than, while Continental polluted, continental clean are also independent of RH but greater than one. Secondly, The Maritime tropical and Maritime polluted have the same type of behaviour, that is why they increase slightly with the increase in RH and are slightly more sensitive at higher RHs (90% - 99%).



Figure 5: A graph of mass based water activity against relative humidity usingeqn(16b)

The ambient relative humidity is used to compare how the mass based affect the water activity of atmospheric aerosols. It can also see from the above graph show that the water activity lowers the ambient relative humidity for maritime polluted, maritime clean and maritime tropical. The continental clean, continental average rises.



Figure 6: A graph of mass based kelvin effect against relative humidity using eqn (15b)

According to the graph above it can seen that, the entire aerosols component shows curve and maritime tropical and maritime clean and maritime pollued rises while continental clean and continental average and continental polluted has lower the relative humidity.



Figure 7: A graph of mass grow factor against relative humidity using eqn (17)

Figure 7 shows that, the mass growth has less effect at high relative humidity and shows steep curve at deliquescence point (90% to 99%) of the all aerosols. They are more sensitive at deliquescence point; however there is slight increase of the mass growth of the particle at low relative humidity, meaning the particle at this region are more hygroscopic in nature.

CONCLUSION

The result obtained shows that bulk hygroscopicity (B) is more dominant than Kelvin radii (A) for maritime clean, maritime polluted and maritime tropical due to their p-value for volume based. From table 4,it shows that the mass growth factor (G_m) increases as the relative humidity increases but there are much increment for maritime clean and maritime polluted at deliquescence point (98% -99%). The effective radii (r_{eff}) of the aerosols decrease as the relative humidity increases for maritime and also there are good linear relationship between Kelvin radii and bulk hygroscopicity for continental. Finally it discovered that, Kelvin radii for mass based is greater than that of volume based ($A_m > A_v$). Meanwhile the bulk hygroscopicity for mass based is less than that of volume based ($B_m < B_v$).

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