

## COMPARATIVE ANALYSIS OF MASS AND VOLUME BASED HYGROSCOPICITIES PARAMETER INTERACTION MODELS FOR ATMOSPHERIC AEROSOLS

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### *Abstract*

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*A mass based hygroscopicity parameter ( $k_m$ ) and volume based hygroscopicity parameter ( $k_v$ ) models were applied to the data extracted from OPAC (Optical Properties of Aerosols and Clouds). The microphysical properties obtained were radii of the atmospheric aerosols of Antarctic, arctic, continental average, continental clean, continental polluted maritime tropical, maritime polluted, maritime clean, Sahara and urban at eight different relative humidity of 0%, 50%, 70%, 80%, 90%, 95%, 98% and 99%. The parameters  $k_m$  and  $k_v$  of the aerosols were determined using multiple regression analysis with SPSS at each relative humidity. Although it was discovered that  $k_v$  is greater than  $k_m$ , the  $R^2$  for all the models are greater than 90%, The  $p$ -values and significances are less than 0.05. Therefore it can be concluded that any of the two parameters can be used for atmospheric modeling.*

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**Keywords:** OPAC, RHs, SPSS, mass based hygroscopicity, volume based hygroscopicity.

### **1. Introduction**

Atmospheric aerosols are suspensions of tiny particles in air that originate from a wide variety of natural and anthropogenic sources. Depending on their sources, aerosols appear in different sizes and have different physical and chemical properties. Aerosols play a significant role in the Earth's climate system and are a major contributor to air pollution. Despite considerable advances in recent decades, significant gaps still remain in our knowledge of aerosols and their impacts on our planet and human health. Atmospheric aerosols are also defined as liquid or solid particles suspended in a gas [1] are tiny and usually invisible to our eyes. Nevertheless, they have an immense impact on our health and on our global climate. Aerosols scatter and absorb solar radiation, and, by doing so, they directly affect the Earth's radiation budget [2,3,4]. In addition, anthropogenic aerosol particles modify cloud properties, causing brighter clouds with longer lifetimes and changed precipitation behavior [5,6,7]. The net effect of anthropogenic aerosols on the Earth's climate is cooling, in contrast to greenhouse gases, which have a warming effect. However, the Intergovernmental Panel on Climate Change [8] concludes that the high uncertainty in the net radiative forcing of all main climate agents is mainly dominated by the large uncertainty in the aerosol radiative forcing. This is mainly caused by the high temporal, spatial, and compositional variability of the aerosol and the poorly understood and quantified aerosol effects. Since aerosol particles can take up water, they can change in size and chemical composition depending on the ambient relative humidity (RH)

The equation that is often used to describe both hygroscopic growth of aerosol particles and their activation to cloud droplets is the Kohler equation. The Kelvin effect and water activity are the major parameters that made hygroscopic growth of aerosols to be size and composition dependent. The growth of aqueous droplets in ambient RH is commonly described by Kohler theory [9]

Kohler theory is the main tool to describe the hygroscopic growth of particles as a function of relative humidity [10,1] and various types of Kohler models have been developed and applied for the analysis of results as well as in numerical models of the atmosphere and climate [11,12,13,14,15,16,17,18,19,20,21,22].

The aim of hygroscopicity measurements and Kohler model studies is to compare the critical super saturation of CCN activation to the hygroscopic growth factors [23,24,25,26,27,28,29,30,31,32,33,34,35,36]. Petters and Kreidenweis (2007) proposed a single-parameter Kohler model where the hygroscopicity parameter  $k$  provides a volume-based measurement of aerosol water uptake characteristics and CCN activity. The practical applicability of volume based hygroscopicity models, however, is often limited by deviations from volume additivity and by the effects of capillary condensation and restructuring of porous and irregularly shaped particles [18,37,38,39,40,41,42].

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2. Methodology

Table1: the composition and some microphysical properties of atmospheric aerosols extracted from OPAC at 0 RHs [43]

Model Number	Aerosols Model Types	Aerosols Components	Number Concentrations (cm <sup>-3</sup> )	Rmin (µm)	Rmax (µm)	Sigma σ	Rmod (µm)
1	Antarctic	Ssam	0.0470	0.0050	20.0000	2.0300	0.2090
		Mitr	0.0053	0.0200	5.0000	2.2000	0.5000
		Suso	42.9000	0.0050	20.0000	2.0300	0.0695
2	Arctic	Inso	0.0100	0.0050	20.0000	2.5100	0.4710
		Waso	1,300.0000	0.0050	20.0000	2.2400	0.0212
		Soot	5,300.0000	0.0050	20.0000	2.0000	0.0118
		Ssam	1.9000	0.0050	20.0000	2.0300	0.2090
		Waso	15,700.0000	0.0050	20.0000	2.2400	0.0212
		Soot	34,300.0000	0.0050	20.0000	2.0000	0.0118
3	Desert	Waso	2,000.0000	2.2400	0.0212	0.0212	0.0050
		Minm	269.5000	1.9500	0.0700	0.0700	0.0050
		Miam	30.5000	2.0000	0.3900	0.3900	0.0050
		Micm	0.1420	2.1500	1.9000	1.9000	0.0050
4	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 *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 modes particles that consist of the various kinds 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* is sulfate component (75% H<sub>2</sub>SO<sub>4</sub>) 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. They consist of mixtures of quartz and clay minerals and are modeled with these three modes to allow considering increasing relative amount of large particles for increasing turbidity [9].

2.1 VOLUME BASED HYGROSCOPICITY PARAMETER K<sub>v</sub>

The Kohler theory that states the water activity (a<sub>w</sub>), the partial molar volume of water (V<sub>w</sub>), the surface tension (σ<sub>w</sub>), and the diameter (D) of a spherical aqueous droplet under equilibrium conditions to the water vapor saturation of the surrounding gas phase (S<sub>w</sub> = RH/100%) [1,10]

$$s_w = a_w \exp\left(\frac{4\sigma_w V_w}{RTD}\right) \text{----- (1)}$$

Where R is the universal gas constant and T is the temperature. According to [27], the hygroscopic growth of aerosol particles can be efficiently approximated by a simplified version of Eq. (3.1), the so-called κ<sub>v</sub>- Kohler equation:

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

$$= \left[\frac{\kappa_v}{\sigma_w^3} + 1\right]^{-1} \exp\left(\frac{4\sigma_w M_w}{RT\rho_w g_d d}\right) \text{----- (3)}$$

Here M<sub>w</sub>, σ<sub>w</sub> and ρ<sub>w</sub> are the molar mass, surface tension, and density of pure water. The aerosol's hygroscopicity growth factor g(S), [44, 45] is defined as:

$$g(S) = \frac{r(S)}{r(S=0)} \text{----- (4)}$$

Where S is taken for eight values 0%, 50%,70%,80%,90%,95%,98% and 99% RH Substituting equation (4) into equation (3) we obtained

$$\ln S = \frac{A}{r(S)} + \frac{B}{1-(g(S))^3} \text{----- (5)}$$

Where r<sub>K</sub> =  $\frac{2\sigma v_w}{RT}$  = 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<sub>w</sub> and v<sub>s</sub> are the coefficient. The product of v and φ is equivalent to the so called van't Hoff factor

The first term on the right hand side of equation (5) is

$$\ln K_e = \frac{2\sigma\phi_w}{RT r(S)} = \frac{A}{r(S)} = g_\sigma(r(S)) \text{----- (6)}$$

Where a characteristic length for the effect of surface tension (or Kelvin radius)  $r_k = \frac{2\sigma\theta_w}{RT} = A$ . 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 (5) is

$$\ln a_w = \frac{B}{1-(g(S))^3} \text{---(7)}$$

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:

$$g_{eff}(S) = \left(\sum_k x_k g_k^3(S)\right)^{1/3} \text{---(8)}$$

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

$$r_{eff}(S) = \left(\sum_k x_k r_k^3\right)^{1/3} \text{---(9)}$$

Where the summation is performed over all compounds present in the particles and  $X_k$  represent their respective volume fractions, using [40, 46, 47, 48]. For multi-components equation (5) can be written to represent the property of the bulk components using equations (8) and (9) as:

$$\ln S = \frac{A}{r_{eff}(S)} + \frac{B}{1-g_{eff}^3(s)} \text{---(10)}$$

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

The hygroscopicity parameter  $\kappa_v$  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:

$$\frac{1}{a_w} = 1 + \kappa_v \frac{V_d}{V_w} \text{---(11)}$$

Or

$$\frac{1}{a_w} = \left[\frac{\kappa_v}{g_{eff}^3 - 1} + 1\right] \text{---(12)}$$

Equations (2)–(11) are commonly used to determine effective hygroscopicity parameters from hygroscopic growth or CCN activation measurements using hygroscopicity tandem differential mobility analyzer (HTDMA) and CCN counter techniques [22,26,28,29,34,38,49,50]. The limitations of these assumptions for HTDMA and CCN experiments have been outlined and discussed in earlier studies [18,37,38,39,42,51,52].

**2.2 MASS-BASED HYGROSCOPICITY PARAMETER  $\kappa_m$**

In analogy to the volume based hygroscopicity parameter (Eq. 11), we define a mass based hygroscopicity parameter  $\kappa_m$ :

$$\frac{1}{a_w} = 1 + \kappa_m \frac{m_d}{m_w} \text{---(13)}$$

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).

Since mass  $m$  is proportional to  $r^3$ , this implies that  $g_m$  is proportional to  $g_{eff}^3$ , therefore equation (10) can be written as

$$\ln S = \frac{A_1}{(g_m)^{1/3}} + \frac{B_1}{1-g_m} \text{---(14)}$$

Using multiple regression analysis with SPSS 16.0 for windows, the constants  $A_1$  and  $B_1$  were determined.

The first term on the right hand side of equation (14) can be replaced by equation (6) as

$$\ln k_e = \frac{2\sigma\theta_w}{RT(g_m)^{1/3}} = \frac{A_1}{(g_m)^{1/3}} \text{---(15a)}$$

This implies that

$$k_e = \exp\left(\frac{A_1}{(g_m)^{1/3}}\right) \text{---(15b)}$$

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

$$\ln a_w = \frac{B_1}{1-g_m} \text{---(16a)}$$

This implies

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

Under the assumption of volume additivity, Eq. (11) and Eq. (13) can be combined to

$$\kappa_m = \kappa_v \frac{\rho_w}{\rho_d} \text{ Or } \frac{\kappa_v}{\kappa_m} = \frac{\rho_d}{\rho_w} \text{---(17)}$$

where  $\rho_d$  is the density of the dry particles material and  $\rho_w$  is the density of water and assumed to be 1.0 g/cm<sup>3</sup>. In this case, the knowledge of  $\rho_d$  enables conversion of  $\kappa_v$  into  $\kappa_m$  and vice versa. But since  $\kappa_m$  and  $\kappa_v$  represents the overall effective hygroscopicity of the aerosols, so  $\rho_d$  can be expressed as

$$\rho_d = \sum f_i \rho_{di} \text{---(18)}$$

Where  $f_i$  represent the volume mix ratio of  $i$ th term and  $\rho_{di}$  is the density of the  $i$ th dry aerosols.

By defining the mass growth factor  $G_m$  as

$$G_m = \frac{m_w + m_d}{m_d} \text{---(19)}$$

And combining Eq. (19) and Eq. (13) we obtain

$$\frac{1}{a_w} = \left(\frac{\kappa_m}{G_m - 1} + 1\right) \text{---(20)}$$

RESULT AND DISCURSION

Table .2 Result of volume based hygroscopicity parameter using equation (12)

Aerosol models	$k_v$	p-value	$R^2$	Significance
Antarctic	1.5052	1.20E-10	0.9993	3.06E-09
Arctic	1.60274	4.05E-12	0.99977	1.82E-10
Sahara	0.01943	6.27E-15	0.99997	8.29E-13
Urban	0.38008	1.25E-11	0.99966	4.66E-10

From table 2, it can be seen that, from the values of  $R^2$ , that the data fitted the equation very excellent. By observing the result it can be seen that, volume based hygroscopicity parameter  $k_v$  is higher for arctic aerosols and is less for Saharan due to sufficient mineral at the region. The p-values and significance are all less than 0.05 which shows that the model is good for the entire aerosols.

Table 3: Result of mass based hygroscopicity using equation (20)

Aerosol models	$k_m$	p-value	$R^2$	Significance
Antarctic	0.82995	1.90E-10	0.99915	4.49E-09
Arctic	0.91265	4.87E-10	0.99884	9.86E-09
Sahara	0.0044	1.36E-10	0.99924	3.40E-09
Urban	0.20456	3.59E-10	0.99895	7.65E-09

Table 3 indicates that, from the values of  $R^2$ , that the data fitted the equation very well. The value of arctic is also greater in mass based and lower in Saharan. By comparing table 2 and 3 we can see that, the values of  $k_v$  are greater than the values of  $k_m$  from regression analysis.

Table 4: Result of mass growth factor using equation (19)

RH (%)	50	70	80	90	95	98	99
ANTARCTIC	2.0094	2.5660	3.1330	4.5119	6.9559	12.7293	20.9224
ARCTIC	1.7860	2.2298	2.7186	4.0139	6.3135	12.0880	19.6138
SAHARA	1.0059	1.0095	1.0134	1.0226	1.0374	1.0685	1.0991
URBAN	1.2269	1.3690	1.5204	1.8813	2.4607	3.6579	4.8613

From table 4, it can be observed that, the mass growth factor are increasing as relative humidity increase but there are much increment for Antarctic and arctic at 98% and 99% relative humidity.

Table 5: Results of ratio of densities and hygroscopicities using equation (17)

Aerosols model	$K_v/K_m$	$\rho_d/\rho_w$
Antarctic	1.81357	1.76036
Arctic	1.73456	1.82250
Sahara	4.41499	2.58684
Urban	1.85804	1.74264

From table 5, it can be seen that, there are good linear relationship between the ratio of densities and the ratio of hygroscopicities parameters for Antarctic, arctic and urban aerosols and are non linear for Saharan aerosols, their non-linearity is due to the compositions of the aerosols.

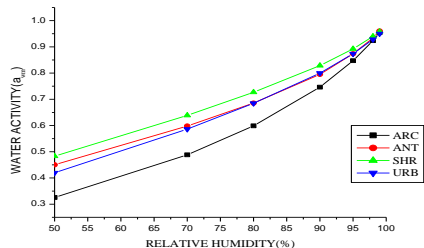


Fig 1.A graph of mass based water activity against relative humidity using eq(16b).

From figure 1, the ambient RH is used to compare how the mass based water activity affect the ambient RH for atmospheric aerosols. From the figure above, it can be seen that the water activity raises the ambient relative humidity of Antarctic, Saharan, Arctic, and Urban and increases slightly with the increase in relative humidity which are more sensitive at higher RHs (80% - 99%) Almost all the aerosols particles are non-linear with steep curve at deliquescence point and the smoothness of the curve shows that the mixtures are internally mixed.

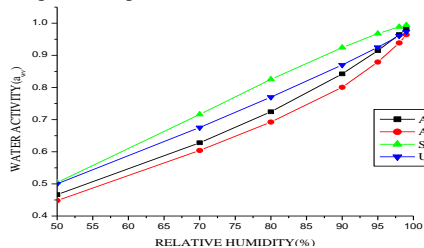


Fig 2.A graph of volume based water activity against relative humidity using eq(7)

From figure 2, the ambient RH 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 Antarctic, Arctic, and Urban which are in non linear form. Lastly, the volume based water activity of Saharan aerosols is raised.

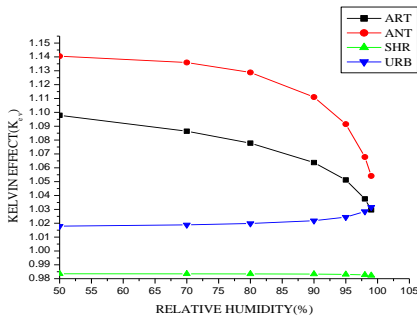


Fig 3.A graph of volume based Kelvin effect against relative humidity using eq (6)

From figure 3, It can be seen that volume based Kelvin effect on Saharan is independent of RH (meaning it is constant) and is less than unity, while Urban is increasing as RH increases and is more sensitive at (95% - 99%). but is greater than one. Secondly, Arctic and Antarctic 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%).

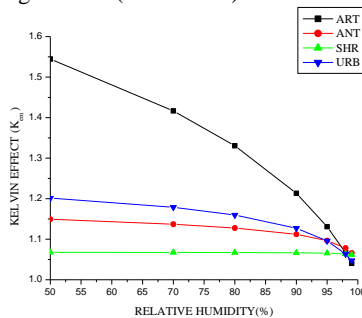


Fig 4.A graph of mass based Kelvin effect against relative humidity using eq(15b)

Figure 4 shows that, the mass based Kelvin effect on Saharan aerosols is linear (independent of RH) and is greater than one. Secondly, the Antarctic, Urban, , have the same character ,in that they increase slightly with the increase in RH with deliquescence observed at relative humidity as from 90% to 99% and the arctic aerosols is non-linear.

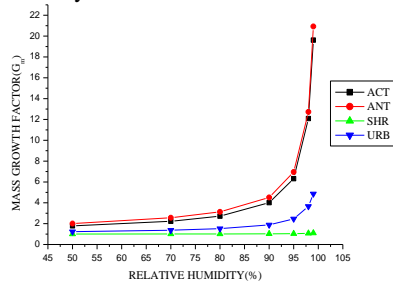


Fig 5.A graph of mass growth factor against relative humidity using eq(19)

Figure 5, 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 except that of Saharan which is independent of relative humidity(that is it is constant). 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.

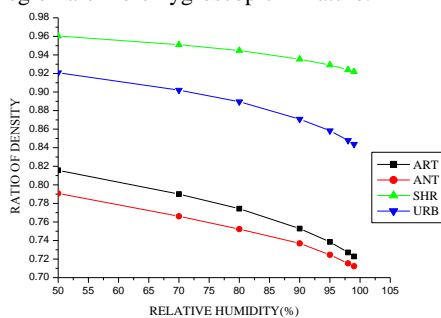


Fig 6.A graph of ratio of density against relative humidity using eq(17)

Figure 6,shows that all the aerosols are non linear, the Saharan increase more as the relative humidity increases while the remaining aerosols increase slightly as the relative humidity increases and are more sensitive at 90%-99%

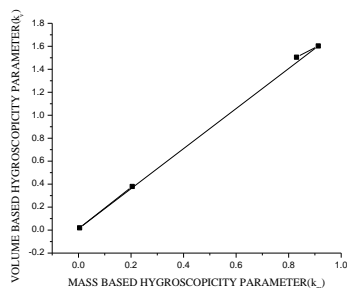


Fig 7.A graph of volume based parameter ( $k_v$ ) against mass based parameter ( $k_m$ ) using eq(17)

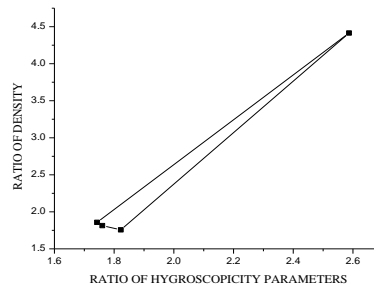


Fig 8.A graph of ratio of density against ratio of hygroscopicity parameters using eq(17)

## CONCLUSION

From the result obtained, it was discovered that, the values of  $k_v$  are greater than the values of  $k_m$  for the entire. From table 4, it can be observed that the mass growth factor are increasing as relative humidity increase but there are much increment for Antarctic and arctic at 98% and 99% relative humidity. By comparing the relationship between densities and hygroscopicities with our graph of fig 8, we can conclude that; there is no linear relationship between mass based and volume based hygroscopicity for Sahara, and the non linearity depends on the aerosols compositions. The regression analysis result shows that, p-values and significance are less than 0.05; R-square are greater than 90% for all the models which means they can be use for atmospheric modeling.

## REFERENCES

- [1] Seinfeld, J. H. and Pandis, S. N.: (2006) *Atmospheric Chemistry and Physics: From Air Pollution to Climate Change*, John Wiley and Sons, Inc., New York.
- [2] Trenberth, K., Fasullo, J., and Kiehl, J.: (2009) *Earth's global energy budget*, B. Amer. Meteor. Soc., 90, 311–323.
- [3] Schwartz, S. (1996) *The Whitehouse Effect—Shortwave radiative forcing of climate by anthropogenic aerosols: An overview*, J. Aerosol Sci., 27, 359–382, 1996.
- [4] Charson R.J., Langner J., Rodhe H., Leovy C.B., Warren S.G. (1991), *Perturbation of the northern hemisphere radiative balance by back scattering from anthropogenic sulphate aerosols*, Tellus B/volume 43, issue 4, doi.org/10.1034/j.1600-0889.
- [5] Lohmann, U. and Leck, C.: (2005) *Importance of submicron surface-active organic aerosols for pristine Arctic clouds*, Tellus B, 57, 261–268.
- [6] Ramaswamy, V., Boucher, O., Haigh, J., Hauglustaine, D., and Haywood, J.: (2001) *Radiative Forcing of Climate Change*, Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA.
- [7] Twomey, S.: (1997) *Atmospheric Aerosols*, Developments in Atmospheric Science, Elsevier, New York, USA.
- [8] IPCC: Climate Change (2007): *The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, Cambridge University Press, 2007.
- [9] Tijjani B.I., Uba S. Koki F.S., Galadanci G.S.M., Nura A.M., Adamu I.D., Saleh M. and Abubakar A.I (2015), *The effect of Kelvin effect on the equilibrium effective radii and hygroscopic growth of atmospheric aerosols*. Journal of Natural Sciences Research. 5,96-111.
- [10] Pruppacher, H. R. and Klett, J. D.: (2000) *Microphysics of clouds and pre-cipitation*, Kluwer Academic Publishers, Dordrecht.
- [11] Junge, C. and McLaren, E.: (1971) *Relationship of cloud nuclei spectra to aerosol size distribution and composition*, J. Atmos. Sci., 28, 382–390.
- [12] Fitzgerald, J. W.: (1973) *Dependence of supersaturation spectrum of CCN on aerosol size distribution and composition*, J. Atmos. Sci., 30, 628–634.
- [13] Shulman, M. L., Jacobson, M. C., Carlson, R. J., Synovec, R. E., and Young, T. E.: (1996) *Dissolution behavior and surface tension effects of organic compounds in nucleating cloud droplets*, Geophys. Res. Lett., 23, 277–280, doi:10.1029/95GL03810.
- [14] Kulmala, M., Laaksonen, A., Charlson, R. J., and Korhonen, P.: (1997) *Clouds without supersaturation*, Nature, 388, 336–337.
- [15] Laaksonen, A., Korhonen, P., Kulmala, M., and Charlson, R. J. (1998) *Modification of the Köhler equation to include soluble trace gases and slightly soluble substances*, J. Atmos. Sci., 55, 853–862.
- [16] Raymond, T. M. and Pandis, S. N. (2003): *Formation of cloud droplets by multicomponent organic particles*, J. Geophys. Res., 108, 4469, doi:10.1029/2003JD003503.
- [17] Bilde, M. and Svenningsson, B. (2004), *CCN activation of slightly soluble organics: the importance of small amounts of inorganic salt and particle phase*, Tellus B, 56, 128–134.
- [18] Mikhailov, E., Vlasenko, S., Niessner, R., and Pöschl, U.: (2004) *Interaction of aerosol particles composed of protein and salt with water vapour: hygroscopic growth and microstructural rearrangement*, Atmos. Chem. Phys., 4, 323–350, doi:10.5194/acp-4-323-2004.
- [19] Huff Hartz, K. E., Rosenørn, T., Ferchak, S. R., Raymond, T. M., Bilde, M., Donahue, N. M., and Pandis, S. N. (2005): *Cloud condensation nuclei activation of monoterpane and sesquiterpene secondary organic aerosol*, J. Geophys. Res., 110, D14208, doi:10.1029/2004JD005754.

- [20] McFiggans, G., Artaxo, P., Baltensperger, U., Coe, H., Facchini, M. C., Feingold, G., Fuzzi, S., Gysel, M., Laaksonen, A., Lohmann, U., Mentel, T. F., Murphy, D. M., O'Dowd, C. D., Snider, J. R., and Weingartner, E.:(2006) *The effect of physical and chemical aerosol properties on warm cloud droplet activation*, Atmos. Chem. Phys., 6, 2593–2649, doi:10.5194/acp-6-2593-2006.
- [21] Svenningsson, B., Rissler, J., Swietlicki, E., Mircea, M., Bilde, M., Facchini, M. C., Decesari, S., Fuzzi, S., Zhou, J., Mønster, J., and Rosenørn, T.:(2006) *Hygroscopic growth and critical supersaturations for mixed aerosol particles of inorganic and organic compounds of atmospheric relevance*, Atmos. Chem. Phys., 6, 1937–1952, doi:10.5194/acp-6-1937-2006.
- [22] Rose, D., Nowak, A., Achtert, P., Wiedensohler, A., Hu, M., Shao, M., Zhang, Y., Andreae, M., and Pöschl, U.(2010): *Cloud condensation nuclei in polluted air and biomass burning smoke near the mega-city Guangzhou, China- Part I: Size-resolved measurements and implications for the modeling of aerosol particle hygroscopicity and CCN activity*, Atmos. Chem. Phys., 10, 3365-3383, 2010.
- [23] Rissler, J., Swietlicki, E., Zhou, J., Roberts, G., Andreae, M. O., Gatti, L. V., and Artaxo, P (2004).: *Physical properties of the sub- micrometer aerosol over the Amazon rain forest during the wet- to-dry season transition – comparison of modeled and measured CCN concentrations*, Atmos. Chem. Phys., 4, 2119–2143, doi:10.5194/acp-4-2119-2004.
- [24] Kreidenweis, S. M., Koehler, K., DeMott, P. J., Prenni, A. J., Carrico, C., and Ervens, B.:(2005) *Water activity and activation diameters from hygroscopicity data – Part I: Theory and application to inorganic salts*, Atmos. Chem. Phys., 5, 1357–1370, doi:10.5194/acp-5-1357-2005.
- [25] Mochida, M., Miyakawa, T., Takegawa, N., Morino, Y., Kawamura, K., and Kondo, Y.: (2008), *Significant alteration in the hygroscopic properties of urban aerosol particles by the secondary formation of organics*, Geophys. Res. Lett., 35.
- [26] Petters, M. D. and Kreidenweis, S. M.(2007): A single parameter representation of hygroscopic growth and cloud condensation nucleus activity, Atmos. Chem. Phys., 7, 1961–1971,doi:10.5194/acp-7-1961-2007, 2007.
- [27] Wex, H., Stratmann, F., Hennig, T., Hartmann, S., Niedermeier, D., Nilsson, E., Ocskay, R., Rose, D., Salma, I., and Ziese, M.:(2008), *Connecting hygroscopic growth at high humidities to cloud activation for different particle types*, Environ. Res. Lett., 3, 1–10, doi:10.1088/1748-9326/3/3/035004.
- [28] Gunthe, S. S., King, S. M., Rose, D., Chen, Q., Roldin, P., Farmer, D. K., Jimenez, J. L., Artaxo, P., Andreae, M. O., Martin, S. T., and Pöschl, U.(2009): *Cloud condensation nuclei in pristine tropical rainforest air of Amazonia: size-resolved measurements and modeling of atmospheric aerosol composition and CCN activity*, Atmos. Chem. Phys., 9, 7551–7575, doi:10.5194/acp-9-7551-2009.
- [29] Petters, M. D., Carrico, C. M., Kreidenweis, S. M., Prenni, A. J., DeMott, P. J., Collett, J. L., and Moosuller, H.:(2009) *Cloud condensation nucleation activity of biomass burning aerosol*, J. Geo- phys. Res., 114, D22205, doi:10.1029/2009JD012353.
- [30] Good, N., Topping, D. O., Allan, J. D., Flynn, M., Fuentes, E., Irwin, M., Williams, P. I., Coe, H., and Mc Figgans, G.:(2010a) *Consistency between parameterisations of aerosol hygroscopicity and CCN activity during the RHaMBLE discovery cruise*, Atmos. Chem. Phys., 10, 3189–3203, doi:10.5194/acp-10-3189-2010.
- [31] Irwin, M., Good, N., Crosier, J., Choulaton, T. W., and McFiggans, G.:(2010) *Reconciliation of measurements of hygroscopic growth and critical supersaturation of aerosol particles in central Germany*, Atmos. Chem. Phys., 10, 11737–11752, doi:10.5194/acp-10-11737-2010.
- [32] Roberts, G. C., Day, D. A., Russell, L. M., Dunlea, E. J., Jimenez, J. L., Tomlinson, J.M.Collins, D. R., Shinozuka, Y., and Clarke, A. D.(2010): *Characterization of particle cloud droplet activity and composition in the free troposphere and the boundary layer during INTEX-B*, Atmos. Chem. Phys., 10, 6627–6644, doi:10.5194/acp-10-6627-2010.
- [33] Cerully, K. M., Raatikainen, T., Lance, S., Tkacik, D., Tiitta, P., Petaja, T., Ehn, M., Kulmala, M., Worsnop, D. R., Laaksonen, A., Smith, J. N., and Nenes, A.(2011): *Aerosol hygroscopicity and CCN activation kinetics in a boreal forest environment during the 2007 EUCAARI campaign*, Atmos. Chem. Phys., 11, 12369–12386, doi:10.5194/acp-11-12369-2011.
- [34] Duplissy, J., DeCarlo, P. F., Dommen, J., Alfarra, M. R., Metzger, A., Barmapadimos, I., Prevot, A. S. H., Weingartner, E., Tritscher, T., Gysel, M., Aiken, A. C., Jimenez, J. L., Canagaratna, M. R., Worsnop, D. R., Collins, D. R., Tomlinson, J., and Baltensperger, U.:(2011),*Relating hygroscopicity and composition of organic aerosol particulate matter*, Atmos. Chem. Phys., 11, 1155– 1165, doi:10.5194/acp-11-1155-2011.
- [35] Fors, E. O., Swietlicki, E., Svenningsson, B., Kristensson, A., Frank, G. P., and Sporre, M.: (2011), *Hygroscopic properties of the ambient aerosol in southern Sweden – a two year study*, Atmos. Chem. Phys., 11, 8343–8361, doi:10.5194/acp-11-8343-2011.
- [36] Metzger, S., Steil, B., Xu, L., Penner, J. E., and Lelieveld, J.(2012) *New representation of water activity based on a single solute specific constant to parameterize the hygroscopic growth of aerosols in atmospheric models*, Atmos. Chem. Phys., 12, 5429–5446, doi:10.5194/acp-12-5429-2012.
- [37] Kramer, L., Pöschl, U., and Niessner, R.(2000): *Microstructural rearrangement of sodium chloride condensation aerosol particles on interaction with water vapor*, J. Aerosol. Sci., 31, 6, 673–5 685, doi:10.1016/S0021-8502(99)00551-0.
- [38] Mikhailov, E., Vlasenko, S., Martin, S. T., Koop, T., and Pöschl, U.:(2009) *Amorphous and crystalline aerosol particles interacting with water vapor: conceptual framework and experimental evidence for restructuring, phase transitions and kinetic limitations*, Atmos. Chem. Phys., 9, 9491–9522, doi:10.5194/acp-9-9491-2009.
- [39] Gysel, M., Weingartner, E., Nyeki, S., Paulsen, D., Baltensperger, U., Galambos, I., and Kiss, G.:(2004) *Hygroscopic properties of water- soluble matter and humic-like organics in atmospheric fine aerosol*, Atmos. Chem. Phys., 4, 35–50, doi:10.5194/acp-4-35-2004.
- [40] Sjogren, S., Gysel, M., Weingartner, E., Baltensperger, U., Cubison, M. J., Coe, H., Zardini, Marcolli, C., Krieger, U. R., and Peter, T.: (2007) *Hygroscopic growth and water uptake kinetics of two- phase aerosol particles consisting of ammonium sulphate, adipic and humic acid mixtures*, J. Aerosol Sci., 38, 157–171.

- [41] Zardini, A. A., Sjogren, S., Marcolli, C., Krieger, U. K., Gysel, M., Weingartner, E., Baltensperger, U., and Peter, T.: (2008), *A combined particle trap/HTDMA hygroscopicity study of mixed in-organic/organic aerosol particles*, *Atmos. Chem. Phys.*, 8, 5589–5601, doi:10.5194/acp-8-5589-2008
- [42] Wang, J., Cubison, M., Aiken, A., Jimenez, J., and Collins, D. (2010): *The importance of aerosol mixing state and size-resolved composition on CCN concentration and the variation of the importance with atmospheric aging of aerosols*, *Atmos. Chem. Phys.*, 10, 7267–7283.
- [43] Hess M., Koepke P., and Schult I (May 1998), *Optical Properties of Aerosols and Clouds: The Software Package OPAC*, *Bulletin of the American Met. Soc.* 79, 5, p831–844.
- [44] Swietlicki, E., Zhou, J. C., Covert, D. S., Hameri, K., Busch, B., Vakeva, M., Dusek, U., Berg, O. H., Wiedensohler, A., Aalto, P., Makela, J., Martinsson, B. G., Papaspiropoulos, G., Mentes, B., Frank, G., and Stratmann, F. (2000): *Hygroscopic properties of aerosol particles in the northeastern Atlantic during ACE-2*, *Tellus*, 52B, 201–227.
- [45] Randles, C. A., Russell L. M. and Ramaswamy V. (2004) Hygroscopic and optical properties of organic sea salt aerosol and consequences for climate forcing, *Geophysical Research Letters*, Vol. 31, L16108, doi:10.1029/2004GL020628.
- [46] Stokes, R. H. and Robinson, R. A. (1966): *Interactions in aqueous nonelectrolyte solutions: I. Solute solvent equilibria*, *J. Phys. Chem.* 70, 2126–2130,
- [47] Meyer, N. K., Duplissy, J., Gysel, M., Metzger, A., Dommen, J., Weingartner, E., Alfarra, M. R., Prevot, A. S. H., Fletcher, C., Good, N., McFiggans, G., Jonsson, A. M., Hallquist, M., Baltensperger, U., and Ristovski, Z. D. (2009): *Analysis of the hygroscopic and volatile properties of ammonium sulphate seeded and unseeded SOA particles*. *Atmospheric Chemistry and Physics*, 9, 721–732, doi:10.5194/acp-9-721-2009.
- [48] Stock M., Y. F. Cheng, W. Birmili, A. Massling, B. Wehner, T. Muller, S. Leinert, N. Kalivitis, N. Mihalopoulos, and A. Wiedensohler, (2011). Hygroscopic properties of atmospheric aerosol particles over the Eastern Mediterranean: implications for regional direct radiative forcing under clean and polluted conditions. *Atmospheric Chemistry and Physics*, 11, 4251–4271, www.atmos-chem-phys.net/11/4251/2011/doi:10.5194/acp-11-4251-2011
- [49] Carrico, C. M., Petters, M. D., Kreidenweis, S. M., Sullivan, A. P., McMeeking, G. R., Levin, E. J. T., Engling, G., Malm, W. C., and Collett Jr., J. L. (2010): *Water uptake and chemical composition of fresh aerosols generated in open burning of biomass*, *Atmos. Chem. Phys.*, 10, 5165–5178, doi:10.5194/acp-10-5165-2010.
- [50] Rose, D., Nowak, A., Achtert, P., Wiedensohler, A., Hu, M., Shao, M., Zhang, Y., Andreae, M. O., and Pöschl, U.: *Cloud condensation nuclei in polluted air and biomass burning smoke near the mega-city Guangzhou, China – Part 1: Size-resolved measurements and implications for the modeling of aerosol particle hygroscopicity and CCN activity*, *Atmos. Chem. Phys.*, 10, 3365–3383, doi:10.5194/acp-10-3365-2010, 2010.
- [51] Rose, D., Gunthe, S. S., Su, H., Garland, R. M., Yang, H., Berghof, M., Cheng, Y. F., Wehner, B., Achtert, P., Nowak, A., Wiedensohler, A., Takegawa, N., Kondo, Y., Hu, M., Zhang, Y., Andreae, M. O., and Pöschl, U. (2011): *Cloud condensation nuclei in polluted air and biomass burning smoke near the mega-city Guangzhou, China – Part 2: Size-resolved aerosol chemical composition, diurnal cycles, and externally mixed weakly CCN-active soot particles*, *Atmos. Chem. Phys.* 11, 2817–2836, doi:10.5194/acp-11-2817-2011.
- [52] Mikhailov E, Merkulov V., Vlasenko S., Rose D., and Pöschl U., (2011), *Mass-based hygroscopicity parameter interaction model and measurement of atmospheric aerosol water uptake*; *Atmos. Chem. Phys. Discuss.*, 11, 30877–30918, 2011, doi:10.5194/acpd-11-30877-2011.