The Effect of Relative Humidity on the Hygroscopic Growth Factor and Bulk Hygroscopicity of water Soluble Aerosols

[1]D.O. Akpootu and [2]N. N. Gana

[1]*Department of Physics, Usmanu Danfodiyo University, Sokoto, Nigeria.*
[2]*Department of Physics, College of Education (Technical), Lafiagi, Kwara State, Nigeria.*

---**ABSTRACT**---
In this paper, the hygroscopicity properties of water soluble aerosols component based on microphysical properties of urban aerosols using data extracted from Optical Properties of Aerosols and Clouds (OPAC) incorporated with FORTRAN program were modeled to determine the effect of relative humidity on hygroscopic growth factor and bulk hygroscopicity at spectral range of 0.25-1.00 μm for eight relative humidities (RHs) (0, 50, 70, 80, 90, 95, 98 and 99%). The extracted microphysical properties are number mix ratio and volume mix ratio as a function of RH. The hygroscopic growth was observed to increase with RH from 0-99% RHs while the bulk hygroscopicity decreases with increase in RH from 0-99% RHs. We observed that the different models adopted excellently fits the data in terms of their coefficient of determination. The analysis further obtain the varying particles hygroscopicity from the proposed one-parameter equation.

KEYWORDS: *Relative humidity, hygroscopic growth factor, bulk hygroscopicity, aerosols, water soluble.*

Date of Submission**:** 21, October - 2013　　　　　　Date of Acceptance: 10, November - 2013

I. INTRODUCTION

The hygroscopic growth of aerosol particles are of current interest due to their effects on light scattering and absorption properties of particles which thereby affect the air quality, visibility, Earth's radiation and the climate (Charlson *et al*., 1992; IPCC, 2007). On a global basis, sulfates, nitrates and chlorides contribute the largest to the mass budget of fine atmospheric particles (IPCC, 2007; Li *et al.*, 2009; Shen *et al.*, 2009). These inorganic salt aerosols are hygroscopic by nature, thus their size, phase and subsequently the optical properties would be strongly influenced by the ambient relative humidity (RH). Based on recent studies, the effect of sulfate particles on the annual-average global direct radiative forcing, arising from the fluctuation of atmospheric particles between aqueous and solid state, is estimated up to as much as 24% (Martin *et al.*, 2004; Wang *et al.*, 2008a, 2008b). Thus, close attention has been paid to study the hygroscopic properties of these inorganic salts.

The interaction of aerosol particles with water vapor is among the central issues of current research in atmospheric and climate science, and numerous studies have investigated the hygroscopicity of aerosol particles and their ability to serve as cloud condensation nuclei (CCN). Köhler theory is the main tool to describe the hygroscopic growth of particles as a function of relative humidity (Pruppacher and Klett, 2000; Seinfeld and Pandis, 2006), and various types of Köhler models have been developed and applied for the analysis of laboratory and field measurement results as well as in numerical models of the atmosphere and climate (Junge and McLaren, 1971; Fitzgerald, 1973, Shulman *et al*., 1996; Kulmala *et al*., 1997; Laaksonen *et al*., 1998, Raymond and Pandis, 2003; Bilde and Svenningsson, 2004; Mikhailov *et al*., 2004, 2009; Huff Hartz *et al*., 2005; McFiggans *et al*., 2006; Svenningsson *et al*., 2006; Rose *et al*., 2008).

A central aim of hygroscopicity measurements and Köhler model studies is to relate the critical supersaturation of CCN activation to the hygroscopic growth factors observed at subsaturated conditions (Rissler *et al*., 2004; Kreidenweis *et al*., 2005; Mochida *et al*., 2006; Petters and Kreidenweis, 2007; Wex *et al*., 2008; Gunthe *et al*., 2009; Petters *et al*., 2009; Good *et al*., 2010a, b; Irwin *et al*., 2010; Roberts *et al*., 2010; Cerully *et al*., 2011; Duplissy *et al*., 2011; Fors *et al*., 2011; Metzger *et al*., 2012). For this purpose, Petters and Kreidenweis (2007) proposed a single-parameter Köhler model where the hygroscopicity parameter provides a volume-based measure of aerosol water uptake characteristics and CCN activity. For water soluble organic and inorganic compounds, the relative differences between values derived from CCN and growth factor measurements are usually less than 30% and can be explained by non-ideal behavior of concentrated solutions (Wex *et al*., 2008; Mikhailov *et al*., 2009).

This paper provides the impact of microphysical properties and relative humidity on the hygroscopic growth factor and bulk hygroscopicity of atmospheric water soluble aerosols. However, it is important to note that it is still less details till now to distinguish the impact of size-effect and RH on the growth factor of inorganic aerosols. Thus, to consider the impact of size-effect and RH synchronous will provide a new insight into visibility degradation and air quality improvement under ambient atmospheric condition.

II. METHODOLOGY

The models extracted from OPAC are given in table 1

Table 1: Compositions of aerosols types (Hess *et al.*, 1998)

Components	Model 1	Model 2	Model 3	Model 4	Model 5
	No.density (cm^{-3})	No.density (cm^{-3})	No.density (cm^{-3})	No.density (cm^{-3})	No.density (cm^{-3})
Insoluble	1.50	1.50	1.50	1.50	1.50
Water soluble	15,000.00	20,000.00	25,000.00	30,000.00	35,000.00
Soot	120,000.00	120,000.00	120,000.00	120,000.00	120,000.00
Total	135,001.50	140,001.50	145,001.50	150,001.50	155,001.50

The data used for the urban aerosols in this paper are derived from the Optical Properties of Aerosols and Clouds (OPAC) data set (Hess *et al.*, 1998). In this, a mixture of three components is used to describe Urban aerosols: a water soluble (WASO) components consist of scattering aerosols that are hygroscopic in nature, such as sulphates and nitrates present in anthropogenic pollution) water insoluble (INSO) and Soot.

The principal parameter used to characterize the hygroscopicity of the aerosol particles is the aerosol hygroscopic growth factor gf(RH), which indicates the relative increase in mobility diameter of particles due to water absorption at a certain RH and is defined as the ratio of the particle diameter at any RH to the particle diameter at RH=0 and RH is taken for seven values 50%, 70%, 80%, 90%, 95%, 98% and 99%. (Swietlicki *et al.*, 2008; Randles *et al.*, 2004):

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

The HGF can be subdivided into different classes with respect to hygroscopicity. One classification is based on diameter growth factor by Liu *et al.*,(2011) and Swietlicki *et al.*, (2008) as barely Hygroscopic (gf(RH) =1.0–1.11), Less Hygroscopic (gf(RH) =1.11–1.33), More Hygroscopic (gf(RH) = 1.33–1.85) and most hygroscopic growth (gf(RH) >1.85)

Most atmospheric aerosols are externally mixed with respect to hygroscopicity, and consist of more and less hygroscopic sub-fractions (Swietlicki *et al.*, 2008). The ratio between these fractions as well as their content of soluble material determines the hygroscopic growth of the overall aerosol. Particle hygroscopicity may vary as a function of time, place, and particle size (McMurry and Stolzenburg, 1989; Swietlicki *et al.*, 2008).

Prediction of hygroscopic growth factors with Köhler theory requires detailed knowledge of particle composition as well as a thermodynamic model, which describes the concentration dependence of the water activity for such a mixture. The hygroscopic growth factor of a mixture, gf$_{mix}$(RH), can be estimated from the growth factors of the individual components of the aerosol and their respective volume fractions, V_k, using the Zdanovskii-Stokes-Robinson relation (ZSR relation; Sjogren *et al.*, 2007; Stokes and Robinson, 1966; Meyer *et al.*, 2009; Stokes *et al.*, 1966; Stock *et al.*, 2011):

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

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

It can also be computed using the corresponding number fractions n_k as (Duplissy *et al.*, 2011; Meier *et al.*, 2009):

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

Where n_k is the number fraction of particles having the growth factor gf$_k$.

The RH dependence of $gf_{mix}(RH)$ can be parameterized in a good approximation by a one-parameter equation, proposed e.g. by Petters and Kreidenweis (2007):

$$gf_{mix}(a_w) = \left(1 + \kappa \frac{a_w}{1-a_w}\right)^{\frac{1}{3}}$$ (4)

Here, a_w is the water activity, which can be replaced by the relative humidity RH at equilibrium (Seinfeld and Pandis, 2006), if the Kelvin effect is negligible, as for particles with sizes more relevant for light scattering and absorption. The coefficient κ is a simple measure of the particle's hygroscopicity and captures all solute properties (Raoult effect).

Humidograms of the ambient aerosols obtained in various atmospheric conditions showed that $gf_{mix}(RH)$ could as well be fitted well with a γ-law (Swietlicki *et al*., 2000; Gysel *et al*., 2009; Putaud, 2012) as

$$gf_{mix}(RH) = \left(1 - \frac{RH}{100}\right)^{\gamma}$$ (5)

Particle hygroscopicity is a measure that scales the volume of water associated with a unit volume of dry particle (Petters and Kreidenweis, 2007) and depends on the molar volume and the activity coefficients of the dissolved compounds (Christensen and Petters, 2012).
The bulk hygroscopicity factor B under subsaturation RH conditions was determined using the following relation:

$$B = (1 - gf_{mix}^3)\ln a_w$$ (6).

III. RESULTS AND DISCUSSION

Table 2: The growth factor and bulk hygroscopicity of the aerosols using number mix ratio for model 1-5

RH	Model 1		Model 2		Model 3		Model 4		Model 5	
	gf_{mix}	Bulk Hyg	gf_{mix}	Bulk Hyg	gf_{mix}	Bulk Hyg	gf_{mix}	Bulk Hyg	gf_{mix}	Bulk Hyg
50	1.03185	0.06836	1.04061	0.08792	1.04861	0.10607	1.05598	0.12305	1.06278	0.13892
70	1.05037	0.05665	1.06392	0.07287	1.07620	0.08791	1.08744	0.10198	1.09774	0.11514
80	1.06941	0.04976	1.08770	0.06401	1.10413	0.07722	1.11907	0.08958	1.13269	0.10113
90	1.11342	0.04007	1.14195	0.05154	1.16720	0.06218	1.18988	0.07213	1.21032	0.08144
95	1.17677	0.03229	1.21864	0.04154	1.25506	0.05011	1.28732	0.05813	1.31608	0.06563
98	1.28996	0.02316	1.35260	0.02979	1.40593	0.03594	1.45241	0.04170	1.49331	0.04707
99	1.38634	0.01673	1.46448	0.02152	1.53018	0.02596	1.58691	0.03011	1.63651	0.03400

Generally, there is an overall increase in aerosol hygroscopic growth factor for number mix ratio model with increase in RHs from 50-99% RH. However, the rate of increase in the aerosol hygroscopic growth factor with RHs is more pronounced as from 90-98% RHs and subsequently decrease at 99% RH; this shows that the aerosols absorbs more water vapour at 90-98% RHs and becomes saturated at this point of deliquescence. The bulk hygroscopicity, B decreases with increase in RH.

More so, we observed that as we moved from model 1-5, the increase in growth factor with RHs from 50-99% RHs increases as well, and this implies that the particles absorb more water vapour. Similarly, the bulk hygroscopicity, B increases as we moved from model 1-5.
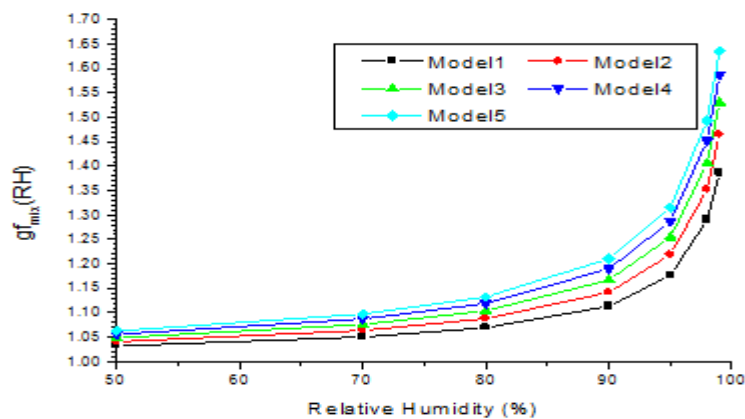

Figure 1: A graph of growth factor of the mixture using number mix ratio (model 1-5)
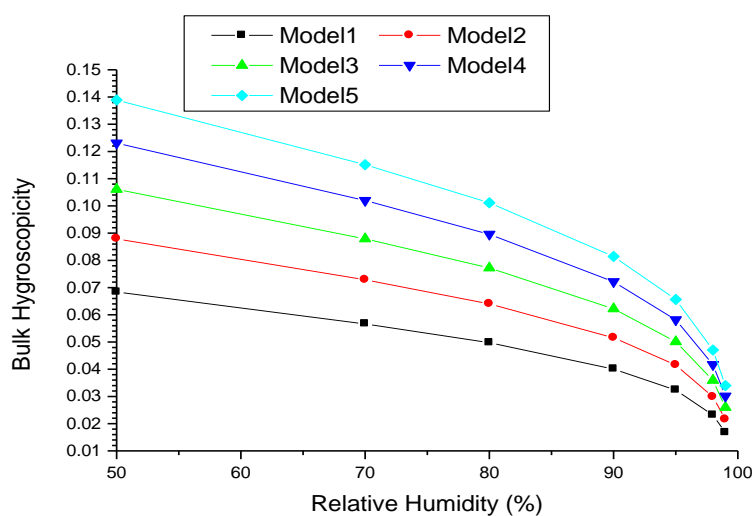
Figure 2: A graph of bulk hygroscopicity of the mixture using number mix ratio (model 1-5)

Figure 1 shows a non-linear increase in aerosol hygroscopic growth factor with RHs. The deliquescence point was observed as from 90-98% RHs. However, from 50-80% RHs indicates that the smallest particles are found in this region; this is line with that reported by (Dawei *et al*., 2010) that smallest particles show the highest growth factor. It is important to note that as seen from the figure the growth factor gradually lift up with increasing RH. This may be attributed that higher RH means more moisture in the atmosphere leads the aerosol particles to absorb more water vapour on particle surface. From the range of values observed, for the gf_{mix} the mixture as depicted in figure 1 can be described as barely hygroscopic, less hygroscopic and more hygroscopic growth (Liu *et al*., 2011; Swietlick *et al*., 2008). The bulk hygroscopicity, B decreases with increase in RHs as displayed in figure 2

Table 3: The growth factor for number mix ratio using model 1

RH (%)	gf_{mix}	RH/1-RH	gf_{mix}^3		ln(1-RH/100)	$lngf_{mix}$
50	1.031847	1.000000	1.098617		-0.69315	0.031351
70	1.050366	2.333333	1.158835		-1.20397	0.049138
80	1.069407	4.000000	1.223007		-1.60944	0.067104
90	1.113423	9.000000	1.380321		-2.30259	0.107439
95	1.176771	19.000000	1.629582		-2.99573	0.162774
98	1.289956	49.000000	2.146467		-3.91202	0.254608
99	1.386338	99.000000	2.664450		-4.60517	0.326666

Table 4: The growth factor for number mix ratio using model 2

RH (%)	gf_{mix}	RH/(1-RH)	gf_{mix}^3		ln(1-RH/100)	$lngf_{mix}$
50	1.040609	1.000000	1.12684		-0.69315	0.039806
70	1.063925	2.333333	1.204294		-1.20397	0.061965
80	1.087696	4.000000	1.286834		-1.60944	0.084062
90	1.141954	9.000000	1.489177		-2.30259	0.132741
95	1.218640	19.00000	1.809783		-2.99573	0.197736
98	1.352600	49.000000	2.474615		-3.91202	0.302028
99	1.464478	99.00000	3.140860		-4.60517	0.381499

Table 5: The growth factor for number mix ratio using model 3

RH (%)	gf$_{mix}$	RH/1-RH	gf$_{mix}$^3	ln(1-RH/100)	lngf$_{mix}$
50	1.048607	1.000000	1.153022	-0.69315	0.047462
70	1.076201	2.333333	1.246466	-1.20397	0.073437
80	1.104129	4.000000	1.346045	-1.60944	0.099057
90	1.167204	9.000000	1.590159	-2.30259	0.154611
95	1.255062	19.000000	1.976950	-2.99573	0.227185
98	1.405932	49.000000	2.779029	-3.91202	0.340701
99	1.530176	99.000000	3.582812	-4.60517	0.425383

Table 6: The growth factor for number mix ratio using model 4

RH (%)	gf$_{mix}$	RH/1-RH	gf$_{mix}$^3	ln(1-RH/100)	lngf$_{mix}$
50	1.05598	1.000000	1.177518	-0.69315	0.05447
70	1.087439	2.333333	1.285921	-1.20397	0.083825
80	1.119073	4.000000	1.401442	-1.60944	0.112501
90	1.189877	9.000000	1.684637	-2.30259	0.17385
95	1.287323	19.000000	2.133351	-2.99573	0.252565
98	1.452408	49.000000	3.063837	-3.91202	0.373223
99	1.586911	99.000000	3.996300	-4.60517	0.46179

Table 7: The growth factor for number mix ratio using model 5

RH (%)	gf$_{mix}$	RH/1-RH	gf$_{mix}$^3	ln(1-RH/100)	ln gf$_{mix}$
50	1.062781	1.000000	1.200416	-0.69315	0.060889
70	1.097737	2.333333	1.322804	-1.20397	0.093251
80	1.132690	4.000000	1.453227	-1.60944	0.124595
90	1.210317	9.000000	1.772953	-2.30259	0.190882
95	1.316083	19.000000	2.279551	-2.99573	0.27466
98	1.493314	49.000000	3.330071	-3.91202	0.400998
99	1.636507	99.000000	4.382821	-4.60517	0.492564

Tables (3-7) shows the data obtained using equations (4) and (5) for the number mix ratio
The results of the modelling using equations (4) and (5) are displayed in the table below

Table 8: Summary of the results of R^2, k, c and γ for the number mix ratio.

Used equation	R^2	k	Constant	γ	Model used
4	0.962	0.016	1.198		Model 1
5	0.973		-0.047	-0.077	
4	0.962	0.020	1.255		Model 2
5	0.978		-0.048	-0.048	
4	0.962	0.025	1.307		Model 3
5	0.982		-0.048	-0.098	
4	0.962	0.029	1.356		Model 4
5	0.984		-0.047	-0.106	
4	0.962	0.032	1.402		Model 5
5	0.986		-0.045	-0.113	

The fitted curve can be represented by one of the empirical parameters fit in the form of either equations (4) or (5) However, we observed that equation (5) gives a higher coefficient of determination, R^2

Table 9: The growth factor and bulk hygroscopicity of the aerosols using volume mix ratio for model(1-5)

| RH	Model 1		Model 2		Model 3		Model 4		Model 5	
	gf_{mix}	Bulk Hyg	gf_{mix}	Bulk Hyg	gf_{mix}	Bulk Hyg	gf_{mix}	Bulk Hyg	gf_{mix}	Bulk Hyg
50	1.12039	0.28170	1.13706	0.32587	1.14951	0.35971	1.15920	0.38654	1.16688	0.40814
70	1.20384	0.26560	1.22672	0.30175	1.24315	0.32857	1.25555	0.34928	1.26524	0.36576
80	1.29096	0.25695	1.31790	0.28763	1.33670	0.30980	1.35054	0.32654	1.36126	0.33972
90	1.48494	0.23963	1.51575	0.26155	1.53643	0.27678	1.55127	0.28796	1.56237	0.29646
95	1.73739	0.21771	1.76819	0.23227	1.78820	0.24201	1.80222	0.24896	1.81256	0.25416
98	2.12661	0.17410	2.15357	0.18158	2.17057	0.18640	2.18226	0.18975	2.19075	0.19221
99	2.41833	0.13209	2.44184	0.13628	2.45641	0.13891	2.46639	0.14074	2.47359	0.14206

Generally, there is an overall increase in aerosol hygroscopic growth factor for volume mix ratio model with increase in RHs from 50-99% RH. However, the rate of increase in the aerosol hygroscopic growth factor with RHs is more pronounced as from 90-98% RHs and subsequently decrease at 99% RH; this shows that the aerosols absorbs more water vapour at 90-98% RHs and becomes saturated at this point of deliquescence. The bulk hygroscopicity, B decreases with increase in RH.

More so, we observed that as we moved from model 1-5, the increase in growth factor with RHs from 50-99% RHs increases as well and this implies that the particles absorb more water vapour. Similarly, the bulk hygroscopicity, B increases as we moved from model 1-5.

Though, we observed that the relative differences between the hygroscopicity parameter, k, values derived from the growth factor are less than 30% using the volume based mix ratio measure of aerosol water uptake and this is in good agreement with the single-parameter Köhler model proposed by Petters and Kreidenwis (2007) and can be explained by non-ideal behavior of concentrated solutions (Wex *et al*., 2008; Mikhailov *et al*., 2009). However, it is slightly above 30% at RH between 95% and 98% RHs.
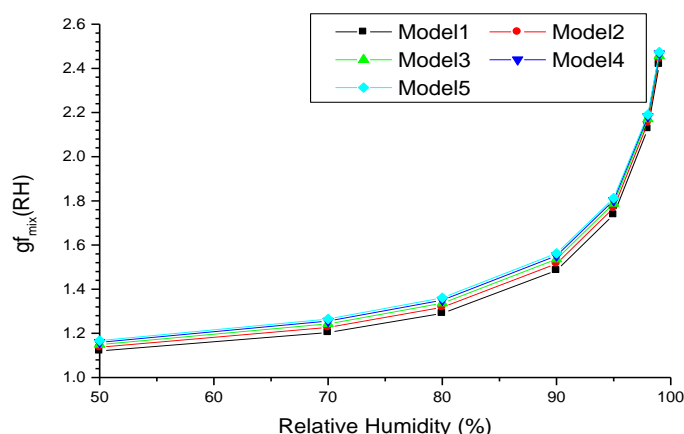
Figure 3: A graph of growth factor of the mixture using volume mix ratio (model 1-5)
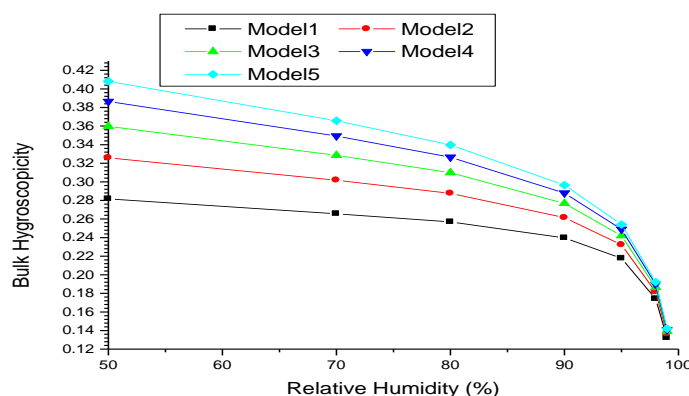
Figure 4: A graph of bulk hygroscopicity of the mixture using volume mix ratio (model 1-5)

Figure 3 shows a non-linear increase in aerosol hygroscopic growth factor with RHs. The deliquescence point was observed as from 90-98% RHs. However, from 50-80% RHs indicates that the smallest particles are found in this region; this is line with that reported by (Dawei *et al*., 2010) that smallest particles show the highest growth factor. It is important to note that as seen from the figure the growth factor gradually lift up with increasing RH. This may be attributed that higher RH means more moisture in the atmosphere leads the aerosol particles to absorb more water vapour on particle surface. From the range of values observed, for the gf$_{mix}$ the mixture as depicted in figure 3 can be described as less hygroscopic, more hygroscopic and most hygroscopic growth (Liu *et al*., 2011; Swietlick *et al*., 2008). The bulk hygroscopicity, B decreases with increase in RHs as displayed in figure 4

Table 10: The growth factor for volume mix ratio using model 1

RH (%)	gf$_{mix}$	RH/1-RH	gf$_{mix}$^3		ln(1-RH/100)	lngf$_{mix}$
50	1.120392	1.000000	1.406405		-0.69315	0.113679
70	1.203843	2.333333	1.744656		-1.20397	0.185519
80	1.290965	4.000000	2.151508		-1.60944	0.25539
90	1.48494	9.000000	3.274361		-2.30259	0.395374
95	1.737393	19.000000	5.244381		-2.99573	0.552386
98	2.126606	49.000000	9.617480		-3.91202	0.754527
99	2.418332	99.000000	14.14320		-4.60517	0.883078

Table 11: The growth factor for volume mix ratio using model 2

RH (%)	gf$_{mix}$	RH/(1-RH)	gf$_{mix}$^3		ln(1-RH/100)	lngf$_{mix}$
50	1.137065	1.000000	1.470130		-0.69315	0.12845
70	1.226718	2.333333	1.846012		-1.20397	0.204342
80	1.317898	4.000000	2.288999		-1.60944	0.276038
90	1.515746	9.000000	3.482407		-2.30259	0.415908
95	1.768195	19.000000	5.528282		-2.99573	0.569959
98	2.153567	49.000000	9.987927		-3.91202	0.767126
99	2.441843	99.000000	14.559720		-4.60517	0.892753

Table 12: The growth factor for volume mix ratio using model 3

RH (%)	gf$_{mix}$	RH/1-RH	gf$_{mix}$^3		ln(1-RH/100)	lngf$_{mix}$
50	1.149513	1.000000	1.518945		-0.69315	0.139339
70	1.243153	2.333333	1.921206		-1.20397	0.217651
80	1.336697	4.000000	2.388353		-1.60944	0.290201
90	1.536433	9.000000	3.626943		-2.30259	0.429463
95	1.788204	19.000000	5.718097		-2.99573	0.581212
98	2.170568	49.000000	10.22634		-3.91202	0.774989
99	2.456412	99.000000	14.82189		-4.60517	0.898702

Table 13: The growth factor for volume mix ratio using model 4

RH (%)	gf$_{mix}$	RH/1-RH	gf$_{mix}$^3		ln(1-RH/100)	lngf$_{mix}$
50	1.159196	1.000000	1.557653		-0.69315	0.147727
70	1.25555	2.333333	1.979256		-1.20397	0.227574
80	1.350544	4.000000	2.463351		-1.60944	0.300508
90	1.551275	9.000000	3.733071		-2.30259	0.439077
95	1.802217	19.000000	5.853571		-2.99573	0.589017
98	2.182261	49.000000	10.39251		-3.91202	0.780362
99	2.466388	99.000000	15.00321		-4.60517	0.902755

Table 14: The growth factor for volume mix ratio using model 5

RH (%)	gf_{mix}	RH/1-RH	$gf_{mix}{}^3$		ln(1-RH/100)	$lngf_{mix}$
50	1.166878	1.000000	1.588824		-0.69315	0.154331
70	1.265245	2.333333	2.025461		-1.20397	0.235266
80	1.361256	4.000000	2.522432		-1.60944	0.308408
90	1.56237	9.000000	3.813747		-2.30259	0.446204
95	1.812563	19.000000	5.954965		-2.99573	0.594742
98	2.190749	49.000000	10.51424		-3.91202	0.784244
99	2.47359	99.000000	15.13503		-4.60517	0.905671

Tables (10-14) shows the data obtained using equations (4) and (5) for the volume mix ratio
The results of the modelling using equations (4) and (5) are displayed in the table below

Table 15: Summary of the results of R^2, k, c and γ for the volume mix ratio.

Used equation	R^2	k	Constant	γ	Model used
4	0.974	0.131	1.938		Model 1
5	0.997		-0.054	-0.203	
4	0.971	0.134	2.077		Model 2
5	0.998		-0.033	-0.201	
4	0.969	0.136	2.177		Model 3
5	0.998		-0.018	-0.199	
4	0.968	0.138	2.251		Model 4
5	0.999		-0.006	-0.198	
4	0.967	0.138	2.310		Model 5
5	0.999		0.003	-0.197	

The fitted curve can be represented by one of the empirical parameters fit in the form of either equations (4) or (5) However, we observed that equation (5) gives a higher coefficient of determination, R^2

IV. CONCLUSION

In this manuscript, we present a number mix and volume mix microphysical properties based hygroscopicity parameter model approach for efficient description of water uptake by mixed particles water soluble atmospheric aerosols.

The analysis for the relative differences between the hygroscopicity parameter values obtained from the growth factor using the volume based mix ratio measure of aerosol water uptake are found to be less than 30% and this is in good agreement with the single-parameter Köhler model proposed by Petters and Kreidenwis (2007), though, we observed that it is slightly above 30% at RH between 95% and 98% RHs, however, there was relatively small percentage values for number based mix ratio model.

The results shows that the growth factor increases with increase in RH and is more pronounced at 90-98% RHs, similarly, the bulk hygroscopicity decreases with increase in RHs. From this fact, we can safely conclude that there is a direct and inverse variation between the hygroscopic growth factor and bulk hygroscopicity with RHs. The growth factor indicates that the mixture are described to be barely hygroscopic, less hygroscopic, more hygroscopic and most hygroscopic growth.

REFERENCES

[1]. 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.

[2]. Cerully, K. M., Raatikainen, T., Lance, S., Tkacik, D., Tiitta, P., Petäjä, 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.

[3]. Charlson, R.J., Schwartz, S.E., Hales, J.M., Cess, R.D., Coakley, J.A., Hansen, J.E. and Hofmann, D.J. (1992): Climate Forcing by Anthropogenic Aerosols. *Science.* 255: 423–430.

[4]. Christensen, S. I. and Petters, M. D. (2012): The role of temperature in cloud droplet activation, *J. Phys. Chem.* A 116(39): 9706–9717.

[5]. Dawei, H., Liping, Q., Jianmin, C., Xingnan, Y., Xin, Y., Tiantao, C., Wen, F. (2010): Hygroscopicity of Inorganic Aerosols: Size and Relative Humidity Effects on the Growth Factor, *Aerosol and Quality Research*., 10, 255-264.

[6]. Duplissy, J., DeCarlo, P. F., Dommen, J., Alfarra, M. R., Metzger, A., Barmpadimos, 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

[7]. aerosol particulate matter, *Atmos. Chem. Phys*., 11, 1155– 1165, doi:10.5194/acp-11-1155-2011.

[8]. Fitzgerald, J. W. (1973): Dependence of supersaturation spectrum of CCN on aerosol size distribution and composition, *J. Atmos. Sci*., 30, 628–634.

[9]. 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.

[10]. Good, N., Topping, D. O., Allan, J. D., Flynn, M., Fuentes, E., Irwin, M., Williams, P. I., Coe, H., and McFiggans, 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.

[11]. Good, N., Topping, D. O., Duplissy, J., Gysel, M., Meyer, N. K., Metzger, A., Turner, S. F., Baltensperger, U., Ristovski, Z., Weingartner, E., Coe, H., and McFiggans, G. (2010b): Widening the gap between measurement and modelling of secondary organic aerosol properties?, *Atmos. Chem. Phys*., 10, 2577–2593, doi:10.5194/acp-10-2577-2010.

[12]. 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 P¨oschl, 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.

[13]. Gysel, M., McFiggans, G. B., and Coe, H. (2009): Inversion of tandem differential mobility analyser (tdma) measurements, *J. Aerosol Sci*., 40, 134–151, doi:10.1016/j.jaerosci.2008.07.013.

[14]. Hess, M., Koepke, P and Schult, I (1998): Optical Properties of Aerosols and Clouds. *American Meteorology Society.*

[15]. 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 monoterpene and sesquiterpene secondary organic aerosol, *J. Geophys. Res*., 110, D14208, doi:10.1029/2004JD005754.

[16]. IPCC (2007): Climate Change 2007: The Scientific Basis. In Solomon, S., Ding, Y., Griggs, D.G., Noguer, M., Vanderlinden, P.G., Dai, X., Maskell, K. and Johnson, C.A. (Eds). Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change. Cambridge University Press. Cambridge.

[17]. Irwin, M., Good, N., Crosier, J., Choularton, 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.

[18]. Junge, C. and McLaren, E. (1971): Relationship of cloud nuclei spectra to aerosol size distribution and composition, J. Atmos. Sci., 28, 382–390

[19]. 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.

[20]. Kulmala, M., Laaksonen, A., Charlson, R. J., and Korhonen, P. (1997): Clouds without supersaturation, Nature, 388, 336–337.

[21]. [20] Laaksonen, A., Korhonen, P., Kulmala, M., and Charlson, R. J. (1998):Modification of the K¨ohler equation to include soluble trace gases and slightly soluble substances, *J. Atmos. Sci*., 55, 853–862.

[22]. Li, W.F, Bai, Z.P., Liu, A.X., Chen, J. and Chen, L. (2009): Characteristics of Major PM2.5 Components during Winter in Tianjin, China. *Aerosol Air Qual. Res*.,9: 105–119.

[23]. Liu, P. F., Zhao, C. S., Gobel, T., Hallbauer, E., Nowak, A., Ran, L., Xu, W. Y., Deng, Z. Z., Ma, N., Milderger, K., Henning, S., Stratmann, F., and Wiedensohler, A. (2011): Hygroscopic Properties of Aerosol Particles at High Relative Humidity and their Diurnal Variations in the North China Plain, *Atm. Chem. Phys.*, Discuss., 11, 2991-3040.

[24]. Martin, S.T., Hung, H.M., Park, R.J., Jacob, D.J., Spurr, R.J.D., Chance, K.V. and Chin, M. (2004): Effects of the Physical State of Tropospheric Ammonium-Sulfate-Nitrate Particles on Global Aerosol Direct Radiative Forcing. *Atmos. Chem. Phys.*, 4: 183–214.

[25]. 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.*

[26]. *Phys.*, 6, 2593–2649, doi:10.5194/acp-6-2593-2006.

[27]. McMurry, P.H., and Stolzenburg, M.R. (1989): On the sensitivity of particle size to relative humidity for Los Angeles aerosols. *Atmos Environ*., 23:497–507. doi:10.1016/0004-6981(89) 90593-3

[28]. Meier J., Wehner, B., Massling, A., Birmili, W., Nowak, A., Gnauk, T., Br¨uggemann, E., Herrmann, H., Min, H., and Wiedensohler, A. (2009): Hygroscopic growth of urban aerosols particles in Beijing (China) during wintertime: A comparison of three experimental methods., *Atmos. Chem. Phys*., 9, 6865–6880, www.atmos-chem-phys.net/9/6865/2009/

[29]. 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.

[30]. 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, *Atmos. Chem. Phys.*, 9, 721–732, doi:10.5194/acp-9-721-2009.

[31]. Mikhailov, E., Vlasenko, S., Niessner, R., and P˘oschl, U.(2004): Interaction of aerosol particles composed of protein and saltswith water vapor: hygroscopic growth and microstructural rearrangement, *Atmos. Chem. Phys.*, 4, 323–350, doi:10.5194/acp-4-323-2004

[32]. 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.

[33]. Mochida, M., Kuwata, M., Miyakawa, T., Takegawa, N., Kawamura, K., and Kondo, Y. (2006): Relationship between hygroscopicity and cloud condensation nuclei activity for urban aerosols in Tokyo, *J. Geophys. Res.*, 111, D23204, doi:10.1029/2005JD006980.

[34]. 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.

[35]. Petters, M. D., Carrico, C. M., Kreidenweis, S. M., Prenni, A. J., DeMott, P. J., Collett, J. L., and Moosm¨uller, H. (2009): Cloud condensation nucleation activity of biomass burning aerosol, *J. Geophys. Res.*, 114, D22205, doi:10.1029/2009JD012353.

[36]. Pruppacher, H. R. and Klett, J. D. (2000): Microphysics of clouds and precipitation, Kluwer Academic Publishers, Dordrecht,

[37]. Putaud, J.P. (2012): Aerosol hygroscopicity at Ispra EMEP-GAW station by M. Adam et al., *Atmos. Chem. Phys. Discuss.*, 12, C1316–C1322

[38]. Randles, C. A., Russell, L. M., and Ramaswamy, V. (2004): Hygroscopic and optical properties of organic sea salt aerosol and consequences for climate forcing, *Geographical Research Letters.*, Vol. 31, LI6108, doi: 10, 1029/2004GL020628.

[39]. 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.

[40]. Rissler, J., Swietlicki, E., Zhou, J., Roberts, G., Andreae, M. O., Gatti, L. V., and Artaxo, P. (2004): Physical properties of the submicrometer aerosol over the Amazon rain forest during the wetto- dry season transition – comparison of modeled and measured CCN concentrations, *Atmos. Chem. Phys.*, 4, 2119–2143, doi:10.5194/acp-4-2119-2004.

[41]. 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.

[42]. Rose, D., Gunthe, S. S., Mikhailov, E., Frank, G. P., Dusek, U., Andreae, M. O., and P˘oschl, U.(2008): Calibration and measurement uncertainties of a continuous-flow cloud condensation nuclei counter (DMT-CCNC): CCN activation of ammonium sulfate and sodium chloride aerosol particles in theory and experiment,

[43]. *Atmos. Chem. Phys.*, 8, 1153–1179, doi:10.5194/acp-8-1153-2008.

[44]. 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.

[45]. Shen, Z.X., Cao, J.J, Tong, Z., Liu, S.X., Reddy, L.S.S., Han, Y.M, Zhang, T. and Zhou, J. (2009): Chemical Characteristics of Submicron Particles in Winter in Xi'an. *Aerosol Air Qual. Res.* 9: 80–93.

[46]. 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.

[47]. Sjogren, S., Gysel, M., Weingartner, E., Baltensperger, U., Cubison, M. J., Coe, H., Zardini, A. A., Marcolli, C., Krieger, U. K., and Peter, T. (2007): Hygroscopic growth and water uptake kinetics of two-phase aerosol particles consisting of ammonium sulfate, adipic and humic acid mixtures, *J. Aerosol Sci.*, 38, 157–171, doi:10.1016/j.jaerosci.2006.11.005.

[48]. Stock, M., Cheng, Y. F. Birmili, W., Massling, A., Wehner, B., Muller, T., Leinert, S., Kalivitis, N., Mihalopoulos, N., and Wiedensohler. A. (2011): *Atmos. Chem. Phys.*, 11, 4251–4271, www.atmos-chemphys.

[49]. net/11/4251/2011/ doi:10.5194/acp-11-4251-2011

[50]. Stokes, R. H., and Robinson, R. A. (1966): Interactions in aqueous nonelectrolyte solutions. I. Solute solvent equilibria, *J. Phys. Chem.*, 70, 2126–2130.

[51]. 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

[52]. 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.

[53]. [48] Swietlicki, E., Zhou, J., Covert, D. S., Hameri, K., Busch, B., Vakeva, M., Dusek, U., Berg, O. H.,Wiedensohler, A., Aalto, P., Makela, J., Marinsson, B. G., Papaspiropoulos, G., Mentes, B., Frank, G. and Stratch. F.(2000):Hygroscopic properties of aerosol particles in the north-eastern Atlantic during ACE-2.*Tellus* 52B, 201–227.

[54]. Swietlicki, E., Hansson, H.-C., H¨ameri, K., Svenningsson, B., Massling, A., McFiggans, G., McMurry, P. H., Petäjä, T., Tunved, P., Gysel, M., Topping, D., Weingartner, E., Baltensperger, U., Rissler, J., Wiedensohler, A., and Kulmala, M. (2008): Hygroscopic properties of submicrometer atmospheric aerosol particles measured with H-TDMA instruments in various environments 10 – a review, *Tellus* B, 60(3), 432–469,. 6892, 6898, 6906, 6909

[55]. Wang, J., Hoffmann, A.A., Park, R.J., Jacob, D.J. and Martin, S.T. (2008a). Global Distribution of Solid and Aqueous Sulfate Aerosols: Effect of the Hysteresis of Particle Phase Transitions. *J. Geophys. Res.* 113: D11206. doi: 10.1029/2007JD009367

[56]. Wang, J., Jacob, D.J. and Martin S.T. (2008b). Sensitivity of Sulfate Direct Climate Forcing to the Hysteresis of Particle Phase Transitions. *J. Geophys. Res.* 113: D11207. doi: 10.1029/2007JD009368.

[57]. 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.