



THE EFFECT OF WATER SOLUBLES ON KELVIN EFFECTS OF THE MARITIME POLLUTED AEROSOLS

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ABSTRACT

In this work microphysical properties of Maritime Polluted aerosols were extracted from Optical Properties of Aerosols and Clouds (OPAC) after varying the concentrations of water soluble at five different levels. The analytical expressions for the changes in the equilibrium relative humidity (RH), effective radii, effective hygroscopic growth, their magnitudes and fractional changes on the effects of surface tension (the Kelvin effect) on the ambient aerosols were numerically analyzed. The expressions were applied to two - one parameter models. It was discovered that the increase in water soluble concentration caused decrease in the Kelvin effects which implies decrease in surface tension. The numerical analysis of the data showed that to the lowest order error. Increase in the overestimation of the effective hygroscopic growth and decrease in the overestimation of the effective radii. On the two models applied, we discovered that the fractional changes in hygroscopic growth increase in overestimation while there is decrease in overestimation of fractional changes in effective radii. Finally, decrease in the underestimation of the fractional changes in the equilibrium pressure it was discovered.

Keywords: Kelvin effect, effective radius, effective hygroscopic growth, water soluble, ambient Relative Humidity, overestimation.

INTRODUCTION

In the real atmosphere, RH is the main component of atmospheric aerosols and its amount depends on the particle physical and chemical compositions and their effective sizes. The growth of aqueous droplets in humid air is commonly described by Köhler theory (Köhler, 1936; Gysel et al., 2002; Kreidenweis et al., 2005, Tijjani et al., 2015). It states that the ambient RH equals the product of water activity for a plane solution surface, and that of the Kelvin effect of the curved surfaces such as aerosol particles. The water activity considers the lowering of the equilibrium vapor pressure with increasing solute concentration at a given RH, is complemented by the Kelvin-term, which accounts for the increase in the water vapor pressure due to the curvature of the particle surface. The Kelvin factor which causes the increase in the surface energy with the decreasing particle diameter (Russell & Ming, 2002), causes the hygroscopic growth to decrease with decreasing dry size for particles of identical composition, depending on RH, chemical composition, and the RH history of the particles (Martin, 2000). Water soluble organic and inorganic can make up an important fraction of atmospheric aerosols. They can dominate the CCN activity and hygroscopic growth depending on chemical compositions (Laaksonen et al., 1998; Nenes et al., 2002; Anttila and Kerminen, 2002; Kreidenweis et al., 2006; McFiggans et al., 2006; Sun and Ariya 2006) by enhancing the Raoult effect and particle size (Kelvin effect; Thomson, 1871). Water-soluble aerosol particles grow by condensation of water vapor simply because the water activity of the aqueous solution

strives to equilibrate to the RH in the surrounding air in accordance with the Köhler equation. The Köhler theory presented the theory of cloud condensation nuclei (CCN) activation of atmospheric aerosol particles as the activation due to the thermodynamic balance of two effects: a decrease in water vapor pressure due to the solute (Raoult effect) and an increase in water vapor pressure due to the curvature at the air-liquid interface (Kelvin effect). These two effects are the major parameters that made hygroscopic growth to be size and composition dependent.

The modern concepts of Köhler equations enable the determination of the equilibrium size of an aerosol droplet for a given dry size, chemical composition, RH and Temperature, by accounting for the dissolution of gases into droplets, changes in surface tension, ion charges, or density of the droplet solutions (e.g. Russell and Ming, 2002; Mikhailov et al., 2004; Biskos et al., 2006a, b; Seinfeld and Pandis, 2006; McFiggans et al., 2006; Rose et al., 2008; Mikhailov et al., 2009; Ruehl et al., 2010) which is always smaller than the ambient RH.

In this paper some microphysical properties of maritime polluted aerosols were extracted from OPAC by varying the concentrations of water soluble at relative humidities of 0, 50, 70, 80, 90, 95, 98 and 99% to determine the effects of water soluble on Kelvin effect and water activity on these aerosols.

The microphysical properties extracted are the radii of the aerosols and volume mix ratios and were used to determine the effective hygroscopic and effective radii of the mixtures.

The analytical expressions derived by Lewis (2006) for the decrease in the equilibrium radius of a solution drop due to the Kelvin effect with dependences on surface tension, particle size and the estimates for the magnitude of these effects in various situations are numerically analyzed. The analytical expressions derived by Lewis (2006) for the changes in the equilibrium radius of solutions drops, the hygroscopic growth, estimates in the changes of their magnitudes and their fractional changes due to the Kelvin effect with dependences on surface tension, particle sizes are also numerically analyzed. We then applied the expressions to two known one parameter models. They are the power law dependence (γ -model) and the model as proposed by Petters and Kreidenweis (2007).

Though in our paper, Tijjani, et al (2015), we discovered that, the Kelvin effect and its consequences on the atmospheric aerosol depends on the hygroscopicity of the aerosol but in this work, we want to determine the effect of varying only the water soluble part.

METHODOLOGY

The *water-soluble* part of aerosol particles originates from gas to particle conversion and consists of various kinds of sulfates, nitrates, and other, also organic, water-soluble substances. 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. *Sea-salt* particles consist of the various kinds of salt contained in seawater. Two sea-salt modes are given as *sea salt accumulation* and *sea salt coarse* modes (Koepke et al. 1997).

The equilibrium water vapor saturation ratio S as described by Kohler theory is given as

$$S = a_w K_e \tag{1}$$

where a_w denotes the water activity or Raoult term, and K_e is the Kelvin effect.

The relationship between droplet radius and RH at equilibrium can also be given as:

$$S = a_w \exp\left(\frac{2\sigma v_w}{RT r(S)}\right) \tag{2}$$

where v_w is the partial molar volume of water, σ is the surface tension of the solution at the composition of the droplet, R is the universal gas constant, T is the temperature and $r(S)$ is the equilibrium radius.

For single-solute particles, the equilibrium water vapor saturation ratio S for a droplet can be described by (Mochida et al., 2006):

$$\ln S = \frac{A}{r(S)} - \frac{B r^3(S=0)}{r^3(S) - r^3(S=0)} \tag{3}$$

where $A = \frac{2\sigma v_w}{RT}$ and $B = v\phi \frac{v_w}{v_s}$ are assumed to be constant, $r(S)$ and $r(S=0)$ are the effective radii of the wet and dry solute, respectively, S is taken for seven values 50%, 70%, 80%, 90%, 95%, 98% and 99%. σ is the surface tension of the solution, v_w and v_s are the molar volumes of pure water and solute, respectively, v is the degree of dissociation, and ϕ is

the osmotic coefficient. The product of v and ϕ is equivalent to the so-called van't Hoff factor (Pruppacher and Klett, 1997), R and T are the gas constant and temperature, respectively.

But atmospheric aerosols usually comprised mixtures of soluble and insoluble 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:

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

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

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

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

For multi components, equation (3) can be written to represents the property of the bulk components using equations (4) and (5) as:

$$\ln S = \frac{A}{r_{eff}(S)} + \frac{B}{1 - g_{eff}^3(S)} \tag{6}$$

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

The first term on the right hand side of equation (6) can replace the first term of equation (3) as

$$\ln K_e = \frac{2\sigma v_w}{RT r_{eff}(S)} = \frac{r_k}{r_{eff}(S)} = g_\sigma(r_{eff}(S)) \tag{7a}$$

This implies

$$K_e = \exp(g_\sigma(r_{eff}(S))) \tag{7b}$$

where a characteristic length for the effect of surface tension on the mixture or the effective Kelvin radius

$$r_k = \frac{2v_w \sigma}{RT} = A \tag{7c}$$

and

$$g_\sigma(r_{eff}(S)) = \frac{r_k}{r_{eff}(S)} = \frac{A}{r_{eff}(S)}.$$

The second term on the right hand side of equation (6) can also replace the second term of equation (3) as

$$\ln a_w = \frac{B}{1 - g_{eff}^3(S)} \tag{8a}$$

The lowest correction on the effective hygroscopic growth due to the Kelvin effect was also obtained by Lewis (2006) as:

$$\Delta g_{eff}(S, g_\sigma(r_{eff})) = - \frac{r_k}{r_{eff}(0,0) g_{eff}(S,0)} \frac{S}{dS} \frac{dg_{eff}(S,0)}{dS} \tag{11}$$

Similarly, the lowest-order correction to the effective radii due to the Kelvin effect was determined by Lewis (2006) as:

$$\Delta r_{eff}(S, g_a) = -r_k \frac{S}{g_{eff}(S, 0)} \frac{dg_{eff}(S, 0)}{dS} \quad (12)$$

The data extracted from OPAC were then applied to two models equations to determine the effect of water solubles on the Kelvin effects of the models. The first model is a one-parameter power law, the empirical γ -model that is frequently used in literature and researchers (Swietlicki et al., 2000; Birmili et al., 2004; Kasten, 1969; Gysel et al., 2009; Putaud, 2012; Tijjani, 2013a; Tijjani, and Uba, 2013a; Tijjani and Uba, 2013b, Tijjani, 2013b; Tijjani et al., 2013) to describe the hygroscopic growth of atmospheric aerosol particles as

$$g_{eff}(S, 0) = (1 - S)^{-n} \quad (13)$$

The exponent n was determined using regression analysis with SPSS 16.0.

The errors due to Kelvin effect on equation (13) were determined by substituting this equation into equations (11) and (12), Lewis (2006) as:

$$\Delta g_{eff}(S, g_a(r_{eff})) = -\frac{r_k}{r_{eff}(0)} \frac{nS}{1 - S} \quad (14)$$

and

$$\Delta r_{eff}(S, g_a(r_{eff})) = -r_k \frac{nS}{1 - S} \quad (15)$$

The fractional changes in the effective hygroscopic growth and effective radii due to the Kelvin effect error from equations (14) and (15) were determined as

$$\frac{\Delta g_{eff}(S, g_a(r_{eff}))}{g_{eff}(S, 0)} = -\frac{r_k}{r_{eff}(0)} \frac{nS}{(1 - S)^{1-n}} \quad (16)$$

and

$$\frac{\Delta r_{eff}(S, g_a)}{r_{eff}(S, 0)} = -\frac{r_k}{r_{eff}(S)} \frac{nS}{1 - S} \quad (17)$$

The second model is the relation between $g_{eff}(S, 0)$ and S that has been parameterized in a good approximation by a one-parameter equation, proposed e.g. by Petters and Kreidenweis (2007) and used by some researchers (Tijjani, 2013a; Tijjani, and

Uba, 2013a; Tijjani and Uba, 2013b, Tijjani, 2013b; Tijjani et al., 2013) as:

$$g_{eff}(S, 0) = \left(1 + k \frac{S}{1 - S}\right)^{\frac{1}{3}} \quad (18)$$

The values of k were determined using regression analysis using SPSS 16.0. The coefficient k is defined as the effective tendency for the ensembles to absorb water (Petters and Kreidenweis, 2007; Sullivan et al., 2007). It accounts not only for the reduction of water activity by the solute but also for surface tension effects (Rose et al., 2008; Gunthe et al., 2009) and depends on the molar volume and the activity coefficients of the dissolved compounds (Christensen and Petters, 2012).

The errors due to Kelvin effects on equation (18) were determined by substituting this equation into equations (11) and (12) as:

$$\Delta g_{eff}(S, g_a(r_{eff})) = -\frac{Sk}{3(1 - S)^2} \frac{r_k}{r_{eff}(0, 0)} \left(1 + \frac{kS}{1 - S}\right)^{-1} \quad (19)$$

and

$$\Delta r_{eff}(S, g_a) = -\frac{Sk}{3(1 - S)^2} \frac{r_k}{r_{eff}(0, 0)} \left(1 + \frac{kS}{1 - S}\right)^{-1} \quad (20)$$

The fractional changes in the effective hygroscopic growth and effective radii due to the Kelvin effect error from equations (19) and (20) were determined as

$$\frac{\Delta g_{eff}(S, g_a(r_{eff}))}{g_{eff}(S, 0)} = -\frac{Sk}{3(1 - S)^2} \frac{r_k}{r_{eff}(0, 0)} \left(1 + \frac{kS}{1 - S}\right)^{-4/3} \quad (21)$$

$$\frac{\Delta r_{eff}(S, g_a)}{r_{eff}(S, 0)} = -\frac{Sk r_k}{3 r_{eff}(S, 0) (1 - S)^2} \left(1 + \frac{kS}{1 - S}\right)^{-1} \quad (22)$$

The fractional changes in the equilibrium pressure $S(g_{eff}, g_a(r_{eff}))$ due to the Kelvin effect error on equation (9) were determined as

$$\frac{\Delta S(g_{eff}, g_a(r_{eff}))}{S(g_{eff}(S, 0), 0)} = \frac{S(g_{eff}, g_a(r_{eff}))}{S(g_{eff}(S, 0), 0)} g_a(r_{eff}) \quad (23)$$

where $S(g_{eff}(S, 0), 0)$ is the ambient RH.

RESULTS AND DISCUSSIONS

The models extracted from OPAC are given in Tables 1 and 2.

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

Components	ModelA Con.(cm ⁻³)	ModelB Con.(cm ⁻³)	ModelC Con.(cm ⁻³)	ModelD Con.(cm ⁻³)	ModelE Con.(cm ⁻³)
water soluble	3500.0000	4000.0000	4500.0000	5000.0000	5500.0000
Soot	5000.0000	5000.0000	5000.0000	5000.0000	5000.0000
sea salt (acc)	20.0000	20.0000	20.0000	20.0000	20.0000
sea salt (coa)	0.0032	0.0032	0.0032	0.0032	0.0032

Table2 :Microphysical properties of aerosol components at 0% RH (Hess et al., 1998).

Components	R _{min} (μm)	R _{max} (μm)	Sigma	R _{mod} (μm)
water soluble	0.0050	20.0000	2.2400	0.0212
Soot	0.0050	20.0000	2.0000	0.0118
sea salt (acc)	0.0050	20.0000	2.0300	0.2090
sea salt (coa)	0.0050	60.0000	2.0300	1.7500

Table 3: The results of the regression analysis of equation (6), the effective radii of the aerosols at 0% RH using equation (5).

equation (6)				
	R ²	A=Kelvin radii(μm)	B=Bulk hygroscopicity factor	Effective radii at 0% RH (μm)
ModelA	0.9901	0.048415	2.012913	0.556633
ModelB	0.9902	0.047928	1.986528	0.550215
ModelC	0.9903	0.047482	1.961530	0.544083
ModelD	0.9904	0.047038	1.937194	0.538194
ModelE	0.9905	0.046610	1.913666	0.532565

From Table 3, by observing the values of R², it can be seen that the data fitted the model very well. It can also be observe that the Kelvin radii and Bulk hygroscopicity factors decrease with the increase in the water soluble concentrations. This is because by looking at equation (3), it can be observe that the

Kelvin radii is proportional to the surface tension and this implies decrease in surface tension, while for B, it can be observe that it is inversely proportional to the volume of the solute. It can also be observe that the effective radii at 0% RH decreases with the increase in water soluble.

Table 4: The results of the regression of equation (13), using the ambient RHs and the water activity.

Models	R ²	N
ModelA	0.9883	0.3502
ModelB	0.9886	0.3493
ModelC	0.9889	0.3484
ModelD	0.9891	0.3475
ModelE	0.9894	0.3466

From Table 4, by observing the values of R², it can be seen that the data fitted the model very well. By observing the values of N, it can be seen that it decreases with the increase in water soluble

concentrations. This signifies inverse relation between the effective growth, g_{eff} and the equilibrium water saturation ratio, S, (1-S).

Table 5: The results of the regression of equation (18), using the ambient RHs the water activity.

Equation (18)	R ²	K
ModelA	0.9983	1.0329
ModelB	0.9983	1.0246
ModelC	0.9984	1.0164
ModelD	0.9984	1.0084
ModelE	0.9985	1.0005

From Table 5, by observing the values of R², it can be seen that the data fitted the model very well. It can also be seen that the values of k decrease with the

increase in water soluble. This signifies increase in the effective hygroscopic growth with water soluble.

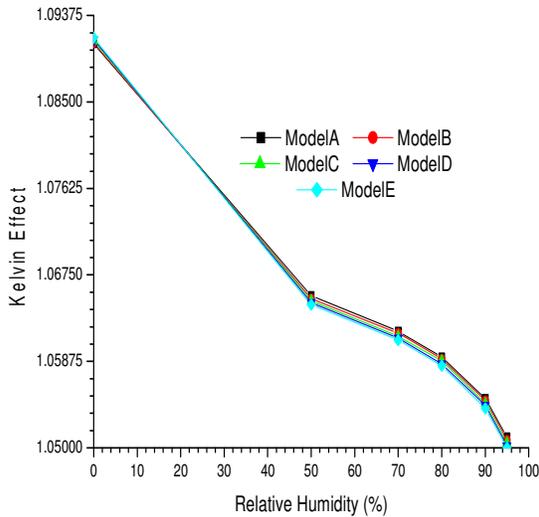


Figure 1: A plot of Kelvin effect of the aerosols against Relative Humidity using equation (7b).
 From Figure 1, it can be seen that kelvin effect decreases with the increase in RH. It also decreases with the increase in the water soluble concentrations most notably at higher RHs.

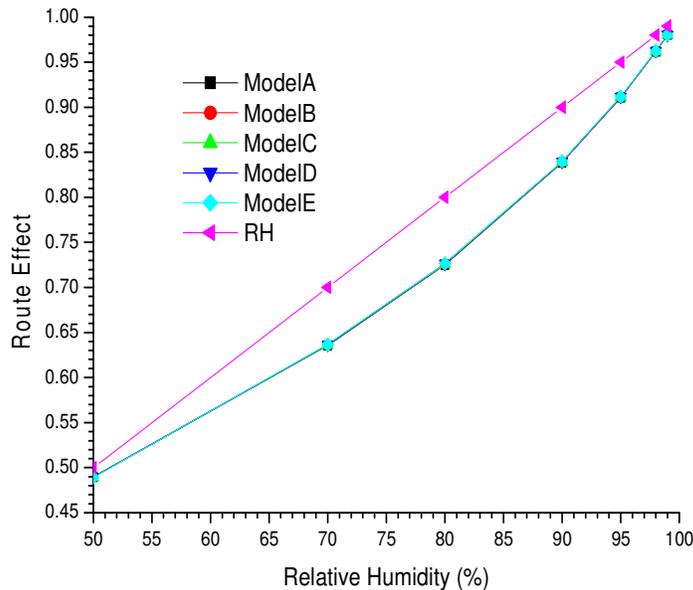


Figure 2: A plot of Route effect (water activity) of the aerosols against Relative Humidity using equation (8b).

From Figure 2, it can be seen that increase RH caused increase in water activity and increase in water soluble concentration has little effect on the Route effect. It increases with the increase in RH in almost quadratic

form. It is also less than the ambient RH. This also signifies the importance of water activity in the ambient RH, most especially at intermediate RH.

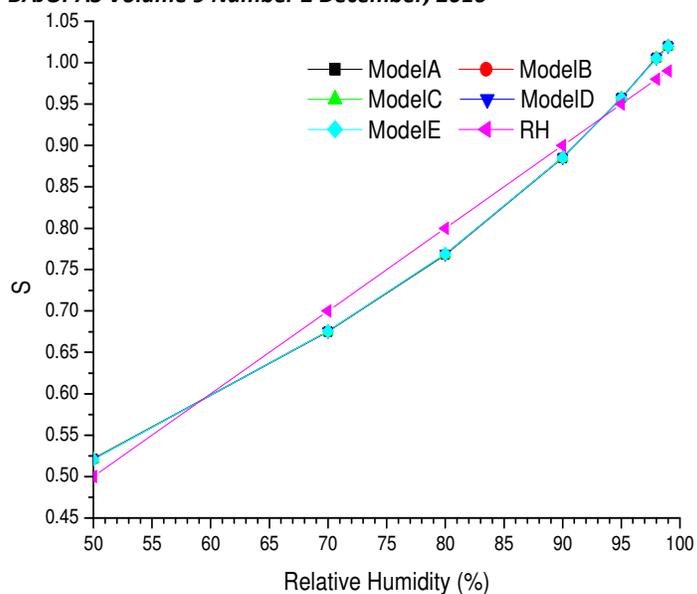


Figure3: A plot of $S(RH)$ of the aerosols against Relative Humidity using equation (9).

From Figure 3, comparing Figure 3 with Figures 1 and 2, it can be seen that at RHs 50, 95, 98 and 99 Kelvin effect dominated, but at RHs 70, 80, and 90, the

route effect dominated. It also shows the importance of water activity at the intermediate RH.

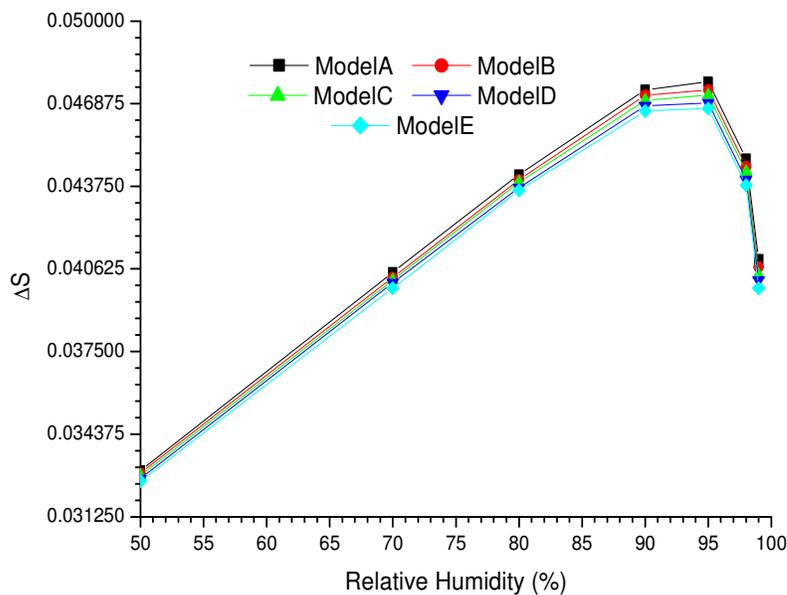


Figure4: A plot of ΔS of the aerosols against Relative Humidity using equation (10).

Figure 4, shows the amount by which the equilibrium relative humidity of a given model is underestimated due to Kelvin effect. From the figure, it can be seen that it decreases with the increase in water solublesand is very sensitive with the increase in the

ambient RH. It is in the range of between 0.033 to 0.05. This shows that the more dilute the solution is, the more the underestimation due to kelvin effect.

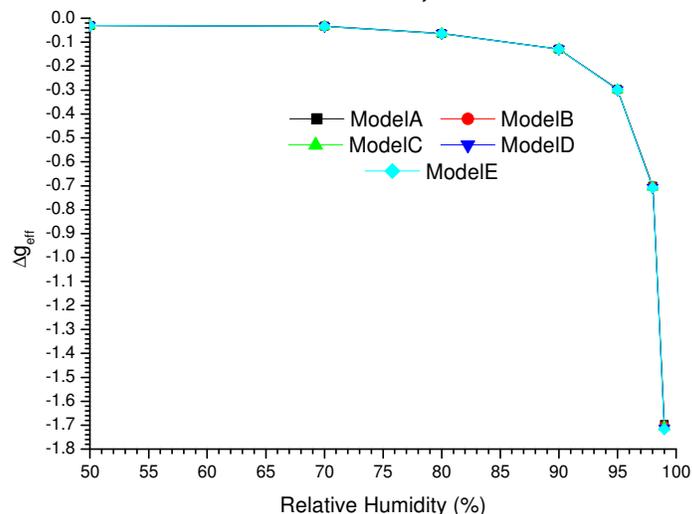


Figure 5: A plot of ΔG_{eff} of the aerosols against Relative Humidity using equation (11).

Figure 5 shows the amount by which the equilibrium effective hygroscopic growth of the models are overestimated by Kelvin effect. From the figure, it can be observe that it increases very little with the increase in the concentrations of the water solubles, but is very sensitive to RH most especially from the intermediate to higher RHs.

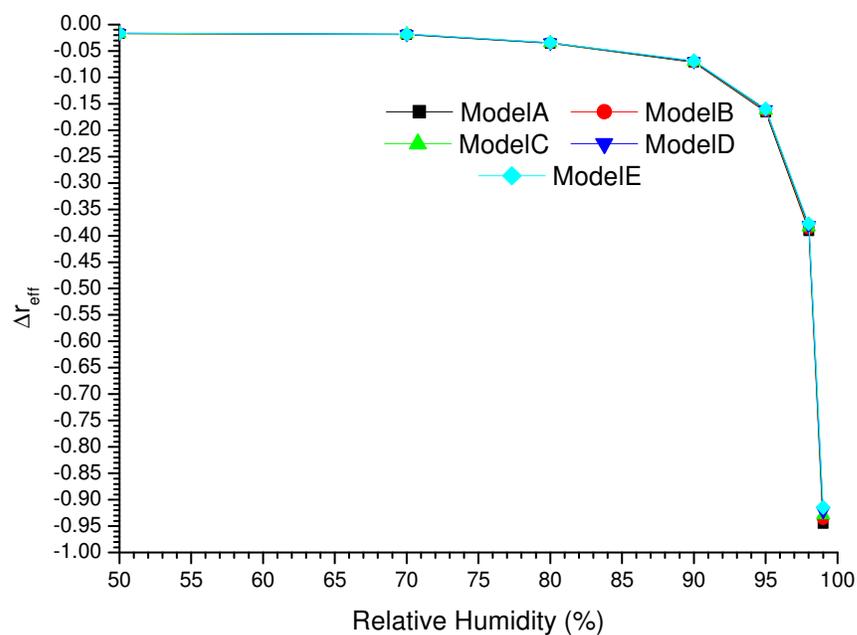


Figure 6: A plot of Δr_{eff} of the aerosols against Relative Humidity using equation (12).

Figure 6 shows the amount by which the equilibrium effective radii of the models are overestimated due to Kelvin effect. From the plots, it can be seen that it decreases very little with the increase in the concentration of the water solubles, but is very sensitive to RH most especially from the intermediate to higher RHs.

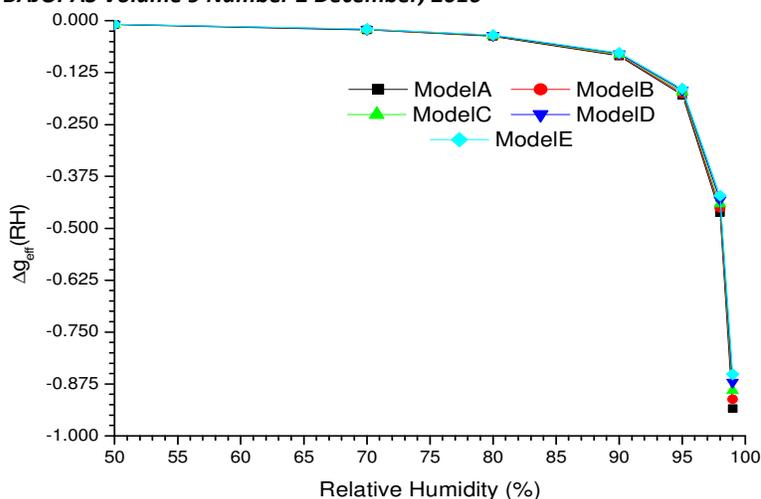


Figure 7: A plot of $\Delta g_{eff}(RH)$ of the aerosols against Relative Humidity using equation (14).

Figure 7 shows the amount by which the equilibrium effective hygroscopic growth of a given model is overestimated by Kelvin effect for our models of equation (13). From the figure it can be seen that the overestimation decreased with the increase in the water soluble concentrations most especially at higher RH.

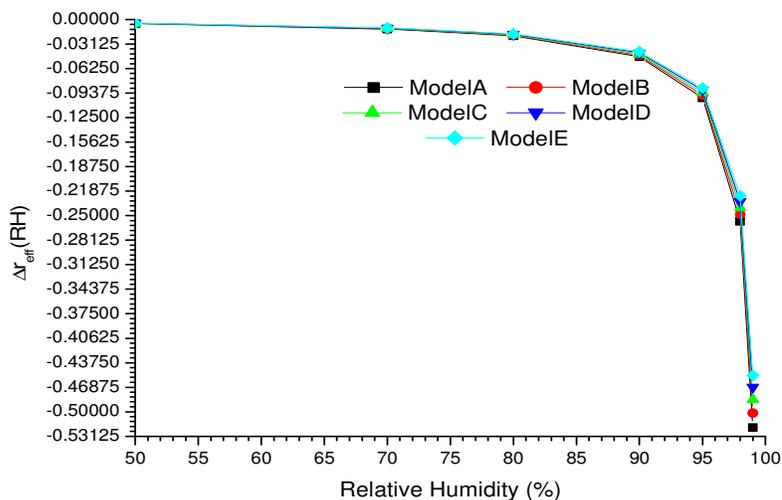


Figure 8: A plot of $\Delta r_{eff}(RH)$ of the aerosols against Relative Humidity using equation (15).

Figure 8 shows the amount by which the equilibrium effective radii of the various models are overestimated when Kelvin effect is neglected from equation (13). From the figure it can be seen that the overestimation decreases with the increase in the concentrations of water soluble and is more important at higher RHs.

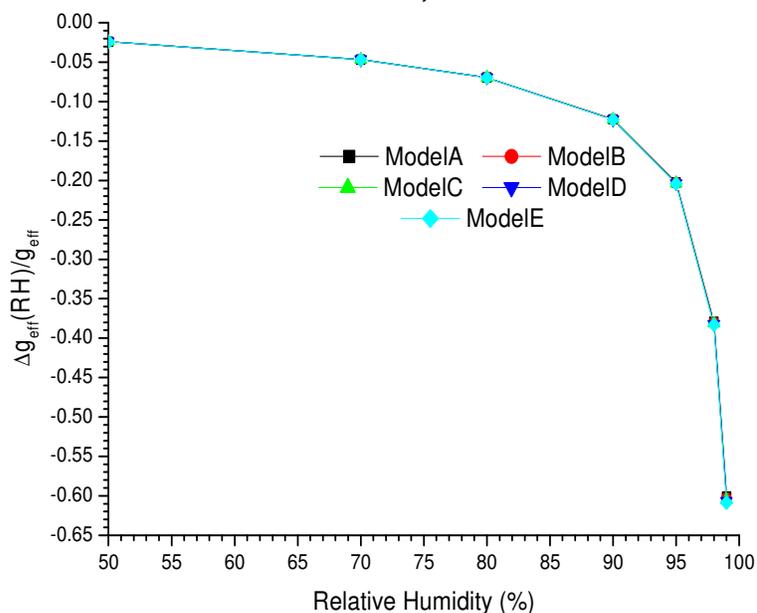


Figure9: A plot of $\Delta g_{eff}(RH)/g_{eff}$ of the aerosols against Relative Humidity using equation (16).

Figure 9 shows the plots of the fractional change in the effective hygroscopic growth when Kelvin effect is neglected from equation (13). From the figure it can

be seen that the fractional overestimation increases with the increase in water soluble concentration most especially at higher RHs.

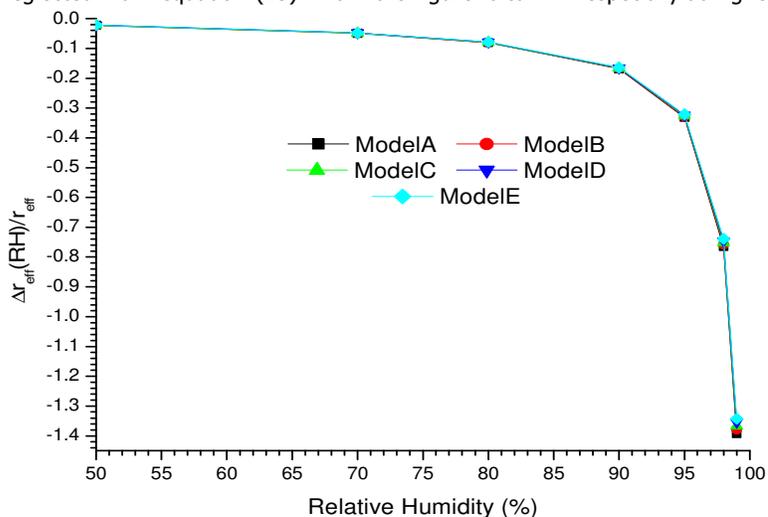


Figure10: A plot of $\Delta r_{eff}(RH)/r_{eff}$ of the aerosols against Relative Humidity using equation (17).

Figure 10 shows the plots of the fractional change in the effective radius when Kelvin effect is neglected from equation (13). From the figure it can be seen

that the fractional overestimation decreases with the increase in water soluble concentration most especially at higher RHs.

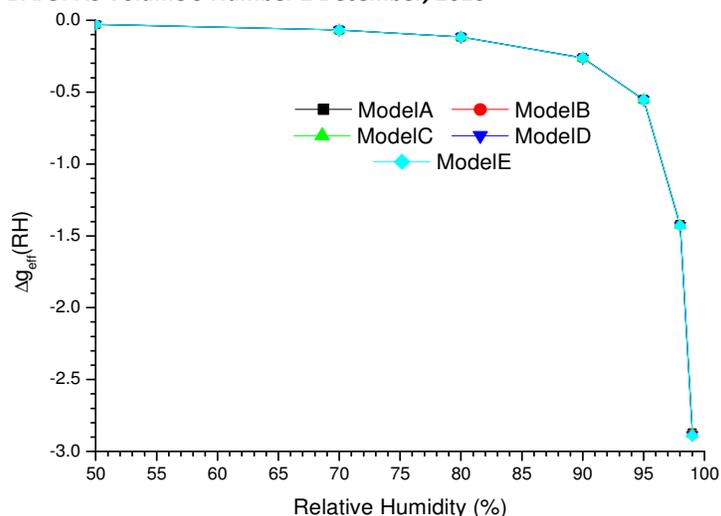


Figure11: A plot of $\Delta g_{eff}(RH)$ of the aerosols against Relative Humidity using equation (19).

Figure 11 shows the amount by which the equilibrium effective hygroscopic growth of a given mixture of aerosols is overestimated when Kelvin effect is neglected from equation (18). From the figure it can

be seen that the error due to the Kelvin effect increases with the increase in water soluble concentration most especially at higher RHs.

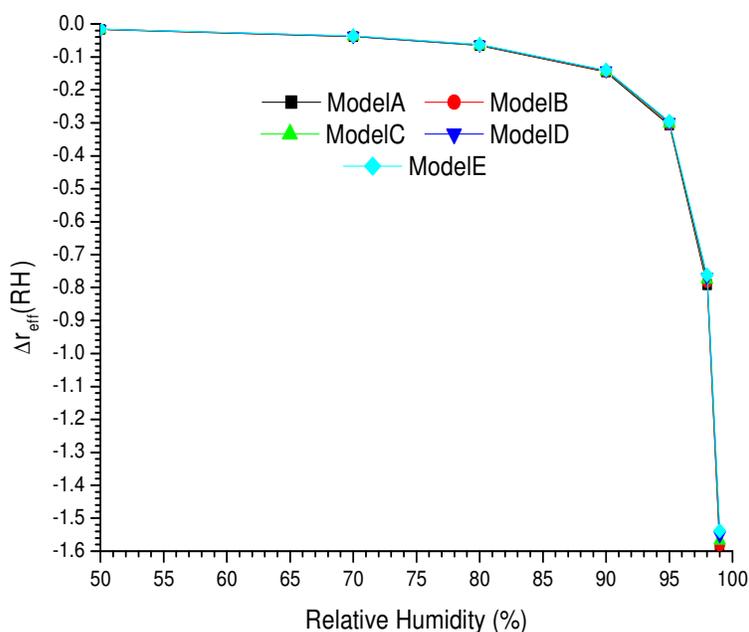


Figure12: A plot of $\Delta r_{eff}(RH)$ of the aerosols against Relative Humidity using equation (20).

Figure 12 shows the amount by which the equilibrium effective radius of a given mixture of aerosols is overestimated when Kelvin effect is neglected from

equation (18). From the figure it can be seen that the error decreases with the increase in the concentration of water soluble most especially at higher RHs.

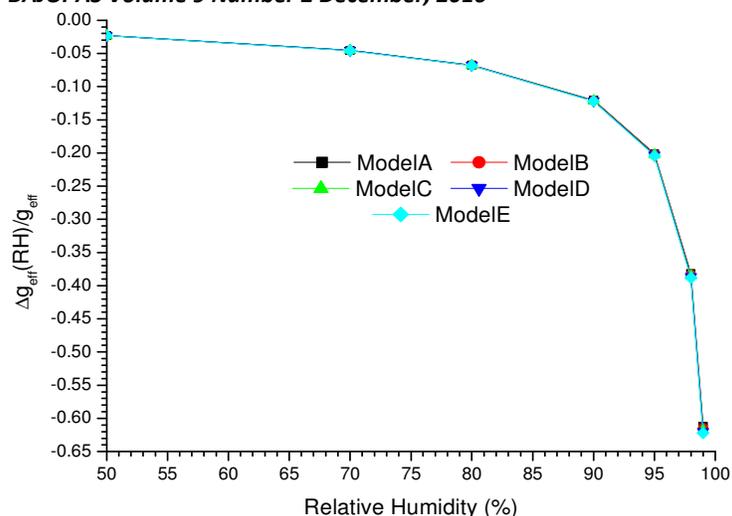


Figure13: A plot of $\Delta g_{eff}(RH)/g_{eff}$ of the aerosols against Relative Humidity using equation (21).

Figure 13 shows the amount by which the equilibrium fractional effective hygroscopic growth of are overestimated when Kelvin effect is neglected from equation (18). From the figure it can be seen that the error increases with the increase in the concentration of water solubles most especially at the higher RHs.

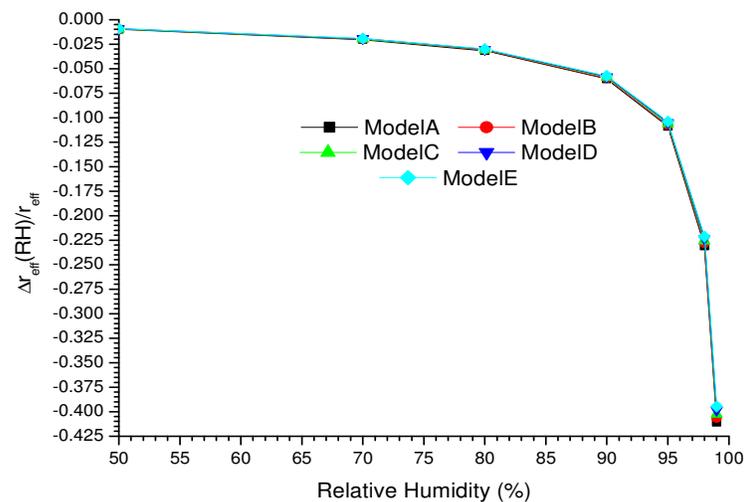


Figure14: A plot of $\Delta r_{eff}(RH)/r_{eff}$ of the aerosols against Relative Humidity using equation (22).

Figure 14 shows the amount by which the equilibrium fractional effective radius of a given mixture of aerosols is overestimated when Kelvin effect is neglected from equation (18). From the figure it can be observed that it decreases with the increase in the concentration of water solubles, most especially at higher RHs.

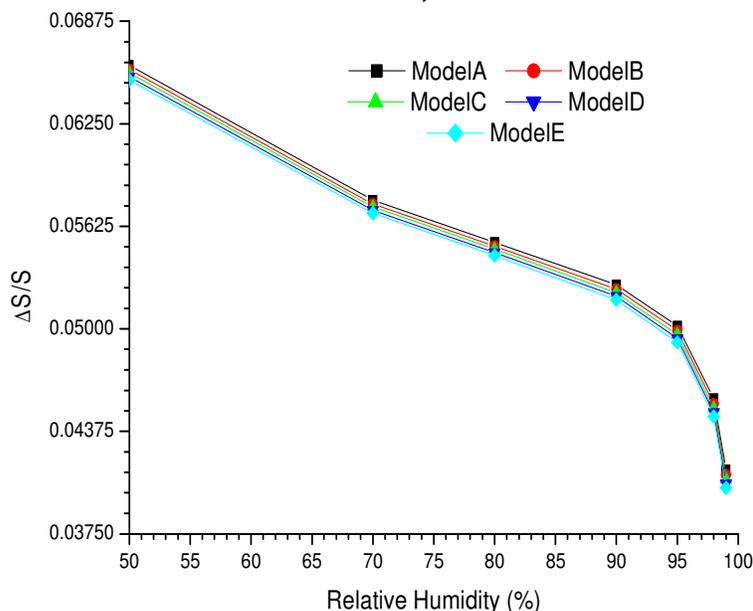


Figure 15: A plot of $\Delta S/S$ of the aerosols against Relative Humidity using equation (23).

Figure 15 shows the plots of the fractional change in the ambient RH due to the Kelvin effect from equation (9). From the figure it can be seen that the error decreases with the increase in the concentration of water soluble.

CONCLUSION

From the observations of the results obtained it can be concluded that increase in the concentrations of water soluble in aerosols has a great impact on its

effective radii, effective hygroscopic growth, in the ambient RHs, and their corresponding error due to the Kelvin effects.

However, there are some researchers that reported an erroneous implementation of the hygroscopic growth within OPAC, especially at intermediate RH ranges with the exception of Saharan dust (Zieger et al., 2013). This can probably be attributed to the behavior of Figure 2 of equation (8).

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