Single-parameter estimates of aerosol water content

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Abstract
Water can represent a substantial fraction of the mass of tropospheric non-cloud particulate matter, and can also serve as a medium for aqueous-phase reactions in such particles. Aerosol water contents are highly dependent upon aerosol hygroscopicity and ambient relative humidities (RH). In this work we evaluate a recently proposed parameterization of composition-dependent aerosol hygroscopicity that predicts the volume of liquid water associated with a unit volume of dry aerosol. The predictions over the range 10% < RH < 99% are compared with those from a rigorous thermodynamic model and shown to predict water volumes within ∼20% for the high water contents (RH > 85%) expected to have the most significant effects on tropospheric chemistry and radiation balance. Water contents for most of the compounds studied are generally represented within experimental uncertainties over the entire range of relative humidity examined, with the exception of marine-type particles dominated by sodium chloride and sodium sulfate.

Keywords: aerosol water content, aerosol hygroscopicity, growth factor

1. Introduction
It is well known that atmospheric particulate matter has important influences on tropospheric chemistry, human health, visibility, and direct and indirect climate forcing. Liquid water can represent a substantial fraction of the tropospheric aerosol volume, even for weakly hygroscopic particles, particularly at ambient relative humidities higher than ∼90%. The additional, and variable, particulate mass that water represents acts to reduce visibility and increase the mass extinction efficiencies of tropospheric aerosol components, and modifies the reflectivity and optical depth of aerosol layers. The high liquid water contents of clouds and fogs enable them to play important roles in tropospheric chemistry, particularly in S(IV)-to-S(VI) conversion processes and possibly in production of secondary organic aerosol (Gelencser and Varga 2005, Ervens and Kreidenweis 2007). Although condensed-phase water mass concentrations at subsaturated relative humidities are much lower than those found in clouds and fogs, and depend strongly on the mass concentrations of hygroscopic dry particulate matter, partitioning of trace species to the aerosol phase and reactions occurring in or on particulate matter may have significant impacts. For example, Arellanes et al (2006) showed that hydrogen peroxide was present in aerosol liquid water in much higher concentrations than expected from Henry’s Law, and that the measured concentrations far exceeded those known to cause damage to lung cells. Relative humidity can have a strong diurnal variation, and varies with synoptic weather patterns. Dry aerosol mass concentrations and compositions also respond to sources, sinks, and transport. Thus the aerosol water mass concentrations and their associated impacts can also be highly variable in time and space.

The water contents associated with various identified components of the atmospheric aerosol have been measured in the laboratory and, particularly for ionic species, can be well modeled using available thermodynamic models and codes (e.g. Wexler and Clegg 2002, e.g. Topping et al 2005a, 2005b). Many measurement campaigns over the past several decades have quantified the water uptake of complex atmospheric aerosols, up to about 95% relative humidity. Water contents are often measured as diameter growth factors (DGF), that is, the ratio of the particle diameter in a humidified environment...
Table 1. Assumed properties of each dry compound.

| Compound               | Molecular weight | Density (g cm\(^{-3}\)) | DRH, % (298.15 K) | \(\kappa\) | References (DRH values) |
|------------------------|------------------|---------------------------|-------------------|---------|------------------------|
| NaCl                   | 58.44            | 2.165                     | 75.3              | 1.27    | Marcoli et al (2004)    |
| \((\text{NH}_4)_2\text{SO}_4\) | 132.14          | 1.769                     | 80.2              | 0.50    | Marcoli et al (2004)    |
| \((\text{NH}_4)_2\text{H} (\text{SO}_4)_2\) | 247.25          | 1.83                      | 69.0              | 0.56    | Tang and Munkelwitz (1994) |
| \(\text{NH}_4\text{HSO}_4\) | 115.11          | 1.78                      | 40.0              | 0.56    | Tang and Munkelwitz (1994) |
| \(\text{Na}_2\text{SO}_4\) | 142.04          | 2.68                      | 82.4              | 0.74    | Tang and Munkelwitz (1993) |
| \((\text{HOOC})_2\text{CH}_3(\text{COOH})\) | 132.11          | 1.41                      | 95.5              | 0.15    | Marcoli et al (2004)    |
| \((\text{HOOC})_–(\text{COOH})\) | 90.03           | 1.9                       | 97.8              | 0.48    | Marcoli et al (2004)    |



\[(D_{\text{wet}}) \text{ to the diameter at dry conditions (}D_{\text{dry}}):\]

\[
\text{DGF}(\text{RH}) = \frac{D_{\text{wet}}}{D_{\text{dry}}} = \left(\frac{V_{\text{wet}}}{V_{\text{dry}}}\right)^\frac{1}{d} = \left(\frac{V_{\text{w}} + V_s}{V_s}\right)^\frac{1}{d}
\]

where \(V\) refers to particle volume, \(V_w\) to the volume of water in the particle, \(V_s\) to the volume of the dry particle, and it has been assumed that the volumes of water and dry particulate matter are additive. Observations in a variety of air mass types (Swietlicki et al 2008; Massling et al 2007) have shown that DGFs at 85–90% RH range from close to 1 (indicating a nonhygroscopic aerosol) to over 2, the highest values being observed for some marine particles dominated by sea salt.

The objective of this work is to examine the predictions of simple, computationally efficient one-parameter representations of aerosol water content as a function of RH (Rissler et al 2006, Petters and Kreidenweis 2007). To date, these parameterizations have primarily been used to successfully predict the critical supersaturations at which dry particles of specified size and composition activate to cloud drops, using subsaturated growth factors measured at RH ~90% (e.g. Carrico et al 2008, Chan et al 2008, Vestin et al 2007). They thus appear useful for incorporation into large-scale models that simulate aerosol activation to cloud drops, where computational speed is an important consideration; they could also be used in such models to compute aerosol water contents in the subsaturated regime, so that the treatment of aerosol hygroscopicity would be consistent throughout the simulated atmosphere. However, predicted aerosol water contents have not heretofore been evaluated at RH < 90%. Here we estimate the error in water content over the range 10% < RH < 99% by comparing one-parameter predictions with those from a validated, rigorous thermodynamic model that simulates solution water activities.

2. Methods

The composition-dependent water activities of several aqueous solutions of atmospheric interest were computed using the online version of the Aerosol Inorganic Model (AIM; Wexler and Clegg 2002; http://www.aim.env.uea.ac.uk/aim/aim.html). Model III (Clegg et al 1998), applicable at 298.15 K, was used for the inorganic species sodium chloride (NaCl), ammonium sulfate (\((\text{NH}_4)_2\text{SO}_4\)), letovicite (\((\text{NH}_4)_3\text{H} (\text{SO}_4)_2\)), ammonium bisulfate (\((\text{NH}_4)\text{HSO}_4\)), ammonium nitrate (\((\text{NH}_4)\text{NO}_3\)), and sodium sulfate (\((\text{Na}_2\text{SO}_4)\)). For the two dicarboxylic acids considered, methylsuccinic acid ((\(\text{HOOC})_2\text{CH}_3(\text{COOH})\)) and oxalic acid ((\(\text{HOOC})(\text{COOH})\)), the available online model following Clegg and Seinfeld (2006), including dissociation equilibria, was used. The model outputs are shown in figure 1 (for clarity, we omitted calculations for letovicite and ammonium bisulfate in this figure), where solution composition has been expressed as the volume of water (\(V_w\)) per unit volume dry compound (\(V_s\)).

![Figure 1. Dotted lines: aerosol water contents for the indicated metastable binary solutions as a function of solution water activity (fractional relative humidity), computed using AIM (Wexler and Clegg 2002) and expressed as the ratio of volume of water (\(V_w\)) per volume of dry compound (\(V_s\)). Solid lines: values above the deliquescence RH at 298.15 K (table 1).](image-url)
was suppressed, so that the water contents represent those for metastable solutions. This choice is intended to represent behavior of aqueous solution particles, which have been observed to retain water to low ambient relative humidities beyond known deliquescence points (e.g., Rood et al. 1989, Tang et al. 1997). The reported deliquescence relative humidity for each compound, below which only metastable solutions exist, is also shown in table 1.

If the aerosol is assumed to be at equilibrium with its environment, and if the effects of curvature on vapor pressure over the particles are neglected, then the solution water activity is equal to the fractional ambient relative humidity. This approximation is good to within a few per cent for particles larger than about 100 nm and for subsaturated conditions (Topping et al. 2005a), and in this discussion we will use water activity and relative humidity interchangeably. For some compositions and at relative humidities approaching saturation, however, kinetic limitations to water uptake (Nenes et al. 2001, Mikhailov et al. 2004, Sjögren et al. 2007), interactions between solution and curvature effects (Sorjamaa et al. 2004), and water mass balance (Metzger and Lelieveld 2007) become important and this equality may not be valid. Here, calculations are restricted to the range 0.1 < $a_w < 0.99$ for these reasons. Figure 1 demonstrates that the volume of the total aerosol that is attributable to water reaches 50% ($V_w/V_s = 1$) well before 90% RH, and that water is the dominant aerosol component at 95% RH ($V_w/V_s > 10$), except for the organic species methylsuccinic acid.

Recently, Petters and Kreidenweis (2007) proposed that the water activity of aqueous atmospheric particulate matter be represented by the functional form,

$$a_w^{-1} = 1 + \kappa \frac{V_s}{V_w}$$  \hspace{1cm} (2)

where $\kappa$ is an adjustable parameter, and was called the hygrosopicity parameter. Rearranging,

$$\frac{V_w}{V_s} = \kappa \frac{a_w}{1 - a_w}. \hspace{1cm} (3)$$

Equation (3) demonstrates that $\kappa$ serves as a scaling variable for the assumed functional dependence of aerosol volumetric water content on water activity, $a_w/(1 - a_w)$: hygroscopic material is characterized by large values of $\kappa$, whereas nonhygroscopic but wettable compounds correspond to $\kappa = 0$. Petters and Kreidenweis (2007) presented tabulated ranges of values of $\kappa$ for a variety of pure compounds and atmospheric aerosol types, computed by fitting laboratory-determined hygroscopic growth factors at $a_w \sim 0.9$, critical supersaturation-dry diameter data, and/or modeled solution water activity data to equation (2). In table 1 we summarize the values of $\kappa$ selected for each species in this work. Petters and Kreidenweis (2007) did not report $\kappa$ values for ammonium bisulfate, oxalic acid or methylsuccinic acid; the AIM water activity calculations shown in figure 1 were fit to equation (2) to obtain the values shown in table 1. Figure 2 shows the computed aerosol water contents for the parameterization over the range 0.1 < $a_w < 0.99$, compared with the AIM model predictions.

3. Discussion

From figure 2 it is apparent that the parameterization is not able to fit well the aerosol water contents over the full range of relative humidity considered, and deviates particularly at low RH. The percentages difference between parameterization- and AIM-derived water contents (100 × $[(V_w/V_s)_{\text{aim}} - (V_w/V_s)_{\text{aim}}]/(V_w/V_s)_{\text{aim}}$) at several RH values are summarized in table 2. Above about 85% RH, the two

![Figure 2](image-url)
Various water activities. 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Alternative dashed and dotted lines indicate the ranges in water activity corresponding to the derived water contents. We have also assumed that organic species contribute between ~20 and 70% of the hygroscopic mass concentration, guided by the aerosol mass spectrometer (AMS) observations reported in Zhang et al (2007) from a range of locations around the globe. Further stratified the organic (OC) mass fraction into hydrocarbon-like (HOA) and oxygenated (OOA) components, with relative contributions from 60%/40% to 0%/100% of the total OC. To develop the models in table 3, we assigned zero hygroscopicity for the ammonium sulfate and oxygenated (OOA) components, but are intended as reasonable representations of variations in aerosol composition that might affect total aerosol water contents, and also allow us to compute aerosol water contents using AIM. Our choices of models are also based on obtaining a reasonable range of overall hygroscopicities that are similar to observations reported in the literature.

Most of the overall $\kappa$ values in table 3 correspond to the range $0.1 < \alpha_w < 0.9$, with the remainder attributed to organic carbon. In Model 2, the organic carbon is dominated by the $\kappa = 0$ component, whereas in Model 3, all of the organic carbon is assumed to be as hygroscopic as methylsulfonylic acid. Models 6, 9, and 10 have volume fractions of ammonium nitrate of 0.8, with the remainder attributed to organic carbon. This is partly due to the ratios ranging over 4 orders of magnitude, and is particularly accurate at high water contents (RH > 0.9). This is partly due to the choices of $\kappa$ for each individual compound. We have chosen $\kappa$ to emphasize agreement with AIM for dilute solutions with high $\alpha_w$, accepting poorer agreement in the metastable regimes. A choice of $\kappa$ that minimizes the error over a broader range of solution composition generally introduces larger errors at high water contents, which are most important for atmospheric applications. For Models 6, 9, and 10, representative of marine aerosol, the parameterization agrees well with AIM for RH > 0.9, but significantly underpredicts water contents at lower RH, as expected.

In figure 4 we present the relative differences in $V_w/V_s$ between AIM and the parameterization for the ten aerosol models as a function of aerosol water content. The gray bars are estimated uncertainties in DGF of ±0.5%. This is partly due to compensating effects of under- and over-prediction of various mixture components (see figure 2), but is primarily due to the choices of $\kappa$ for each individual compound. We have chosen $\kappa$ to emphasize agreement with AIM for dilute solutions with high $\alpha_w$, accepting poorer agreement in the metastable regimes (figure 2). A choice of $\kappa$ that minimizes the error over a broader range of solution composition generally introduces larger errors at high water contents, which are most important for atmospheric applications. For Models 6, 9, and 10, representative of marine aerosol, the parameterization agrees well with AIM for RH > 0.9, but significantly underpredicts water contents at lower RH, as expected.

Models 2 and 3 have volume fractions of ammonium nitrate of 0.8, with the remainder attributed to organic carbon. In Model 2, the organic carbon is dominated by the $\kappa = 0$ component, whereas in Model 3, all of the organic carbon is assumed to be as hygroscopic as methylsulfonylic acid. Both models have similar overall $\kappa$ and similar accuracies in $V_w/V_s$. This example demonstrates that, for particle compositions dominated by hygroscopic inorganic species, the exact treatment of the hygroscopicity of the organic fraction is of only minor importance.

In figure 4 we present the relative differences in $V_w/V_s$ between AIM and the parameterization for the ten aerosol models as a function of aerosol water content. The gray
bars are estimated maximum uncertainties in \( V_\text{w}/V_c \) based on Mikhailov et al (2004), who cited a sizing accuracy of 2% in diameter in their HTDMA instrument, which translates into an estimated ±4% uncertainty in DGF. DGF data are rarely obtained for RH greater than about 95% because of the high sensitivity of water content to slight RH variations in this range (figure 1). Interestingly, at low RH, the ±4% uncertainty in DGF translates into large relative uncertainty in \( V_\text{w}/V_c \). This occurs because DGF becomes very close to one, and thus the very small amount of water associated with the particle is poorly constrained experimentally. When compared with this experimental error, the errors in \( V_\text{w}/V_c \) from the parameterization are within the uncertainty estimate, except for the models representative of marine aerosols and for models 1–3 over small ranges of the domain.

4. Conclusions

We have evaluated the predictions from a simple, single-parameter representation of aerosol water contents against predictions from a rigorous thermodynamic model, over a wide range of stable and metastable inorganic and organic aqueous solution concentrations (0.1 < \( a_\text{w} < 0.99 \)). The fit parameters were specifically chosen for accuracy in the dilute regime, \( a_\text{w} > 0.9 \), because these compositions represent the largest aerosol water contents and are also used in predicting particle activity as a cloud condensation nucleus. We find significant errors, more than 20%, in predicted aerosol water contents for individual species for RH < 85%. When these species are combined into mixtures that are reasonable representations of observed atmospheric aerosols, however, we find that overall predicted water contents are within experimental errors over the full range of RH, except for marine-type aerosols, for which the parameterization significantly underpredicts water content for RH < 90%. We suggest that although a separate parameterization is needed to well represent marine aerosol water contents in other applications, including visibility and direct aerosol climate forcing, the single-parameter representation is satisfactory for predicting CCN activity of marine particles. For other particle types and for applications that require computational simplicity and speed, the results presented here can be used to evaluate whether the parameterization is sufficiently accurate.

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