

Technical note

Size distribution of sea-salt emissions as a function of relative humidity

K. Max Zhang^a, Eladio M. Knipping^d, Anthony S. Wexler^{a,b,c,*}, Prakash V. Bhawe^{e,1}, Gail S. Tonnesen^f

^aDepartment of Mechanical and Aeronautical Engineering, University of California, Davis, CA 95616, USA

^bDepartment of Civil and Environmental Engineering, University of California, Davis, CA 95616, USA

^cDepartment of Land, Air, and Water Resources, University of California, Davis, CA 95616, USA

^dElectric Power Research Institute, 3412 Hillview Ave., Palo Alto, CA 94304, USA

^eAtmospheric Sciences Modeling Division, Air Resources Laboratory, National Oceanic and Atmospheric Administration, Research Triangle Park, NC 27711, USA

^fCenter for Environmental Research and Technology, University of California, Riverside, CA 92521, USA

Received 5 August 2004; received in revised form 11 February 2005; accepted 25 February 2005

Abstract

This note presents a straightforward method to correct sea-salt-emission particle-size distributions according to local relative humidity. The proposed method covers a wide range of relative humidity (0.45–0.99) and its derivation incorporates recent laboratory results on sea-salt properties. The formulas are given as functions of relative humidity and compared with growth factors derived from earlier theoretical work. Application of this method to several common open-ocean and surf-zone sea-salt-particle source functions is described. The resulting sea-salt-emission aerosol distributions can be used directly in atmospheric model simulations without further correction.

© 2005 Elsevier Ltd. All rights reserved.

Keywords: Sea-salt; Emission; Size distribution; Size correction; Relative humidity; Atmospheric modeling; Aerosol thermodynamics

1. Introduction

Sea-salt particles may have profound impacts on urban air quality (Knipping and Dabdub, 2003), ecosystems of estuaries (Paerl et al., 2002; Evans et al., 2004), and global climate change (Laskin et al., 2003),

which make them essential components of atmospheric models at urban, regional and global scales (Gong et al., 1997). Sea-salt generation functions are usually given in the form of a continuous particle-size distribution at a specific relative humidity (RH), usually 80% (Monahan et al., 1986; Smith and Harrison, 1998) or at formation (de Leeuw et al., 2000). Given the hygroscopic nature of sea-salt electrolytes and the wide variability in the relative humidity within the planetary boundary layer, a size correction is needed to represent local conditions when calculating sea-salt emission rates. Traditionally, this correction is carried out inside a mathematical air quality model, i.e. sea-salt particle size distributions are adjusted according to the local relative humidity using

*Corresponding author. Department of Mechanical and Aeronautical Engineering, University of California, Davis, CA 95616, USA.

E-mail addresses: maxzhang@ucdavis.edu (K.M. Zhang), aswexler@ucdavis.edu (A.S. Wexler).

¹On assignment to the National Exposure Research Laboratory, US Environmental Protection Agency.

thermodynamic algorithms within the model at every time step. However, this method results in some deficiencies. For a model with a sectional aerosol representation (i.e., based on particle size bins), sea-salt emissions take a discrete form; the accuracy of the particle size correction will depend largely on how finely the particle size bins are divided and numerical errors will inevitably be introduced. This problem can be reconciled in a modal representation only if the adopted sea-salt emission function is also given by lognormal modes; otherwise, the same problem arises.

Earlier research has produced particle-growth formulas for a variety of particle types including sea salt (Fitzgerald, 1975; Hänel, 1976; Gerber, 1985). Application of these growth formulas to sea-salt emission generation functions is cumbersome because the former are functions of the dry particle size whereas the latter are functions of the particle size at 80% RH or at formation. Moreover, recent experimental evidence indicates that the properties of a “dry” sea-salt particle are not well-defined because water is present in sea-salt particles even at very low relative humidity (Tang et al., 1997). Considering the source of sea-salt particles in nature (namely, marine water bodies), it is intuitive to use the particle’s size at formation as the reference state rather than the dry particle size. Applying knowledge of sea-salt properties developed in recent years (Millero, 1996; Tang et al., 1997), we show that the particle size at the local RH relative to the particle size at formation can be expressed as a function of relative humidity alone. This yields a straightforward method to correct sea-salt-emission particle size distributions according to the local RH, thereby allowing the emissions to be used directly in air quality models without requiring further correction within the model.

2. Size adjustment with respect to RH

Sea-salt particle composition at formation resembles that of seawater, which has been well documented (Millero, 1996) as shown in Table 1. In the present study, seawater composition is approximated as 3.5% solute and 96.5% solvent on a mass basis (i.e., the solute weight fraction of seawater, x_{sw} , is 0.035). The solute weight fraction, x_{sw} , of seawater varies in a narrow range of 0.034 to 0.036; for atmospheric modeling purposes, the assumption above does not add significant uncertainty to aerosol calculations. The additional assumptions applied herein are: (1) thermodynamic equilibrium of water between sea-salt particles and the ambient environment (Andreas, 1992), (2) negligible effect of particle-surface curvature on water equilibrium (i.e., Kelvin effect) because sea-salt-particle diameters are usually above 0.1 μm , (3) conservation of solute within each particle, and (4) negligible dependence of the

Table 1
Composition of seawater (1 kg) with $\sim 19.4\%$ chlorinity (Millero, 1996)

| Species | Concentration (g) | Concentration (mol) |
|--------------------|-------------------|---------------------|
| Cl^- | 19.3529 | 0.54588 |
| Na^+ | 10.7838 | 0.46907 |
| Mg^{2+} | 1.2837 | 0.05282 |
| SO_4^{2-} | 2.7124 | 0.02824 |
| Ca^{2+} | 0.4121 | 0.01028 |
| K^+ | 0.3991 | 0.01021 |
| HCO_3^- | 0.107 | 0.00175 |
| Br^- | 0.0672 | 0.00084 |
| B(OH)_3 | 0.0194 | 0.00031 |
| CO_3^{2-} | 0.0161 | 0.00027 |
| Sr^{2+} | 0.0079 | 0.00009 |
| F^- | 0.0068 | 0.00068 |
| B(OH)_4^- | 0.001 | 0.0001 |
| OH^- | 0.0008 | 0.00008 |
| Overall | 35.172 | 0.56012 |

Table 2
Polynomial coefficients for water activity and density of seawater (Tang et al., 1997)

| | Water activity | | Density |
|-------|-------------------------|-------|------------------------|
| C_1 | -5.872×10^{-3} | A_1 | 7.93×10^{-3} |
| C_2 | 1.24×10^{-4} | A_2 | -4.28×10^{-5} |
| C_3 | -1.688×10^{-5} | A_3 | 2.52×10^{-6} |
| C_4 | 3.105×10^{-7} | A_4 | -2.35×10^{-8} |
| C_5 | -1.44×10^{-9} | A_5 | 0 |

thermodynamic properties of sea-salt electrolytes on ambient temperature. Note that water activity (a_w) is equal to RH under these assumptions.

Tang et al. (1997) used laboratory measurements of sea-salt particles to obtain a best-fit curve of water activity, a_w , in terms of solute weight fraction in natural sea-salt solutions, x , as $a_w = 1.0 + \sum_{i=1}^5 C_i (100x)^i$, where the values of C_i are given in Table 2. To represent x as a function of RH, we fit a polynomial to the inverse of Tang’s equation,

$$x(\text{RH}) = \sum_{i=0}^5 W_i \text{RH}^i, \quad 0.45 < \text{RH} < 0.99, \quad (1)$$

where W_i are polynomial coefficients provided in Table 3. Eq. (1) reproduces the results of Tang’s polynomial within 2% over the 0.45–0.99 RH range.

Fig. 1 depicts the relationship of solute weight fraction as a function of relative humidity as given by Eq. (1). We compare this result in Fig. 1 with similar curves obtained for pure sodium chloride, NaCl, as

Table 3
Polynomial coefficients for solute weight fraction, density and size correction

| i | W_i | D_i | S_i^0 |
|-----|---------|---------|---------|
| 0 | 3.1657 | 3.8033 | 28.376 |
| 1 | -19.079 | -16.248 | -205.44 |
| 2 | 55.72 | 46.085 | 653.37 |
| 3 | -83.998 | -68.317 | -1031.7 |
| 4 | 63.436 | 50.932 | 803.18 |
| 5 | -19.248 | -15.261 | -247.08 |

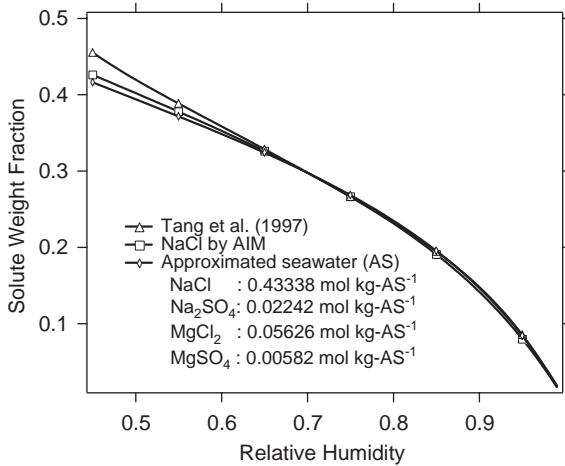


Fig. 1. Solute weight fraction of sea salt as a function of ambient relative humidity.

calculated using the aerosol inorganic model (AIM) (Wexler and Clegg, 2002; <http://mae.ucdavis.edu/wexler/aim>) and for an approximated sea salt (AS) as calculated using the standard Zdanovskii–Stokes–Robinson approach (Stokes and Robinson, 1966). The AS uses only the principal four ions in seawater—chloride, sodium, sulfate and magnesium—to construct an artificial solution similar to that of seawater given in Table 1. Chloride and sulfate are maintained at equal concentrations to that of seawater, whereas the molar concentrations of sodium and magnesium are increased slightly (proportionate to the molar ratio of potassium and calcium found in seawater) to maintain charge balance and an ionic strength similar to seawater. In order to use the standard ZSR approach, the distribution of the ions over the four possible salt combinations—NaCl, MgCl₂, Na₂SO₄ and MgSO₄—is determined using the mixing scheme of Clegg and Simonson (2001).

The curves in Fig. 1 for NaCl using the AIM and for the AS using the standard ZSR approach are fairly similar throughout the relative humidity range of

interest. These two curves diverge slightly from the curve calculated using (1) at low relative humidity (< 0.60 RH). In the case of the AIM results, this difference is due likely to the compositional difference between sea salt and pure sodium chloride. As the standard ZSR approach uses only single-solute data in its calculations, the divergence in the case of the AS is due, at least in part, to the inability of the method to account accurately for interactions among the electrolytes of multiple solutes, particularly in concentrated non-ideal solutions. Given that Eq. (1) is based on direct experimental observations, it is the preferred method for calculating the relation of solute weight fraction of sea salt as a function of relative humidity.

In sea-salt particles, the ratio of water mass at a particular RH to water mass at the point of formation, R_{H_2O} , may be expressed as

$$R_{H_2O} = \frac{M_{H_2O,RH}}{M_{H_2O,0}} = \frac{M_{H_2O,RH}/M_{solute}}{M_{H_2O,0}/M_{solute}} = \frac{1/x(RH) - 1}{1/x_{sw} - 1}, \quad (2)$$

where $M_{H_2O,0}$ and $M_{H_2O,RH}$ are the mass of water in a sea-salt particle at formation and at the local relative humidity, respectively, and M_{solute} is the mass of solute in the sea-salt particle.

The density of a sea-salt particle, ρ_p (kg m⁻³), may be expressed as a function of x using the empirical formula given by Tang et al. (1997):

$$\rho_p = 1000 \left[\left(0.9971 + \sum_{i=1}^5 A_i (100x)^i \right) \right], \quad (3)$$

where the values of A_i are listed in Table 2. Since x in Eq. (1) is expressed as a function of RH, combining Eqs. (1) and (3), ρ_p can be calculated also as a function of RH:

$$\rho_p(RH) = 1000 \sum_{i=0}^5 D_i RH^i, \quad 0.45 < RH < 0.99, \quad (4)$$

using the coefficients, D_i , listed in Table 3. Particle densities calculated using Eq. (4) match those calculated by Eq. (3) within 0.12% over the 0.45–0.99 RH range.

The mass of a spherical sea-salt particle is the product of its volume and density,

$$M_p = \frac{\pi}{6} \rho_p D_p^3. \quad (5)$$

This mass can be expressed also in terms of the particle mass at formation:

$$M_p = \frac{\pi}{6} \rho_{sw} D_{p0}^3 (R_{H_2O} (1 - x_{sw}) + x_{sw}), \quad (6)$$

where ρ_{sw} is the sea-water density (1.027×10^3 kg m⁻³) and D_{p0} is the diameter at formation. Equating Eqs. (5) and (6), we obtain a correction term that relates the

diameter at formation with the diameter at ambient RH.

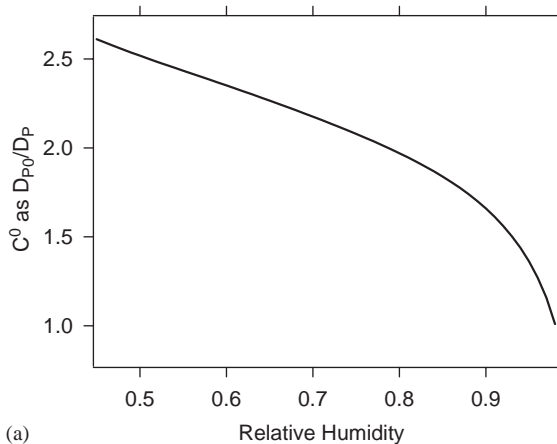
$$C^0 = \frac{D_{p0}}{D_p} = \left(\frac{\rho_p}{\rho_{sw}(RH_0(1 - x_{sw}) + x_{sw})} \right)^{1/3}. \quad (7)$$

Since both R_{H_2O} and ρ_p are functions of RH and x_{sw} is constant, C^0 is solely a function of RH. The curve of C^0 is depicted in Fig. 2a and fit to a polynomial as

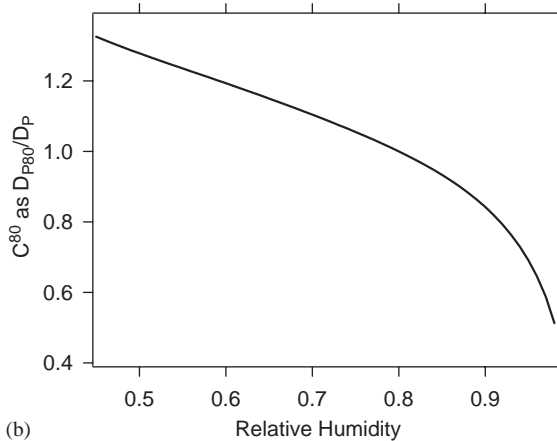
$$C^0(RH) = \sum_{i=0}^5 S_i^0 RH^i, \quad 0.45 < RH < 0.99 \quad (8)$$

using the coefficients listed in Table 3. Note that C^0 is equal to 1.97 at RH = 0.8, in good agreement with the value of ~ 2 reported by Fitzgerald (1975) and Andreas (1989). Therefore, we can express C^{80} , i.e., D_{p80}/D_p , as

$$C^{80}(RH) = C^0(RH)/1.97, \quad 0.45 < RH < 0.99. \quad (9)$$



(a)



(b)

Fig. 2. Particle-size correction factors as functions of ambient relative humidity with respect to the state at (a) formation and (b) 80% RH. Eqs. (8) and (9) reproduce those calculated indirectly from Tang's polynomial expressions within 2.8% over the 0.45–0.98 RH range, and within 8.6% over the 0.98–0.99 RH range.

C^0 and C^{80} should be relatively constant when RH is below the efflorescence point of sea salt ($\sim 45\%$ RH), but some residual water (5–10 wt%) is still found to be present in “solid” sea-salt particles at very low RH (Tang et al., 1997). Due to this uncertainty, we do not extend our formulas below 45% RH; nevertheless, the equation is valid for essentially all RH conditions expected over marine regions.

3. Comparison to other correction formulas

Particle-growth equations have been developed for the calculation of growth factors (GF), which are defined as a particle's size at a specified RH divided by the particle's size when it is dry (Fitzgerald, 1975; Hänel, 1976; Gerber, 1985). These growth equations were derived for a variety of electrolyte solutions in addition to seawater and account for the Kelvin effect. For reasons given above, they cannot be applied directly to adjust sea-salt emission distributions to the local RH. However, they can serve to corroborate the size-correction formulas derived in Section 2. After some algebraic manipulation, C^0 and C^{80} may be expressed as a ratio of GFs:

$$C^0 = \frac{D_{p0}}{D_p} = \frac{r_0/r_{dry}}{r_{RH}/r_{dry}} = \frac{GF(RH_0)}{GF(RH)}$$

and

$$C^{80} = \frac{D_{p80}}{D_p} = \frac{r_{80}/r_{dry}}{r_{RH}/r_{dry}} = \frac{GF(RH_{80})}{GF(RH)}. \quad (10)$$

In Eq. (10), r_{dry} , r_0 , and r_{RH} , are the dry particle radius, the radius at formation, and the radius at a specified relative humidity, respectively, and RH_0 is the relative humidity at formation. By definition of the GF, values of C^0 and C^{80} calculated from Eq. (10) are dependent on r_{dry} . Also, a value of RH_0 must be specified when calculating C^0 by the growth-factor approach.

Values of C^0 and C^{80} are calculated from Eq. (10), using the GFs of Fitzgerald (1975) and Gerber (1985), and plotted over the RH range of interest in Fig. 3. The resulting values of C^0 are quite sensitive to r_{dry} and RH_0 , while the C^{80} values are sensitive to r_{dry} . In contrast, the formulas derived in the present work are independent of r_{dry} because the Kelvin effect is neglected and are independent of RH_0 because a fixed value of x_{sw} is assumed. The lack of dependence on r_{dry} and RH_0 makes the present formulas straightforward to implement in air quality models. The formulas can be readily applied to sea-salt generation functions, as discussed in Section 4. Values of C^0 calculated from the present method match those from Fitzgerald's equation when $r_{dry} = 3.0 \mu\text{m}$ and $RH_0 = 0.98$, and match those from Gerber's equation when $r_{dry} = 0.05 \mu\text{m}$ and $RH_0 = 0.98$ (see Fig. 3a). The C^{80} values from all three methods are

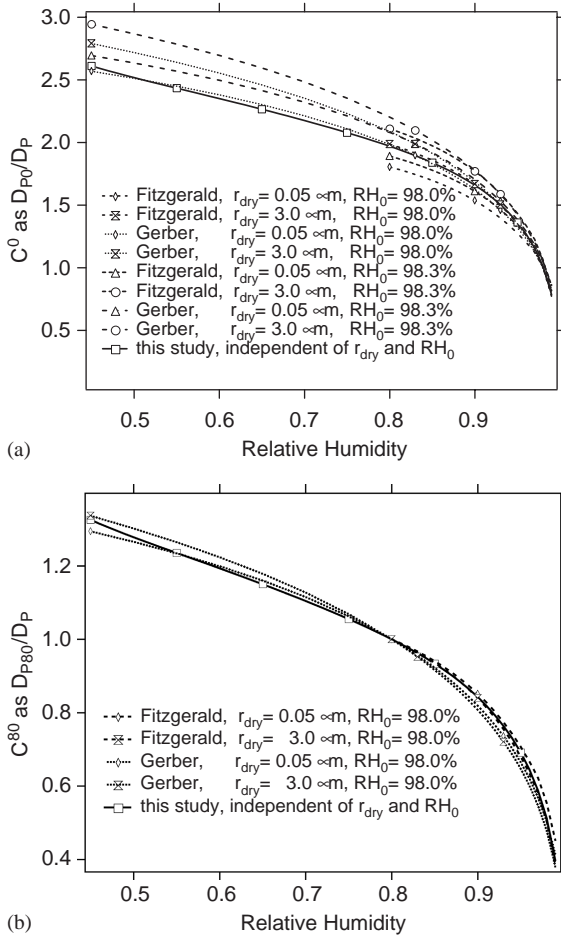


Fig. 3. Comparisons of correction factors from Fitzgerald (1975), Gerber (1985) and this study with respect to the state at (a) formation and (b) 80% RH.

in excellent agreement, as shown in Fig. 3b, further confirming the validity of the proposed method.

4. Application to sea-salt generation functions

Sea-spray droplets come in three varieties: film droplets, jet droplets, and spume droplets. Film droplets (0.5–5 μm) and jet droplets (3–50 μm) derive from the same process: the bursting of air-entrained bubbles from oceanic whitecaps formed during the breaking of wind-induced waves. Spume droplets (>20 μm), on the other hand, derive from the wind shear which tears the droplets directly from the wave crests (Andreas, 1998). In the surf zone, however, wave breaking is dominated by interactions with the sea bottom surface (de Leeuw et al., 2000) which increase the rate of air entrainment and whitecap formation. Consequently, sea-salt emissions

per unit area from the surf zone can be 1–2 orders higher than the oceanic background.

Source functions, formulated from different approaches using laboratory and field experiments, are often presented as continuous particle number-size distributions (dF_N/dr , where F_N is the number flux and r is a characteristic radius; unit: particles $\text{m}^{-2} \text{s}^{-1} \mu\text{m}^{-1}$) as a function of wind speed. Although the magnitude of the number flux is a function of wind speed, the profile of the size distribution generated is independent of wind speed since the source functions are of the form $dF/dr = f(u) \times f(r)$, where u is a characteristic wind speed. An overview of current estimates of the surface fluxes of sea-spray aerosol, beyond the scope of this study, can be found in Andreas (1998). Here we use several common source functions to demonstrate how to apply the method introduced in this paper.

4.1. Open-ocean source functions

Monahan et al. (1986):

$$\begin{aligned} \frac{dF_{N\text{-Open}}}{dr_{80}} &= \frac{dF_0}{dr_{80}}(\text{bubbles}), \\ &= 1.373 U_{10}^{3.41} r_{80}^{-3} (1 + 0.057 r_{80}^{1.05}) \times 10^{1.19} \exp(-B^2), \end{aligned} \quad (11)$$

where r_{80} is the droplet radius (μm) at a reference relative humidity of 80%, U_{10} is the wind speed (m s^{-1}) at an elevation of 10 m from the water surface, and $B = (0.380 - \log r_{80})/0.650$. This function is applicable to $U_{10} < 20 \text{ m s}^{-1}$ and $0.8 < r_{80} < 10 \mu\text{m}$.

Smith and Harrison (1998):

$$\frac{dF_{N\text{-Open}}}{dr_{80}} = \sum_{i=1}^2 A_i \exp \left\{ -f_i \left[\ln \left(\frac{r_{80}}{r_{0i}} \right) \right]^2 \right\}, \quad (12)$$

where $r_{01} = 3 \mu\text{m}$ and $r_{02} = 30 \mu\text{m}$, $f_1 = 1.5$ and $f_2 = 1.0$, and A_1 and A_2 are approximated by $A_1 = 0.2 U_{10}^{3.5}$ and $A_2 = 6.8 \times 10^{-3} U_{10}^3$. This function is applicable to $U_{10} < 20 \text{ m s}^{-1}$ and $0.5 < r_{80} < 150 \mu\text{m}$ (Dr. Michael H. Smith, per. comm.). Note here we use the notation A_i to be consistent with Smith and Harrison (1998). It is different from A_i in Eq. (3).

To apply the proposed method,

$$\begin{aligned} \text{number: } \frac{dF_{N\text{-Open}}}{dD_p} &= \frac{dF_{N\text{-Open}}}{dr_{80}} \frac{dr_{80}}{dD_{p80}} \frac{dD_{p80}}{dD_p} \\ &= \frac{1}{2} C^{80} \frac{dF_{N\text{-Open}}}{dr_{80}} \end{aligned} \quad (13)$$

and

$$\text{mass: } \frac{dF_{M\text{-Open}}}{dD_p} = 10^{-15} \frac{dF_{N\text{-Open}}}{dD_p} \frac{\pi}{6} D_p^3 \rho_p x \quad (14)$$

for dry mass flux. The factor of 10^{-15} accounts for unit conversion and C^{80} , ρ_p and x are given by Eqs. (9), (4) and (1). The units of $dF_{N\text{-Open}}/dD_p$ and $dF_{M\text{-Open}}/dD_p$

are particles $\text{m}^{-2}\text{s}^{-1}\mu\text{m}^{-1}$ and $\text{g}\text{m}^{-2}\text{s}^{-1}\mu\text{m}^{-1}$, respectively. When using Eqs. (13) and (14), the applicable size ranges of Eqs. (11) and (12) should be adjusted to local RH using the relationship, $r_{80} = \frac{1}{2}C^{80}D_p$.

4.2. Surf-zone source function

de Leeuw et al. (2000) provide a source function for sea spray produced by waves breaking in the surf zone from data collected at two locations on the California coast as

$$\frac{dF_{N\text{-Surf}}}{dD_{p0}} = 1.1 \times 10^7 e^{0.23U_{10}} D_{p0}^{-1.65}, \quad (15)$$

where D_{p0} is sea-salt particle diameter at formation. The function is applicable for $U_{10} < 9\text{ m s}^{-1}$ and $1.6 < D_{p0} < 20\mu\text{m}$. (Note that the factor of 10^7 was inadvertently omitted in the original publication.)

To apply the proposed method,

$$\text{number} : \frac{dF_{N\text{-Surf}}}{dD_p} = \frac{dF_{N\text{-Surf}}}{dD_{p0}} \frac{dD_{p0}}{dD_p} = C^0 \frac{dF_{N\text{-Surf}}}{dD_{p0}} \quad (16)$$

and

$$\text{mass} : \frac{dF_{M\text{-Surf}}}{dD_p} = 10^{-15} \frac{dF_{N\text{-Surf}}}{dD_{p0}} \frac{\pi}{6} D_p^3 \rho_p^x \quad (17)$$

for dry mass flux, where C^0 , ρ_p and x are given by Eqs. (8), (4) and (1), respectively. Again, the applicable size range of Eq. (15) should be adjusted to local RH when using Eqs. (16) and (17) following the relationship $D_{p0} = C^0 D_p$.

5. Conclusion

Here we introduced a simple method for correcting sea-salt-emission particle-size distributions as a function of relative humidity. The correction factors, corresponding to the size at formation and the size at 80% RH, are given as polynomial functions of local relative humidity which are straightforward to implement. Without major compromises, these factors are thermodynamically accurate and can be applied between 0.45 and 0.99 RH. Since the thermodynamic properties of sea-salt electrolytes have a weak dependence on ambient temperature, these factors can be regarded as temperature independent. The resultant sea-salt emissions can be used directly in atmospheric model simulations without further correction.

Acknowledgment

K.M.Z., A.S.W and G.S.T. would like to thank their support from the Visibility Improvement State and Tribal Association of the Southeast (VISTAS). We

appreciate the insightful comments from Dr. Ernie Lewis of Brookhaven National Laboratory. A portion of the research presented here was performed under the Memorandum of Understanding between the US Environmental Protection Agency (EPA) and the US Department of Commerce's National Oceanic and Atmospheric Administration (NOAA) and under agreement No. DW13921548. This work constitutes a contribution to the NOAA Air Quality Program. Although it has been reviewed by EPA and NOAA and approved for publication, it does not necessarily reflect their policies or views.

References

- Andreas, E.L., 1989. Thermal and size evolution of sea spray droplets. CRREL Report 89-11, US Army Cold Regions Research and Engineering Laboratory, Hanover, NH (NTIS ADA210484).
- Andreas, E.L., 1992. Sea spray and the turbulent air-sea fluxes. *Journal of Geophysical Research—Oceans* 97, 11,429–11,441.
- Andreas, E.L., 1998. A new sea spray generation function for wind speeds up to 32 m s^{-1} . *Journal of Physical Oceanography* 28, 2175–2184.
- Clegg, S.L., Simonson, J.M., 2001. A BET model of the thermodynamics of aqueous multicomponent solutions at extreme concentration. *Journal of Chemical Thermodynamics* 31, 1457–1472.
- de Leeuw, G., Neele, F.P., Hill, M., Smith, M.H., Vignali, E., 2000. Production of sea spray aerosol in the surf zone. *Journal of Geophysical Research—Atmospheres* 105, 29,397–29,409.
- Evans, M.C., Campbell, S.W., Bhethanabotla, V., Poor, N.D., 2004. Effect of sea salt and calcium carbonate interactions with nitric acid on the direct dry deposition of nitrogen to Tampa Bay, Florida. *Atmospheric Environment* 38 (29), 4847–4858.
- Fitzgerald, J.W., 1975. Approximation formulas for the equilibrium size of an aerosol particle as a function of its dry size and composition and the ambient relative humidity. *Journal of Applied Meteorology* 14, 1044–1049.
- Gerber, H.E., 1985. Relative-Humidity Parameterization of the Navy Aerosol Model (NAM). Naval Research Laboratory, Washington, DC (NTIS ADA1632090).
- Gong, S.L., Barrie, L.A., Blanchet, J.P., 1997. Modeling sea-salt aerosols in the atmosphere. 1. Model development. *Journal of Geophysical Research—Atmospheres* 102, 3805–3818.
- Hänel, G., 1976. The properties of atmospheric aerosol particles as a function of the relative humidity at thermodynamic equilibrium with the scattering moist air. *Advances in Geophysics* 19, 73–188.
- Knipping, E.M., Dabdub, D., 2003. Impact of chlorine emissions from sea-salt aerosol on coastal urban ozone. *Environmental Science and Technology* 37, 275–284.
- Laskin, A., Gaspar, D.J., Wang, W.H., Hunt, S.W., Cowin, J.P., Colson, S.D., Finlayson-Pitts, B.J., 2003. Reactions at interfaces as a source of sulfate formation in sea-salt particles. *Science* 301, 340–344.

- Millero, F.J., 1996. *Chemical Oceanography*, second ed. CRC Press, Boca Raton, FL.
- Monahan, E.C., Spiel, D.E., Davidson, K.L., 1986. In: Monahan, E.C., Mac Niocaill, G. (Eds.), *Oceanic Whitecaps*. Riedel, Norwell, MA, pp. 167–174.
- Paerl, H.W., Dennis, R.L., Whitall, D.R., 2002. Atmospheric deposition of nitrogen: implications for nutrient over-enrichment of coastal waters. *Estuaries* 25, 677–693.
- Smith, M.H., Harrison, N.M., 1998. The sea spray generation function. *Journal of Aerosol Science* 29, S189–S190.
- Stokes, R.H., Robinson, R.A., 1966. Interactions in aqueous nonelectrolyte solutions: I. Solute-solvent equilibria. *Journal of Physical Chemistry* 70, 2126–2130.
- Tang, I.N., Tridico, A.C., Fung, K.H., 1997. Thermodynamic and optical properties of sea salt aerosols. *Journal of Geophysical Research—Atmospheres* 102, 23,269–23,275.
- Wexler, A.S., Clegg, S.L., 2002. Atmospheric aerosol models for systems including the ions H^+ , NH_4^+ , Na^+ , SO_4^{2-} , NO_3^- , Cl^- , Br^- , and H_2O . *Journal of Geophysical Research—Atmospheres* 107 (art. No. 4207).