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ABSTRACT: The direct influence of aerosol particles on atmospheric radiation through light scattering is determined by their radii and refractive index, both of which are affected by uptake of water vapor. Previous expressions for these quantities have typically been given in terms of molality or solute mole or mass fraction rather than relative humidity, RH, which is the pertinent atmospheric variable. Here simple expressions are presented for the dependence of radius and index of refraction on RH for a particle of given dry mass, for several inorganic salts of atmospheric importance. These expressions, which are accurate to a few percent over a wide range of RH, are readily applicable in radiation transfer models and climate madels.

RADIUS AND INDEX OF REFRACTION OF AQUEOUS INORGANIC AEROSOL PARTICLES: DEPENDENCE ON RELATIVE HUMIDITY

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http://www.ecd.bnl.gov/steve/RHparamsARM05.pdf

MOTIVATION

- Calculating aerosol forcing in specific situations and representing aerosol forcing in climate models requires knowledge of the dependence of light scattering on relative humidity *rh*.
- Representing aerosol forcing therefore requires knowledge of the *rh*-dependence of particle radius *r* and index of refraction *n* for measured or modeled aerosol composition and (dry) size distribution.

THE PROBLEM

- Expressions for *r* and *n* are typically presented in terms of solute concentration, expressed as molality *m*, solute mass fraction *x*, or solute mole fraction n_s, none of which are readily available in field situations or to modelers.

THE SOLUTION

- Determine expressions for radius *r* and index of refraction *n* in terms of the fractional relative humidity *rh* (equal to the water activity a_w for situations in which the Kelvin effect can be neglected).

THIS STUDY

- This study develops accurate, readily applied parameterizations of the dependence of *r* and *n* on *rh* for several atmospherically relevant inorganic salts from previously published data.

SUBSTANCES OF INTEREST

Substance	Formula	MW g mol^{-1}	Density g cm^{-3}	ν
Ammonium sulfate	$(NH_4)_2SO_4$	132.14	1.77	3
Ammonium bisulfate	NH_4HSO_4	115.11	1.78	3
Letovicite	$(NH_4)_3H(SO_4)_2$	247.25	1.83	6
Ammonium nitrate	NH_4NO_3	80.04	1.73	2
Sodium nitrate	$NaNO_3$	84.99	2.26	2
Sodium sulfate	Na_2SO_4	142.04	2.68	3
Sodium bisulfate	$NaHSO_4$	120.06	2.44	2
Sodium chloride	NaCl	58.44	2.17	2
Sea salt		62.79	2.2	2

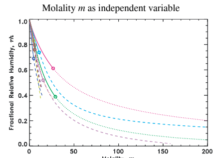
ABOUT THE GRAPHS …

- Each color denotes a different substance
- Solid lines refer to subsaturated region
- Dashed lines refer to supersaturated region
- Dotted lines denote unknown or poorly known data
- Circles denote saturation point

RADIUS, r

DATA AS TYPICALLY REPORTED

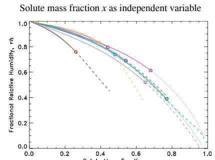
Molality *m* as independent variable



- Definitely not linear
- Independent variable unbounded

DATA AS SOMETIMES REPORTED

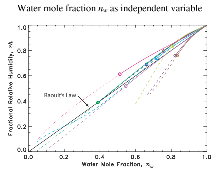
Solute mass fraction *x* as independent variable



- Not linear
- Independent variable restricted

ALTERNATIVE REPRESENTATION

Water mole fraction n_w as independent variable

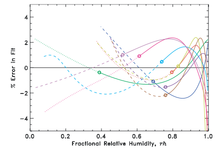


- $1-a_w$ nearly directly proportional to $1-n_w$
- Independent variable restricted
- Most substances follow Raoult's Law (solid line)

DESIRED QUANTITY: Radius r

- $r = r_{dry}\left[\frac{\nu M_w \rho_{dry}}{M_s \rho}\right]^{1/3}\left[\frac{M_s}{\nu M_w} - 1 + \frac{1}{1-n_w}\right]^{1/3}$
- $(1-n_w)$ is nearly directly proportional to $(1-a_w)$
- This suggests an approximation $r = r_{dry} c\left[b + \frac{1}{1-a_w}\right]^{1/3}$

ACCURACY OF FIT



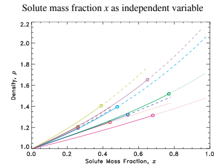
- Two-parameter fit to radius accurate to ~2%

DENSITY, ρ

- Density ρ needed to determine radius *r* from solute mass fraction *x*: $r = r_{dry}\left[\frac{\rho_{dry}}{\rho}\cdot\frac{1}{x}\right]^{1/3}$
- Thus the pertinent quantity is $\rho^{1/3}$

DATA AS TYPICALLY REPORTED

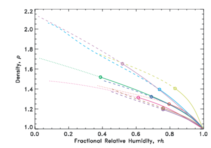
Solute mass fraction *x* as independent variable



- Nearly linear – permits extrapolation

DATA AS DESIRED

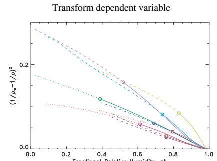
Relative humidity *rh* as independent variable



- Not linear

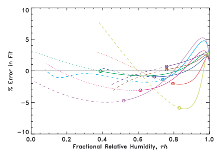
ALTERNATIVE REPRESENTATION

Transform dependent variable



- Nearly linear
- Fit as $\frac{1}{\rho_w} - \frac{1}{\rho} = c\sqrt{1-rh} \Rightarrow \rho = \frac{\rho_w}{1-c\rho_w\sqrt{1-rh}}$

ACCURACY OF FIT

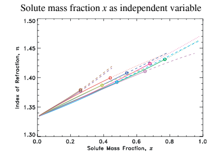


- Single-parameter fit to density accurate to ~5%
- Accuracy in the pertinent quantity $\rho^{1/3}$ is ~2%

REFRACTIVE INDEX, n

DATA AS TYPICALLY REPORTED

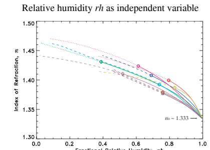
Solute mass fraction *x* as independent variable



- Nearly linear – permits extrapolation

DATA AS DESIRED

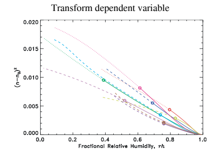
Relative humidity *rh* as independent variable



- Not linear
- For given *rh* the spread in *n* is only ±0.02 among substances

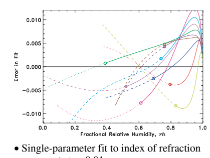
ALTERNATIVE REPRESENTATION

Transform dependent variable



- Nearly linear
- Fit as $n = n_0 + c\sqrt{1-rh}$

ACCURACY OF FIT



- Single-parameter fit to index of refraction accurate to ~0.01 (note expanded scale)

PARAMETERIZATION CONSIDERATIONS

*There is always a tradeoff between **accuracy** and **simplicity**. The approximations presented were guided by the following:*

Measurement issues:
- RH not accurately known
- Temperature not accurately known

Particle composition:
- Particle composition not accurately known
- Particles often internally mixed
- Possible insoluble inclusions
- Surface-active substances may affect properties

Data availability and accuracy:
- Uncertainties in original data
- Few data in supersaturated range
- Temperature dependence often not known

Concerns with complicated expressions (e.g., high-order polynomials)**:**
- Little or no physical insight provided
- Patterns not easily discerned
- Extrapolation not stable
- Mixing rules not possible
- Implied level of precision often not justified
- Mistakes likely in listing coefficients or in coding

Advantages of simple expressions:
- Physical insight
- Patterns readily recognized
- Extrapolation possible
- Path forward to developing mixing rules

SUMMARY

- Simple expressions are presented for:
  - radius r
  - density ρ
  - index of refraction n

  in terms of the relative humidity *rh*

$$r = r_{dry} c\left[b + \frac{1}{1-rh}\right]^{1/3}$$

$$\rho = \frac{\rho_w}{1 - c\rho_w\sqrt{1-rh}}$$

$$n = n_0 + c\sqrt{1-rh}$$

- Accuracy:
  - ~2% in radius r
  - ~5% in ρ, or ~2% in $\rho^{1/3}$
  - ~0.01 in index of refraction n

It is the mark of an educated mind to rest satisfied with the degree of precision which the subject admits, and not seek exactness where only an approximation is possible.

Aristotle