



*Supplement of*

## **Insights into the real part of natural sea spray aerosol refractive index in the Pacific Ocean**

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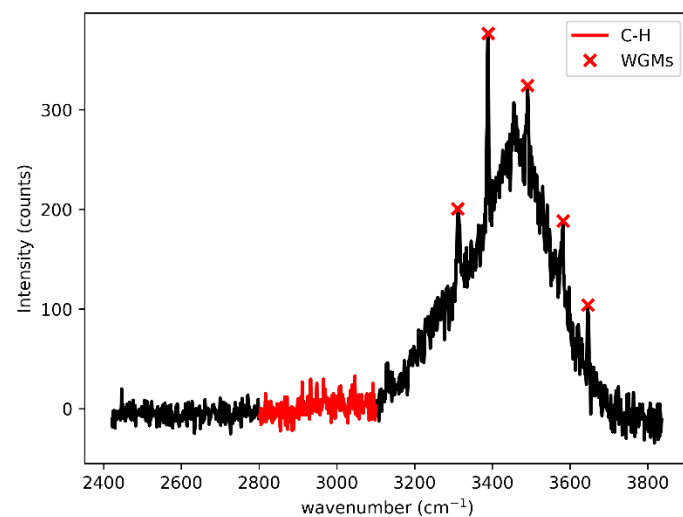


Figure S1. The Raman spectrum of the particles generated by the nebulizer at RH = 70%. The red part represents the typical peak position of the C-H peak, and the red cross marks indicate the particle's significant Whispering Gallery Modes (WGMs).

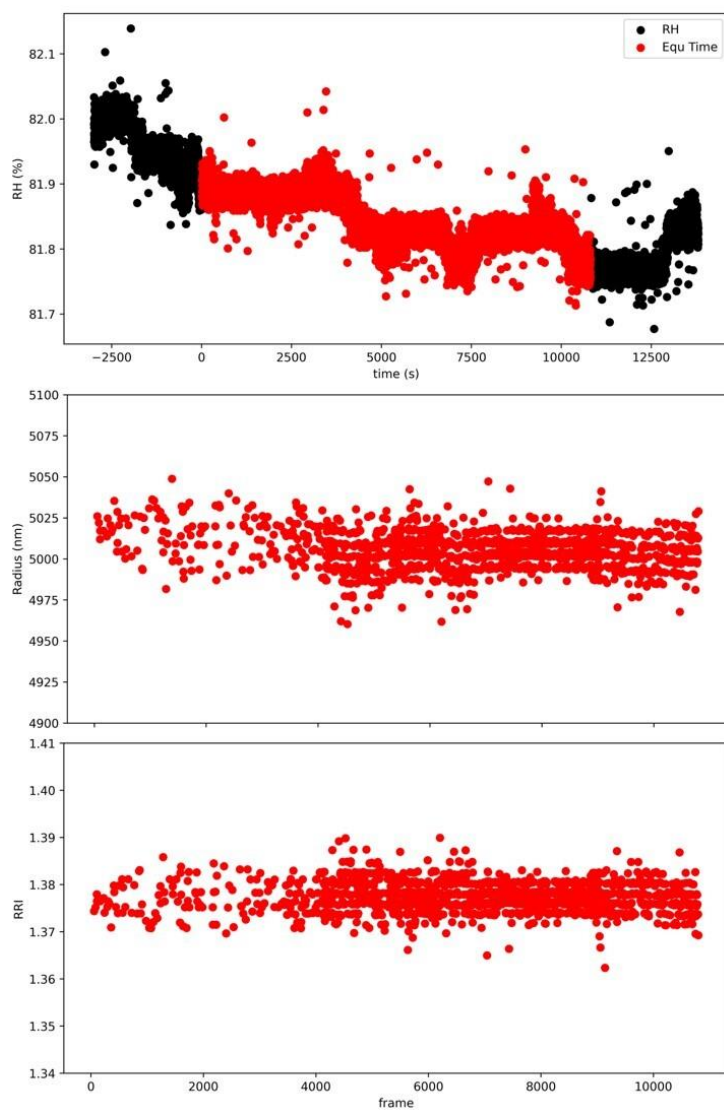


Figure S2. An example of the relative humidity (top) when an artificial sea salt particle is in equilibrium at  $RH = 81.8\%$ , with the fitted radius (middle) and RRI (bottom). The mean values are 5005.4 nm and 1.3773, with a standard deviation of 10.7 nm and 0.0029, respectively.

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### Model IV: $\text{H}^+ - \text{NH}_4^+ - \text{Na}^+ - \text{SO}_4^{2-} - \text{NO}_3^- - \text{Cl}^- - \text{H}_2\text{O}$

• [Description and abstract](#)

• This is the principal *E-AIM* input page for Model IV. Alternative [types of calculation](#) can be selected from the list below:

- Simple (this page)
- [Comprehensive](#)
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  - [temperature](#)
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#### Simple Calculation

Here, the model determines the state of a system containing water, and two or more ions, at equilibrium with an atmosphere of known temperature and relative humidity. Solids are allowed to form, at the choice of the user, but partitioning of trace gases into the vapour phase is not enabled.

- Inputs:** ambient relative humidity (expressed as a fraction), temperature, and the moles of each ion present.
- Outputs:** the amounts of liquid water, dissolved ions, and solids present at equilibrium. The partial pressures or partial pressure products of any  $\text{NH}_3$ ,  $\text{HNO}_3$ ,  $\text{HCl}$ , or  $\text{H}_2\text{SO}_4$  that would be in equilibrium with the condensed phase are also reported.
- Options:** control of the formation of solids, and equilibration of the system to the water vapour pressure above ice.

Specify the problem in the input fields below.

#### Ambient Conditions

Temperature (180 - 330 K):  Relative Humidity (0.1 - 1.0):

(See [inputs](#) for details of restrictions on the temperature range, related to the chemical composition of the system.)

If the temperature is less than 273.15 K, and you wish the system to be equilibrated to the water vapour pressure over ice, then check the box here. (Any relative humidity that has been entered above will be ignored.) ☐

#### Ionic Composition in Moles

$\text{H}^+$ : 
 $\text{NH}_4^+$ : 
 $\text{Na}^+$ :

$\text{SO}_4^{2-}$ : 
 $\text{NO}_3^-$ : 
 $\text{Cl}^-$ :

#### Other Chemical Components

The amounts of organic compounds present can be entered here. First, click the button to select compounds from the library or create new ones for this session.

[Manage Compounds](#)

#### Solid Phases

Check the boxes below to prevent the formation of selected solids, according to the [restrictions](#) that apply for this model. This enables the properties of metastable, supersaturated, aqueous aerosols to be investigated. The default for each calculation is that all solids can form.

No boxes should be checked for any systems containing both  $\text{NH}_4^+$  and  $\text{Cl}^-$  ions.

Omit the following solids:

<input type="checkbox"/> Ice	<input type="checkbox"/> $\text{H}_2\text{SO}_4 - \text{H}_2\text{O}$	<input type="checkbox"/> $\text{H}_2\text{SO}_4 - 2\text{H}_2\text{O}$
<input type="checkbox"/> $\text{H}_2\text{SO}_4 - 3\text{H}_2\text{O}$	<input type="checkbox"/> $\text{H}_2\text{SO}_4 - 4\text{H}_2\text{O}$	<input type="checkbox"/> $\text{H}_2\text{SO}_4 - 6.5\text{H}_2\text{O}$
<input type="checkbox"/> $\text{HNO}_3 - \text{H}_2\text{O}$	<input type="checkbox"/> $\text{HNO}_3 - 2\text{H}_2\text{O}$	<input type="checkbox"/> $\text{HNO}_3 - 3\text{H}_2\text{O}$
<input type="checkbox"/> $\text{HCl} - 3\text{H}_2\text{O}$	<input checked="" type="checkbox"/> $(\text{NH}_4)_2\text{SO}_4$	<input type="checkbox"/> $(\text{NH}_4)_3\text{H}(\text{SO}_4)_2$
<input type="checkbox"/> $\text{NH}_4\text{HSO}_4$	<input type="checkbox"/> $\text{NH}_4\text{NO}_3$	<input type="checkbox"/> $2\text{NH}_4\text{NO}_3 - (\text{NH}_4)_2\text{SO}_4$
<input type="checkbox"/> $3\text{NH}_4\text{NO}_3 - (\text{NH}_4)_2\text{SO}_4$		

To submit your calculation, click here:

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### E-AIM Model Results

Problem no. 1. iFail = 0

System pressure = 1.00000 atm  
 Volume = 1.000 m<sup>3</sup>  
 T = 293.150 K  
 RH = 0.60809 [pH<sub>2</sub>O = 0.14040E-01 atm]  
 (The relative humidity above is the input value. The equivalent partial pressure is also given.)

```

*****
** AQUEOUS PHASE **
*****
Species      Moles      Grams      Molality    Mole Frac.  Act. Coeff.
H(aq)        0.16569E+05    0.1670E+06    0.1920E+04    0.2127E-06    0.9371E+00
NH4(aq)       0.20000E+01      0.3608E+02    0.2317E+02    0.2567E+00    0.8527E+00
HSO4(aq)      0.22783E-04      0.2210E-02    0.2637E-03    0.2922E-05    0.6046E+00
SO4(aq)       0.99998E+00      0.9606E+02    0.1159E+02    0.1283E+00    0.1520E-01
OH(aq)        0.45594E-11      0.7754E-10    0.5282E-10    0.5852E-12    0.1150E+02
H2O(aq)       0.47919E+01      0.8631E+02    0.5551E+02    0.6149E+00    0.9899E+00
NH3(aq)       0.24420E-04      0.4159E-03    0.2829E-03    0.3134E-05    0.1626E+01
  
```

(1) The density of the aqueous phase is 1.33999 g per cm<sup>3</sup>, and its total volume is 1.63028E+02 cm<sup>3</sup>.

(2) The surface tension of the aqueous phase is 96.07 mN per m.

(3) The molal osmotic coefficient of the aqueous phase is 0.7944. (It is equal to  $-\ln(a_{\text{H}_2\text{O}}) / ((\text{MW}/1000) \cdot \text{SUM})$ , where  $a_{\text{H}_2\text{O}}$  is the water activity, SUM is the total molality of all solute species, and MW is the molar mass of water in grams.)

(4) The value of the parameter kappa is 0.7629 for this aqueous solution, and/or other liquid phase, at the specified temperature. See Petters and Kreidenweis (*Atmos. Chem. Phys.* 7, 1961-1971, 2007) for a definition, and the *E-AIM* web page describing the model outputs. The (dry) chemical system is assumed to be made up of the following solid and/or liquid components:  $(\text{NH}_4)_2\text{SO}_4 - 1.0000E+00$  moles (7.4656E+01 cm<sup>3</sup>).

There is no second liquid phase.

Figure S3. A computational example of the E-AIM IV model, a thermodynamic model used to predict aerosol solution properties like chemical composition, water content, and density under different environmental conditions.

Table S1. Geographic coordinates, and salinity data for 7 stations investigated.

Station	Latitude (°N)	Longitude (°E)	Salinity (‰)
M30	28.5	155.0	35.1618
M35	33.0	155.0	34.6105
stn41	36.7	155.0	34.4737
stn43	39.5	155.0	34.2808
KE2	36.6	152.5	34.1683
Kuroshio	26.0	125.8	33.9200
South China Sea	18.0	116.0	33.9478

Table S2. Input parameters for the E-AIM IV model, results, and the RRI calculated using the molar refraction method

Temperature (K)	RH (%)	$x_{(\text{NH}_4)_2\text{SO}_4}$	$\rho$	$M_e^a$	$R_e^b$	n
293.15	60.81	0.173	1.340	37.72	7.05	1.415
293.15	65.82	0.151	1.319	35.26	6.64	1.411
293.15	69.12	0.138	1.305	33.72	6.37	1.408
293.15	72.02	0.126	1.291	32.39	6.15	1.405
293.15	76.81	0.107	1.266	30.23	5.78	1.400
293.15	79.62	0.096	1.249	28.97	5.57	1.396
293.15	82.23	0.086	1.232	27.77	5.37	1.392
293.15	87.03	0.065	1.194	25.49	4.98	1.383

a  $M_e = x_1M_1 + x_2M_2$ , where  $M_e$  is the relative molecular mass of the mixture. The relative molecular mass of ammonium sulfate and water are 132.14 g/mol and 18.02 g/mol, respectively.

b  $R_e = x_1R_1 + x_2R_2$ , where  $R_e$  is the molar refraction of the mixture. The molar refraction of a pure substance is calculated by the following formula:  $R = \frac{n^2 - 1}{n^2 + 2} \frac{M}{\rho}$  and the molar refractions of ammonium sulfate and water are 23.06 and 3.71, respectively.