

## Supporting information for:

# The Acidity of Atmospheric Particles and Clouds

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## 44 S1 Supplementary information for the definition of pH

### 45 S1.1 Operational definition and pH measurements

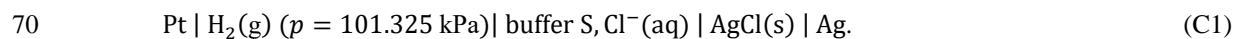
#### 46 *Operational definition of pH*

47 The notional character of the pH definition was recognized decades ago and has led to operational definitions of pH  
48 in terms of primary standard methods for pH measurements. These standard methods have been agreed upon by expert  
49 consensus, such as during the IUPAC-sponsored conference on “Harmonization of pH Scale Recommendations” in  
50 the year 1980 and refinements since then (Covington et al. 1985; Buck et al., 2002). The operational definition of pH  
51 is based on the principle of determining the difference between the pH of a solution of interest and that of a reference  
52 (buffer) solution of known pH by means of measuring the difference in electromotive force, i.e. an electric potential  
53 difference, using two electrochemical cells. High-precision measurement of absolute pH values are made with a so-  
54 called primary method using electrochemical cells without transference as described in great detail by Buck et al.  
55 (2002) and briefly introduced in this section.

56 The main purpose of the rather laborious absolute pH measurements is to establish the pH values and associated  
57 uncertainties of well-defined, stable standard buffer solutions (so-called primary standard buffers) at certain  
58 concentrations and over a meaningful temperature range, e.g. the primary standard of 0.05 mol kg<sup>-1</sup> potassium  
59 hydrogen phthalate (in water) has a determined pH value of 4.005 at 25 °C, 3.997 at 10 °C and 4.000 at 0 °C. Tabulated  
60 values of primary standard buffer solutions, covering acidic to alkaline pH ranges, serve as calibration standards for a  
61 wider range of so-called secondary standard buffer solutions, covering the pH scale from 1.7 to 13.4 (see tables 1 &  
62 2 in Buck et al., 2002). Primary or secondary standard solutions of known pH are then used in instrument calibration  
63 and the quantification of the pH of a sample solution of interest. As stated by Buck et al., (2002), the declaration of a  
64 pH measurement by a “primary method” requires assuring full traceability of the results of all measurements and  
65 consideration of their uncertainties as well as of limitations in the theory employed in the determination of  
66 experimental variables.

#### 67 *Absolute pH measurement with a Harned cell*

68 IUPAC recommends the Harned cell, a cell without liquid junction, as a primary method of pH measurement. Using  
69 the conventional notation from electrochemistry, the Harned cell is defined by:



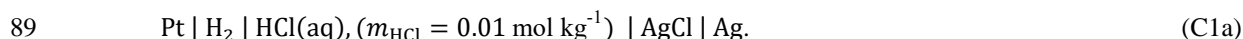
71 Here, cell (C1) contains an aqueous standard buffer solution S of known composition as well as chloride ions (typically  
72 in form of an aqueous solution of KCl or NaCl). The cell includes a silver–silver-chloride electrode and a hydrogen–  
73 platinum electrode (platinum as catalyst), allowing for concentration-dependent measurements of the electrical  
74 potential. The potential difference  $E_1$  of the cell is found by using the cell reaction  $\text{AgCl}(\text{s}) + \frac{1}{2}\text{H}_2(\text{g}) \rightleftharpoons \text{Ag}(\text{s}) +$   
75  $\text{H}^+(\text{aq}) + \text{Cl}^-(\text{aq})$  and application of Nernst’s equation to yield (Buck et al., 2002),

$$76 \quad E_1 = E^\ominus - \frac{RT}{F} \ln \left[ a_{\text{H}^+} \frac{m_{\text{Cl}^-}}{m^\ominus} \gamma_{\text{Cl}^-} \right], \quad (\text{S1})$$

77 where  $E^\ominus$  is the standard potential difference of the cell at absolute temperature  $T$  when the hydrogen gas pressure is  
 78 corrected to 101.325 kPa.  $R$  is the ideal gas constant (8.3144598 J K<sup>-1</sup> mol<sup>-1</sup>) and  $F$  the Faraday constant  
 79 (9.6485332 × 10<sup>4</sup> C mol<sup>-1</sup>). Equation (S1) can be rearranged to express the molality-based acidity function  $p(a_H\gamma_{Cl})$ ,  
 80 which is a linear function of the measured potential difference ( $E_1 - E^\ominus$ ) and the logarithm of the chloride ion  
 81 molality,

$$82 \quad p(a_H\gamma_{Cl}) = -\log_{10}(a_{H^+}\gamma_{Cl^-}) = \frac{E_1 - E^\ominus}{(RT/F)\ln(10)} + \log_{10}\left(\frac{m_{Cl^-}}{m^\ominus}\right). \quad (S2)$$

83 Calculation of the (molal) pH value of solution S from Eq. (S2) seems straightforward,  $pH(S) = p(a_H\gamma_{Cl}) +$   
 84  $\log_{10}(\gamma_{Cl^-})$ . However, it requires an independent assumption about the value of the single-ion activity coefficient of  
 85 the chloride ion (immeasurable). The Bates–Guggenheim convention established a means for a standardized way of  
 86 determining  $\gamma_{Cl^-}$  under specified conditions (outlined below). The standard potential difference  $E^\ominus$  is also determined  
 87 using the Harned cell, with only aqueous HCl present at a specified molality;  $m_{HCl} = 0.01$  mol kg<sup>-1</sup> is recommended.  
 88 This buffer-free Harned cell configuration is



90 The Nernst equation for cell C1a involves the molality of HCl and the mean molal activity coefficient  $\gamma_{\pm,HCl}$ , which  
 91 is known for various temperatures from experimental data, e.g.  $\gamma_{\pm,HCl} = 0.9042$  for  $m_{HCl} = 0.01$  mol kg<sup>-1</sup> (in pure  
 92 water) at 298.15 K (Bates and Robinson, 1980). Hence, a precise measurement of the electrical cell potential  $E_{1a}$   
 93 allows for an unambiguous calculation of  $E^\ominus$  via

$$94 \quad E^\ominus = E_{1a} + \frac{2RT}{F} \ln[0.01 \gamma_{\pm,HCl}]. \quad (S3)$$

95 As mentioned above, to arrive at a pH value by means of the acidity function (Eq. S2),  $\log_{10}(\gamma_{Cl^-})$  needs to be  
 96 quantified independently. In brief, the IUPAC (Buck et al., 2002) recommends the following two-step procedure: (i)  
 97 the value of the acidity function at zero chloride molality  $p(a_H\gamma_{Cl})^0$  is determined as the intercept of Eq. (S2) by  
 98 means of a linear extrapolation using several measurements with cell C1 at different molalities of NaCl or KCl (small  
 99 additions of chloride while maintaining a total molal ionic strength  $I < 0.1$  mol kg<sup>-1</sup>); (ii) the chloride ion activity  
 100 coefficient,  $\gamma_{Cl^-}^0$ , at zero chloride molality (the trace activity coefficient) is determined using Debye–Hückel theory.  
 101 In aqueous solutions of low ionic strength ( $I < 0.1$  mol kg<sup>-1</sup>), the Debye–Hückel equation for single-ion activity  
 102 coefficients is applicable (but imperfect). The Bates–Guggenheim convention adopts the expression

$$103 \quad \log_{10}(\gamma_{Cl^-}^0) = -A \frac{\sqrt{I}}{1 + b\sqrt{I/m^\ominus}}, \quad (S4)$$

104 with parameter  $A$  as the temperature-dependent Debye–Hückel constant; parameter  $b$  set to 1.5, assumed constant for  
 105 temperatures in the range from 5 – 50 °C; and  $I = \frac{1}{2} \sum_i m_i z_i^2$ , the ionic strength of the (standard) solution with  $z_i$  the  
 106 normalized integer charge of ion  $i$ . Values of  $A$  are tabulated in the appendix of Buck et al. (2002), e.g.  $A = 0.5100$

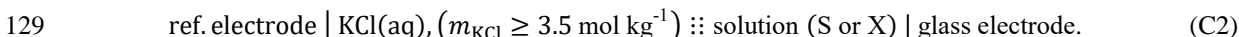
107 mol<sup>-1/2</sup> kg<sup>-1/2</sup> at 298.15 K. Employing this convention and parameters, the primary method allows determining the pH  
 108 of different standard solutions S based on Eqs. (S2 – S4) as follows:

$$109 \quad \text{pH(S)} = \lim_{m_{\text{Cl}^-} \rightarrow 0} \left[ \frac{E_1 - E^\ominus}{(RT/F) \ln(10)} + \log_{10} \left( \frac{m_{\text{Cl}^-}}{m^\ominus} \right) - A \frac{\sqrt{I}}{1 + 1.5 \sqrt{I/m^\ominus}} \right]. \quad (\text{S5})$$

110 The experimental uncertainty for a typical primary pH measurement is of the order 0.004 pH units. However, the  
 111 uncertainty associated with the assumptions made by the Bates–Guggenheim convention add an estimated 0.01 pH  
 112 units of uncertainty (95 % confidence interval) for  $I < 0.1$  mol kg<sup>-1</sup>, while increasing uncertainty is expected towards  
 113 higher ionic strength (Buck et al., 2002). Most of the latter uncertainty arises from assumptions about the true  
 114 dissociation of ions in buffer solutions (affecting the value of  $I$ ) and the effective distance of closest approach of ions  
 115 (factored into  $b = 1.5$ ). We refer to the IUPAC recommendations detailed in Buck et al. (2002) and the pH uncertainty  
 116 evaluation by Meinrath and Spitzer (2000) for recommended measurement protocols, calibration and considerations  
 117 of uncertainties as well as various technical details of the procedures introduced.

#### 118 *Practical pH measurements with glass electrodes*

119 In practice, secondary methods are frequently used, among them cells with a glass electrode instead of a H<sub>2</sub>|Pt  
 120 electrode. These secondary methods use cells with transference, which contain liquid junctions (e.g. a salt bridge),  
 121 leading to irreversible migration of ions and associated non-negligible liquid junction potentials contributing to the  
 122 measured potential difference of interest. The modern pH meters used widely in laboratories and industry are based  
 123 on H<sup>+</sup>-ion-responsive glass electrodes. This choice of electrode is for reasons of convenience and reliability. The key  
 124 part of the electrode is a glass membrane (a thin piece of H<sup>+</sup>-sensitive glass), often shaped in the form of a bulb,  
 125 attached to a stem of glass of high electric resistivity containing an internal reference electrode and filling solution of  
 126 fixed chloride concentration. pH meters come in many shapes and sizes, including flat models, capillary probes and  
 127 devices with ion-selective electrodes specific for other ions, e.g. nitrate, chloride, sodium and ammonium (Kolb, 1979;  
 128 Buck et al., 2002). The glass-electrode electrochemical cell is defined by



130 The reference electrode is usually of the silver–silver chloride type, with a salt bridge, e.g. a porous plug junction in  
 131 contact with the solution to be tested. The working principle of a glass electrode is based on the development of an  
 132 electrical potential at H<sup>+</sup>-sensitive glass–liquid interfaces. The potential at the outside glass surface depends on the pH  
 133 of the sample solution measured, while the potential at the inside surface is established by the constant pH of the  
 134 filling solution (e.g. concentrated KCl<sub>(aq)</sub>). Sometimes the glass and reference electrodes are combined into a single-  
 135 probe *combination electrode*.

136 The pH of sample solution X is determined via the measured potential difference  $E_2(\text{X}) - E_2(\text{S})$  using an adequate  
 137 standard buffer solution of known pH(S),

$$138 \quad \text{pH(X)} = \text{pH(S)} - \frac{E_2(\text{X}) - E_2(\text{S})}{(RT/F) \ln(10)}. \quad (\text{S6})$$

139 The direct application of Eq. (S6) is an example of a simple one-point calibration. Higher precision measurements are  
 140 carried out by using a two-point or multi-point calibration procedure, in which at least two standard buffers are used  
 141 that bracket the (unknown) pH(X). Ideally, the standard buffers chosen are close (above and below) in pH to pH(X),  
 142 leading to reduced uncertainties. Two-point or multipoint calibrations are needed to achieve a target pH uncertainty  
 143 of about 0.02 – 0.03 near 25 °C. Details about such methods, proper instrument calibration procedures and associated  
 144 uncertainties are outlined in Buck et al. (2002) as well as the manuals and guidelines of commercial pH-meter  
 145 manufacturers. Moreover, these references point out that special considerations are necessary for pH measurements  
 146 in non-aqueous solutions or solutions containing substantial amounts of organic components, which may affect the  
 147 behavior of the electrodes and junctions.

## 148 S1.2 Derivation of pH scale conversions

149 Conversions among pH values calculated using different concentration scales (molarity, molality, mole fraction, etc.)  
 150 are possible and necessary for an adequate comparison of model predictions. It is recommended to convert all pH  
 151 values to the molality scale. For clarity, the molality-scale pH is denoted by symbol “pH” while the pH on other scales  
 152 is indicated by a subscript (e.g. pH<sub>x</sub> for the mole-fraction-based pH). Generally, formulas for the conversion of pH  
 153 scales are derived using the equivalence of the (electro-)chemical potential of single ions expressed in any  
 154 concentration scale. For example, in the case of an electroneutral liquid phase (i.e. cancellation of the local electrostatic  
 155 potential within the phase), the chemical potential of H<sup>+</sup>,  $\mu_{\text{H}^+}^l$ , is given by

$$156 \quad \mu_{\text{H}^+}^l = \mu_{\text{H}^+}^{\ominus,(m)} + RT \ln \left( \frac{m_{\text{H}^+}}{m^{\ominus}} \gamma_{\text{H}^+} \right) = \mu_{\text{H}^+}^{\ominus,(x)} + RT \ln (x_{\text{H}^+} f_{\text{H}^+}^* ). \quad (\text{S7})$$

157 Here, the chemical potential is expressed either using the molality scale (first equality on right hand side) or the mole  
 158 fraction scale (second equality). In a general case, neither of the two standard state chemical potentials ( $\mu_{\text{H}^+}^{\ominus,(m)}$ ,  $\mu_{\text{H}^+}^{\ominus,(x)}$ )  
 159 nor the two activity coefficients are of equivalent values, yet the correct combinations according to Eq. (S7) yield an  
 160 equivalent  $\mu_{\text{H}^+}^l$ . By using this fact combined with the detailed definitions of the different activity coefficient scales  
 161 and reference states, a correct mapping between scales is possible, which then allows also for a conversion between  
 162 pH values defined on corresponding scales. Note, when solvents other than water are involved one needs to correctly  
 163 account for the definitions of the activity coefficients in terms of their reference states (e.g. infinite dilution in pure  
 164 water or in a specific water–organic mixed solvent), then follow the derivation steps analogous to those outlined  
 165 below. In the case described here, the reference states of  $\gamma_{\text{H}^+}$  and  $f_{\text{H}^+}^*$  are both that of infinite dilution of H<sup>+</sup> in pure  
 166 water, where  $\gamma_{\text{H}^+} \rightarrow 1$  and  $f_{\text{H}^+}^* \rightarrow 1$  (regardless of whether other solvent compounds are present or not), such that both  
 167 activity coefficients are equivalent at that reference point. This is consistent with models that only treat organic-free  
 168 aqueous electrolyte systems as well as with models treating organic–inorganic electrolyte solutions, such as  
 169 AIOMFAC. Using these definitions leads to (e.g. Zuend, 2007, pp. 45 – 47)

170 
$$\ln(\gamma_{\text{H}^+}) = \ln(f_{\text{H}^+}^*) + \ln\left(\frac{x_{\text{H}^+}}{M_w m_{\text{H}^+}}\right), \quad (\text{S8})$$

171 where  $M_w$  denotes the molar mass of water. Using the definition of molality,  $m_{\text{H}^+} = \frac{n_{\text{H}^+}}{\sum_s n_s M_s}$ , (compatible with the  
172 AIOMFAC model) Eq. (S8) can be expressed as

173 
$$\ln(\gamma_{\text{H}^+}) = \ln(f_{\text{H}^+}^*) + \ln\left(\frac{1}{M_w} \sum_s x_s M_s\right). \quad (\text{S9})$$

174 Here, the summation over index  $s$  covers non-electrolyte species only, i.e. water and organic mixture components,  
175 while  $x_s = \frac{n_s}{\sum_s n_s + \sum_i n_i}$  is the mole fraction of ‘solvent’  $s$  computed by accounting for all solution species, including  
176 molar amounts of  $\text{H}^+$  and other ions ( $\sum_i n_i$ ). For the case of an aqueous electrolyte solution absent any organic  
177 components or when water is defined as the only solvent (e.g. in the E-AIM model), molality is defined by  $m_{\text{H}^+} =$   
178  $\frac{n_{\text{H}^+}}{n_w M_w}$  and  $x_w = \frac{n_w}{n_w + \sum_i n_i}$ . In this case, Eq. (S9) simplifies to

179 
$$\ln(\gamma_{\text{H}^+}) = \ln(f_{\text{H}^+}^*) + \ln(x_w). \quad (\text{S10})$$

180 Using Eq. (S9) with the definitions of pH on molality and mole fraction scales (Eqs. 1 and 2 from main text) results  
181 in

182 
$$\text{pH}_x = -\log_{10}\left(\frac{m_{\text{H}^+}}{m^\ominus} \frac{\sum_s n_s M_s}{M_w n_{\text{H}^+}}\right) \text{pH}, \quad (\text{S11})$$

183 with  $m_{\text{H}^+} = \frac{n_{\text{H}^+}}{\sum_s n_s M_s}$  this simplifies to

184 
$$\text{pH} = \text{pH}_x + \log_{10}(m^\ominus M_w) \approx \text{pH}_x - 1.74436. \quad (\text{S12})$$

185 The exact same conversion formula (Eq. S12) results for solutions for which the molality is defined via  $m_{\text{H}^+} = \frac{n_{\text{H}^+}}{n_w M_w}$   
186 which led to Eq. S10. This is the case because the reference state in both cases is that of infinite dilution of ion “ $i$ ” in  
187 pure water. As a reminder,  $m^\ominus$  stands for unit molality (1 mol  $\text{kg}^{-1}$ ). Therefore, pH values defined on molality and  
188 mole fraction scales are offset by a constant value of about  $-1.74436$ .

189 The analogous conversion between molarity and molality-based pH is given by (Jia et al., 2018)

190 
$$\text{pH}_c = \text{pH} - \log_{10}\left(\frac{c^\ominus}{m^\ominus \rho_0} \cdot 10^3 \frac{\text{dm}^3}{\text{m}^3}\right). \quad (\text{S13})$$

191 Here,  $\rho_0$  is the density of the reference solvent, water, in units of  $\text{kg m}^{-3}$ , while  $10^3 \frac{\text{dm}^3}{\text{m}^3}$  is a conversion factor necessary  
192 when concentrations are expressed in non-SI units of  $\text{mol dm}^{-3}$  (or moles per liter). Because liquid-state density of the  
193 reference solvent depends weakly on temperature, the exact relation between pH and  $\text{pH}_c$  is non-linear. However, in  
194 the case of water near room temperature ( $\rho_0$  close to  $1000 \text{ kg m}^{-3}$ ), the logarithm in Eq. (S13) yields a small number,  
195 resulting in  $\text{pH}_c \approx \text{pH}_m \text{pH}_m \text{pH}$  (Jia et al., 2018).

196 **S2. Supplementary information for sulfate production in Figure 3**

197 The reaction rates for the S(IV) –S(VI) conversion processes listed in Table S1 were calculated using the conditions  
198 for Beijing Winter Haze published in Cheng et al. (2016): 271 K, 40 ppb SO<sub>2</sub>, 66 ppb NO<sub>2</sub>, 3 ppb O<sub>3</sub>, 0.01 ppb  
199 HOOH, 3.16×10<sup>-3</sup> M [Mn<sup>+2</sup>], and

$$200 \quad [\text{Fe}^{+3}] = \begin{cases} 10^{-3} \text{ M for } pH < 3 \\ 10^3 [H^+]^3 \text{ M for } pH \geq 3 \end{cases} \quad (\text{S14})$$

201 Henry's Law constants are available in Table S2.

202 **S3. Supplementary information for proxy estimates in Figure 15**

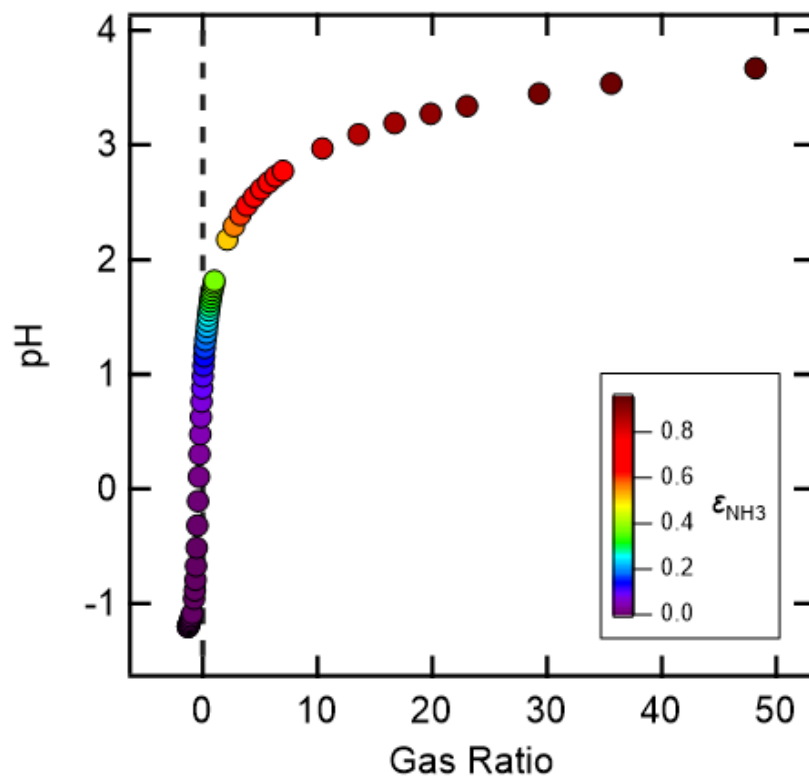
203 (a) *Inorganic ion balance*: There are 5 points for the Tianjin data set off scale. The lines represent the linear fits to  
204 each data set based on a least squares regression analysis. The fit parameters are as follows: northeast USA (slope =  
205 -45.63, intercept = 0.71, R<sup>2</sup> = 0.32, n = 3623), southeast USA (slope = -27.11, intercept = 1.31, R<sup>2</sup> = 0.36, n = 555),  
206 California (slope = -12.27, intercept = 2.25, R<sup>2</sup> = 0.34, n = 102), Tianjin (slope = -1.346, intercept = 3.16, R<sup>2</sup> = 0.02, n  
207 = 241). Note that a positive value for the ion balance represents conditions with Σ(anion equivalents) > Σ(cation  
208 equivalents).

209 (b) *Cation/anion molar equivalents ratio*: There are 7 points for the Tianjin data set off scale. The lines represent the  
210 linear fits to each data set based on a least squares regression analysis. The fit parameters are as follows: northeast  
211 USA (slope = 2.88, intercept = -1.88, R<sup>2</sup> = 0.72, n = 3623), southeast USA (slope = 1.34, intercept = 0.07, R<sup>2</sup> = 0.27, n  
212 = 524), California (slope = 0.69, intercept = 1.70, R<sup>2</sup> = 0.27, n = 102), Tianjin (slope = 0.65, intercept = 2.45, R<sup>2</sup> = 0.28,  
213 n = 241).

214 (c) *Gas ratio*: The lines represent the linear fits to each data set based on a least squares regression analysis. The fit  
215 parameters are as follows: northeast USA (slope = 1.73, intercept = 0.44, R<sup>2</sup> = 0.75, n = 3534), southeast USA (slope  
216 = 0.012, intercept = 0.89, R<sup>2</sup> = 0.21, n = 525), California (slope = 0.76, intercept = 1.93, R<sup>2</sup> = 0.72, n = 102, GR values  
217 below -2 excluded from the fit), Tianjin (slope = 0.03, intercept = 3.10, R<sup>2</sup> = 0.01, n = 241).

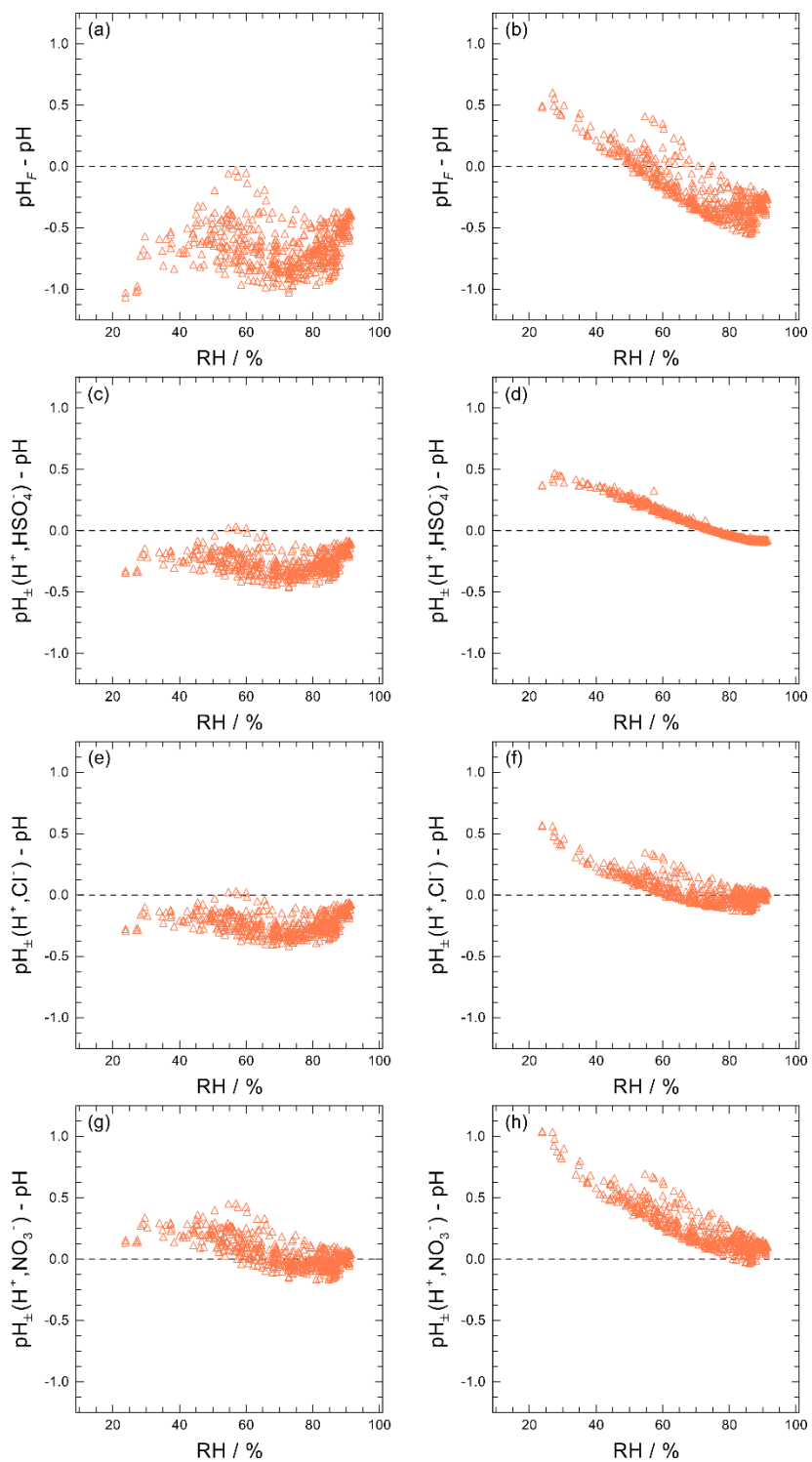
218 (d) *pH from HNO<sub>3</sub> partitioning*: The lines represent the linear fits to each data set based on a least squares regression  
219 analysis. The fit parameters are as follows: northeast USA (slope = 0.54, intercept = -0.06, R<sup>2</sup> = 0.16, n = 3268),  
220 California (slope = 0.27, intercept = 2.01, R<sup>2</sup> = 0.09, n = 102), Tianjin (slope = -0.27, intercept = 4.20, R<sup>2</sup> = 0.00, n =  
221 234, five points with pH<sub>F</sub> > 7 were excluded from the fit).

222 (f) *pH from NH<sub>3</sub> partitioning*: The lines represent the linear fits to each data set based on a least squares regression  
223 analysis. The fit parameters are as follows: southeast USA (slope = 1.15, intercept = 0.25, R<sup>2</sup> = 0.41, n = 486),  
224 California (slope = 0.22, intercept = 2.41, R<sup>2</sup> = 0.22, n = 102), Tianjin (slope = 1.18, intercept = 0.95, R<sup>2</sup> = 0.41, n =  
225 234, five points with pH<sub>F</sub> > 7 were excluded from the fit).

228  
229

230 **Figure S1.** Aerosol pH modeled in E-AIM (model II) versus the Gas Ratio ( $GR = ([\text{TNH}_4] - 2[\text{TSO}_4])/[\text{TNO}_3]$ ).  
231 Conditions for the simulations were: Temperature = 298 K, RH = 0.7,  $[\text{TSO}_4] = 10 \mu\text{g m}^{-3}$ ,  $[\text{TNO}_3] = 10 \mu\text{g m}^{-3}$ ,  
232  $[\text{TNH}_4]$  varied from 0 –  $130 \mu\text{g m}^{-3}$ . Color of the symbols corresponds to the fraction of TNH<sub>4</sub> in the gas phase  
233 ( $\epsilon_{\text{NH}_3} = [\text{NH}_3]/([\text{NH}_3] + [\text{NH}_4^+])$ ). The dotted line at GR = 0 indicates the transition from “ammonia-poor” ( $[\text{TNH}_4]$   
234  $< 2[\text{TSO}_4]$ ) to “ammonia-rich” ( $[\text{TNH}_4] > 2[\text{TSO}_4]$ ) conditions.





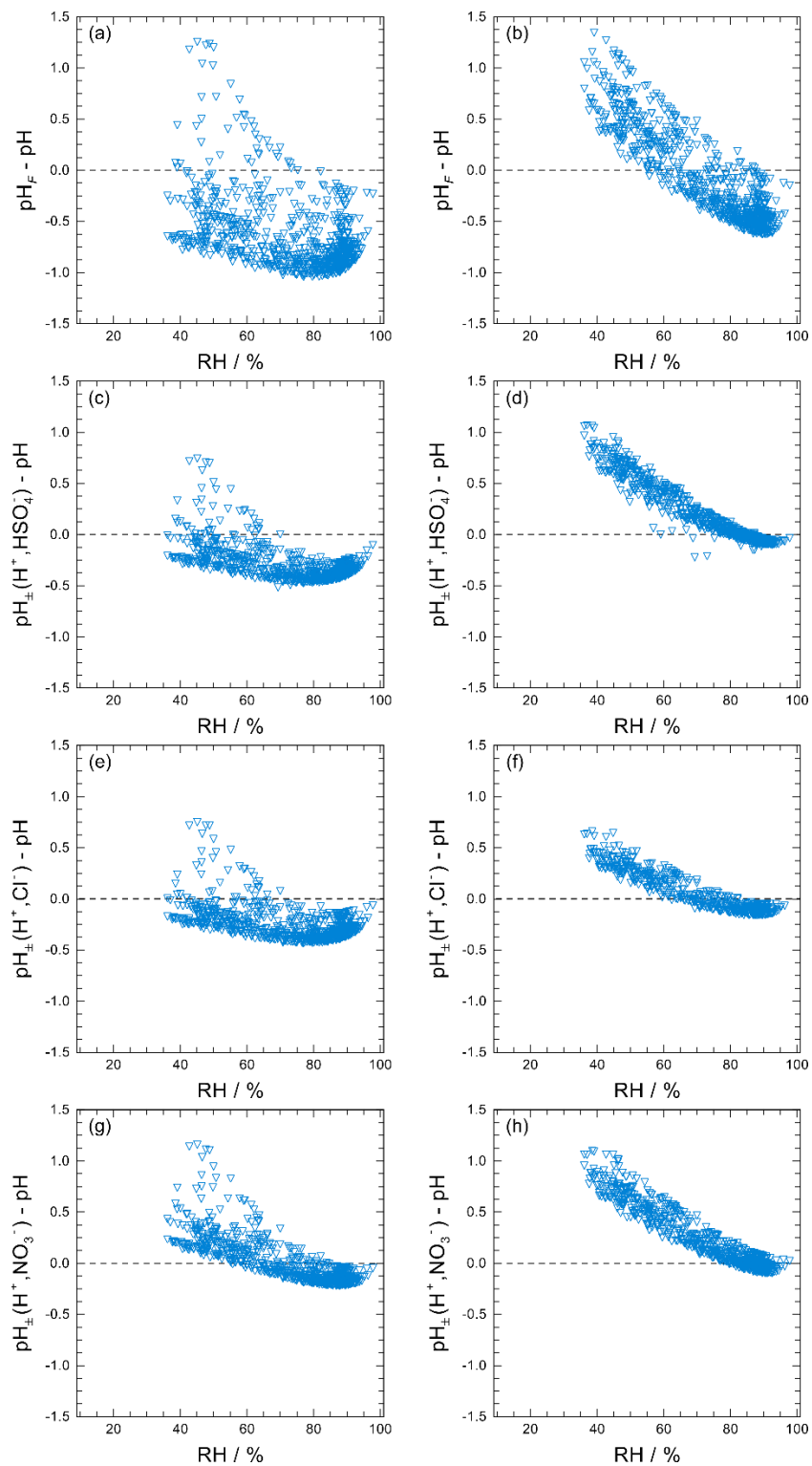
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238 **Figure S2.** Comparison of different metrics of calculating pH using the CalNex data for AIOMFAC-GLE (left

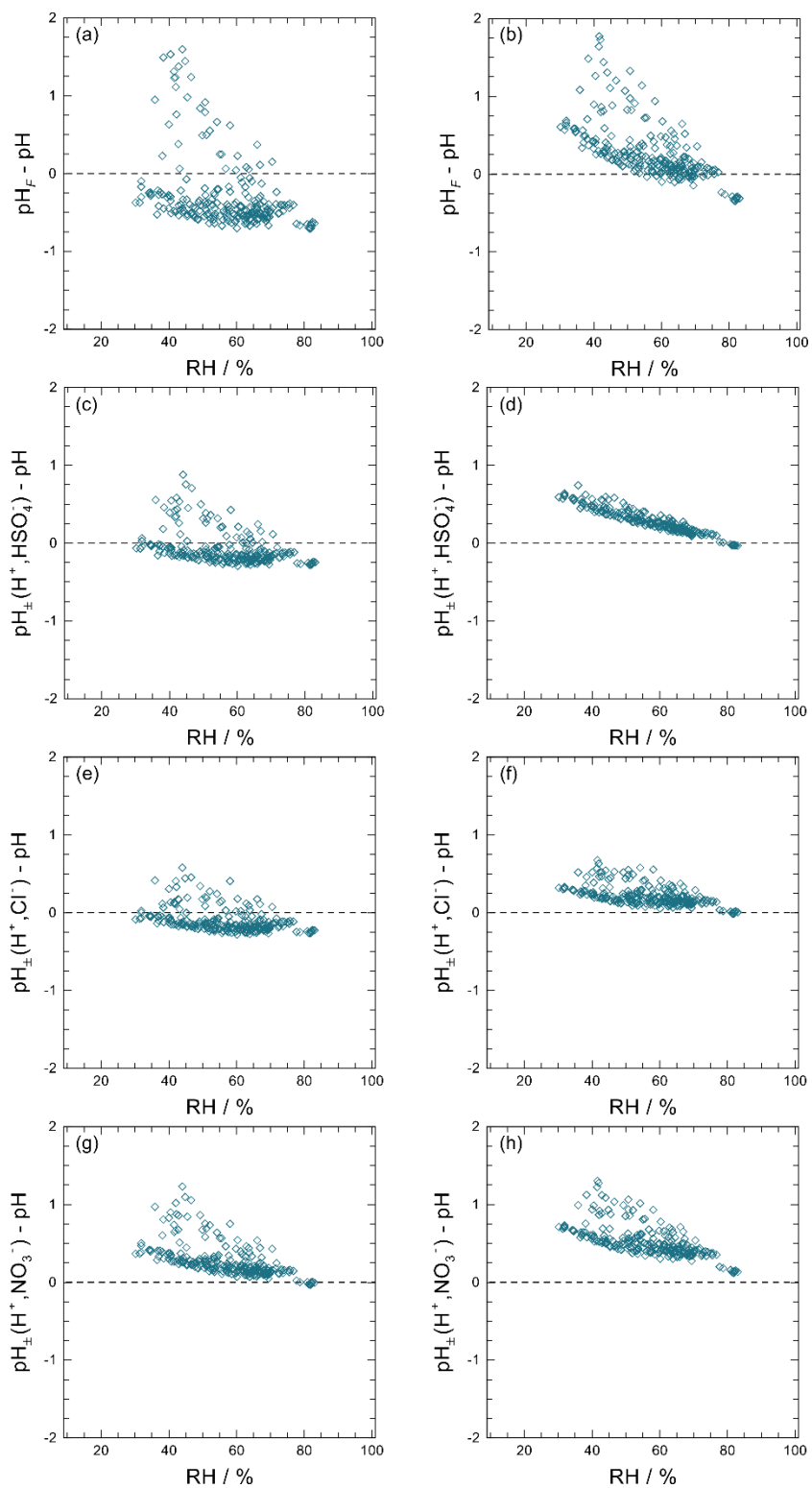
239 column) and E-AIM (right column).

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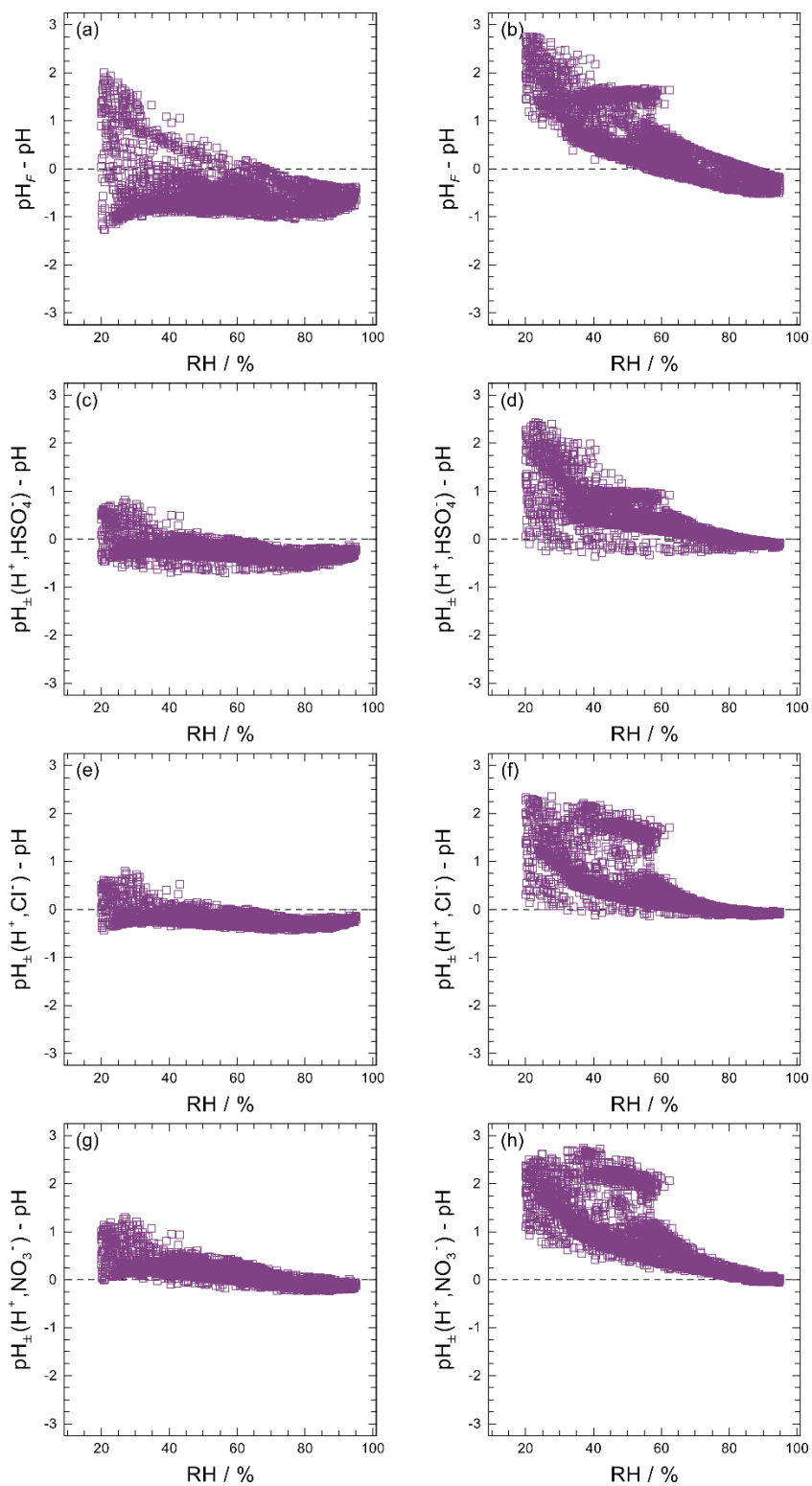
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**Figure S3.** Comparison of different metrics of calculating pH using the SOAS data for AIOMFAC-GLE (left column) and E-AIM (right column).



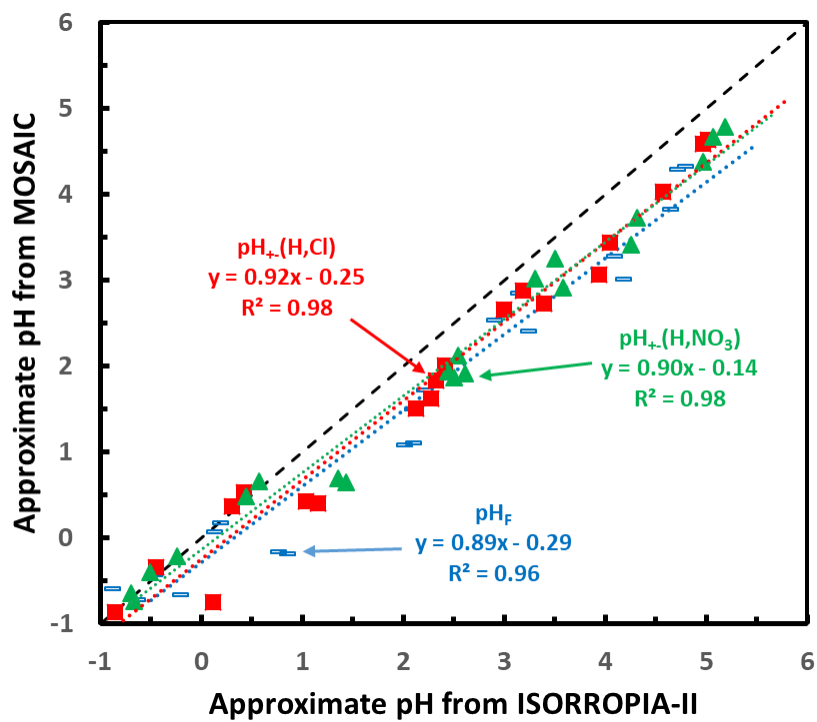
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**Figure S4.** Comparison of different metrics of calculating pH using the Tianjin data for AIOMFAC-GLE (left column) and E-AIM (right column).



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**Figure S5.** Comparison of different metrics of calculating pH using the WINTER data for AIOMFAC-GLE (left column) and E-AIM (right column).

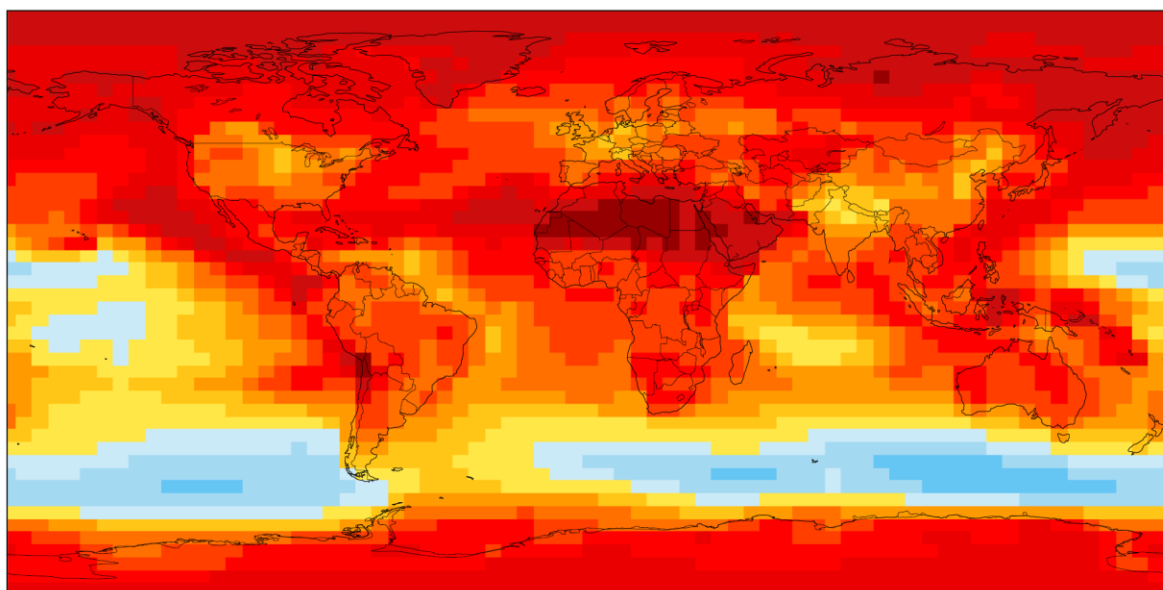


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**Figure S6.** MOSAIC-calculated pH using the different metrics plotted against the corresponding pH values calculated using ISORROPIA II for the data presented in Table S7. Each pH metric is presented with a distinct symbol, and the corresponding linear fit to the data is shown in the corresponding symbol colour.

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(a) Fine aerosol  $pH_F$  without NVC from dust

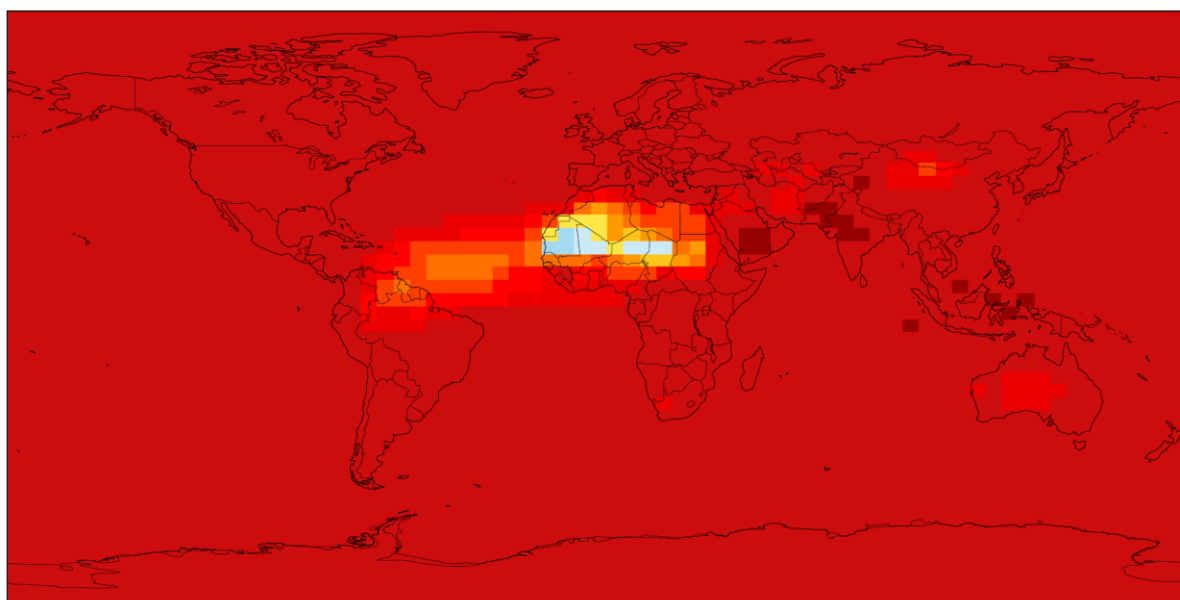


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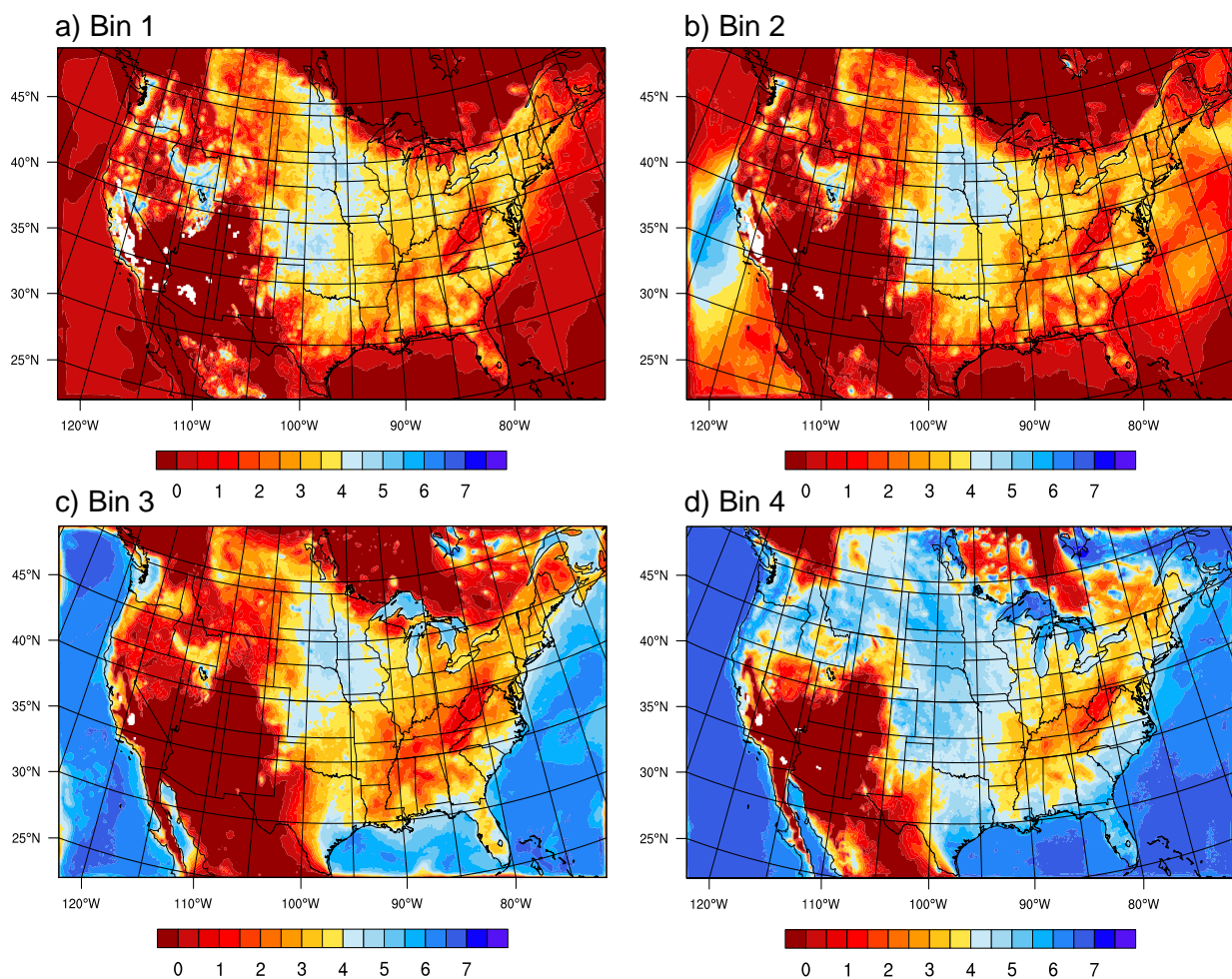
(b) Change in fine aerosol  $pH_F$  due to NVC from dust



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269 **Figure S7.** (a) GEOS-Chem annual-mean predicted fine aerosol  $pH_F$  excluding non-volatile cations (NVCs) from  
270 dust (default GEOS-Chem approach). (b) Increase in  $pH_F$  from the inclusion of NVCs from dust. Main text GEOS-  
271 Chem figures include NVC from dust in calculations of fine aerosol  $pH_F$ .



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**Figure S8.** LWC-weighted surface aerosol pH as predicted by the MOSAIC aerosol scheme in WRF-Chem for four size bins. (a) Bin 1 for 0.039-0.156  $\mu\text{m}$  diameter, (b) Bin 2 for 0.156-0.625  $\mu\text{m}$  diameter, (c) Bin 3 for 0.625-2.5  $\mu\text{m}$  diameter, and (d) Bin 4 for 2.5-10.0  $\mu\text{m}$  diameter.

278 **Supplementary Tables**

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280 **Table S1.** Reaction rates for the S(IV) –S(VI) conversion processes in Figure 3.

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Rate	Parameters	Reference
$R_{HOOH} = k_1 K_1 \frac{[H^+][HSO_3^-]}{1 + K_1[H^+]} [HOOH]$	$k_1 = 7.2 \times 10^7 \exp(-4000 (\frac{1}{T} - \frac{1}{298})) M^{-1}s^{-1}$ $K_1 = 16 M^{-1}$	McArdle and Hoffmann (1983); Zellner (1994)
$R_{TMI} = R_{Fe} + R_{Mn}$  $R_{Fe} = \begin{cases} \frac{k_2 [Fe^{+3}][S(IV)]}{[H^+](1 + K_2[S(IV)]^{2/3})} & M \text{ for } pH < 3 \\ k_3 [Fe^{+3}]^2 [S(IV)] & \text{for } 3 \leq pH < 5 \\ k_4 [S(IV)] & \text{for } pH \geq 5 \end{cases}$  $R_{Mn} = \begin{cases} k_5 [Mn^{+2}][S(IV)] & \text{for } pH < 6 \\ 680 [H^+]^2 & \text{for } pH \geq 6 \end{cases}$	$k_2 = 6 s^{-1}$ $K_2 = 150 M^{-2/3}$ $k_3 = 10^9 M^{-2} s^{-1}$ $k_4 = 10^{-3} s^{-1}$ $k_5 = 10^3 M^{-1} s^{-1}$ $k_6 = 680 M^{-1} s^{-1}$	Martin and Hill (1987); Martin et al. (1991)
$R_{O_3} = (k_7 [SO_2 \cdot H_2O] + k_8 [HSO_3^-] + k_9 [SO_3^{-2}]) [O_3]$	$k_7 = 2.4 \times 10^4 M^{-1} s^{-1}$ $k_8 = 3.7 \times 10^5 \exp(-5530 (\frac{1}{T} - \frac{1}{298})) M^{-1} s^{-1}$ $k_9 = 1.5 \times 10^9 \exp(-5280 (\frac{1}{T} - \frac{1}{298})) M^{-1} s^{-1}$	Hoffmann (1986)
$R_{NO_2} = k_{10} [NO_2][S(IV)]$	$k_{10} = \begin{cases} 1.4 \times 10^5 M^{-1} s^{-1} & \text{for } pH < 5.3 \\ 1.24 \times 10^7 M^{-1} s^{-1} & \text{for } pH = 5.3 \\ 1.6 \times 10^7 M^{-1} s^{-1} & \text{for } pH = 8.7 \\ \text{linear interpolation for } 5.3 < pH < 8.7 \end{cases}$	Lee and Schwartz (1983); Clifton (1988); Cheng et al. (2016)

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284 **Table S2.** Henry's Law constants (Sander 2015) used for the S(IV) –S(VI) conversion processes in Figure 3.  
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Henry's Law Constant	Value
$H_{\text{SO}_2}$	$1.317225 \exp\left(2900\left(\frac{1}{T} - \frac{1}{298}\right)\right) \text{ M atm}^{-1}$
$H_{\text{NO}_2}$	$1.22 \times 10^{-2} \exp\left(2400\left(\frac{1}{T} - \frac{1}{298}\right)\right) \text{ M atm}^{-1}$
$H_{\text{O}_3}$	$1.01 \times 10^{-2} \exp\left(2800\left(\frac{1}{T} - \frac{1}{298}\right)\right) \text{ M atm}^{-1}$
$H_{\text{HOOH}}$	$8.41 \times 10^4 \exp\left(7600\left(\frac{1}{T} - \frac{1}{298}\right)\right) \text{ M atm}^{-1}$

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**Table S3.** Predictions of molality-based pH and related  $H^+$  properties for system 1, water +  $(\text{NH}_4)_2\text{SO}_4 + \text{H}_2\text{SO}_4 + \text{NH}_3$  at 298.15 K. Model calculations include the partial dissociation of  $\text{HSO}_4^-$  and the gas–liquid equilibria of  $\text{NH}_3$  and water.

RH	E-AIM, model III			AIOMFAC–GLE			MOSAIC			ISORROPIA II			EQUISOLV II		
	$m_{\text{H}^+}$ (mol kg <sup>-1</sup> )	$\gamma_{\text{H}^+}$	pH	$m_{\text{H}^+}$ (mol kg <sup>-1</sup> )	$\gamma_{\text{H}^+}$	pH	$m_{\text{H}^+}$ (mol kg <sup>-1</sup> )	$\gamma_{\pm}(\text{H}^+, \text{HSO}_4^-)$	pH $_{\pm}$	$m_{\text{H}^+}$ (mol kg <sup>-1</sup> )	$\gamma_{\pm}(\text{H}^+, \text{HSO}_4^-)$	pH $_{\pm}$	$m_{\text{H}^+}$ (mol kg <sup>-1</sup> )	$\gamma_{\pm}(\text{H}^+, \text{Cl}^-)$	pH $_{\pm}$
<i>moderately acidic; water-free input composition: 99.9% ammonium sulfate by mass; higher NH<sub>3</sub>(g)</i>															
99%	2.466×10 <sup>-4</sup>	4.681×10 <sup>-1</sup>	<b>3.94</b>	2.900×10 <sup>-4</sup>	3.634×10 <sup>-1</sup>	<b>3.98</b>	2.873×10 <sup>-4</sup>	4.331×10 <sup>-1</sup>	<b>3.91</b>	5.740×10 <sup>-5</sup>	5.852×10 <sup>-1</sup>	<b>4.47</b>	1.556×10 <sup>-4</sup>	6.061×10 <sup>-1</sup>	<b>4.03</b>
90%	6.359×10 <sup>-3</sup>	1.336×10 <sup>-1</sup>	<b>3.07</b>	5.639×10 <sup>-3</sup>	1.054×10 <sup>-1</sup>	<b>3.23</b>	6.504×10 <sup>-3</sup>	2.284×10 <sup>-1</sup>	<b>2.83</b>	6.221×10 <sup>-4</sup>	6.015×10 <sup>-1</sup>	<b>3.43</b>	1.675×10 <sup>-3</sup>	4.808×10 <sup>-1</sup>	<b>3.09</b>
80%	1.636×10 <sup>-2</sup>	9.445×10 <sup>-2</sup>	<b>2.81</b>	1.208×10 <sup>-2</sup>	7.936×10 <sup>-2</sup>	<b>3.02</b>	1.591×10 <sup>-2</sup>	1.941×10 <sup>-1</sup>	<b>2.51</b>	1.562×10 <sup>-3</sup>	7.658×10 <sup>-1</sup>	<b>2.92</b>	1.799×10 <sup>-3</sup>	5.879×10 <sup>-1</sup>	<b>2.98</b>
70%	2.738×10 <sup>-2</sup>	8.258×10 <sup>-2</sup>	<b>2.65</b>	1.624×10 <sup>-2</sup>	8.042×10 <sup>-2</sup>	<b>2.88</b>	2.538×10 <sup>-2</sup>	2.003×10 <sup>-1</sup>	<b>2.29</b>	2.732×10 <sup>-3</sup>	9.249×10 <sup>-1</sup>	<b>2.60</b>	1.424×10 <sup>-3</sup>	7.607×10 <sup>-1</sup>	<b>2.97</b>
60%	3.928×10 <sup>-2</sup>	7.733×10 <sup>-2</sup>	<b>2.52</b>	1.814×10 <sup>-2</sup>	9.192×10 <sup>-2</sup>	<b>2.78</b>	3.577×10 <sup>-2</sup>	2.110×10 <sup>-1</sup>	<b>2.12</b>	4.223×10 <sup>-3</sup>	1.100	<b>2.33</b>	9.985×10 <sup>-4</sup>	1.020	<b>2.99</b>
50%	5.261×10 <sup>-2</sup>	7.408×10 <sup>-2</sup>	<b>2.41</b>	1.918×10 <sup>-2</sup>	1.074×10 <sup>-1</sup>	<b>2.69</b>	4.719×10 <sup>-2</sup>	2.205×10 <sup>-1</sup>	<b>1.98</b>	6.210×10 <sup>-3</sup>	1.313	<b>2.09</b>	6.429×10 <sup>-4</sup>	1.418	<b>3.04</b>
40%	6.883×10 <sup>-2</sup>	7.053×10 <sup>-2</sup>	<b>2.31</b>	2.195×10 <sup>-2</sup>	1.136×10 <sup>-1</sup>	<b>2.60</b>	5.861×10 <sup>-2</sup>	2.369×10 <sup>-1</sup>	<b>1.86</b>	9.091×10 <sup>-3</sup>	1.607	<b>1.84</b>	4.017×10 <sup>-4</sup>	1.989	<b>3.10</b>
<i>highly acidic; water-free input composition: 50% ammonium sulfate by mass; moderate NH<sub>3</sub>(g)</i>															
99%	1.447×10 <sup>-2</sup>	4.910×10 <sup>-1</sup>	<b>2.15</b>	1.717×10 <sup>-2</sup>	3.825×10 <sup>-1</sup>	<b>2.18</b>	1.760×10 <sup>-2</sup>	4.529×10 <sup>-1</sup>	<b>2.10</b>	4.890×10 <sup>-3</sup>	5.864×10 <sup>-1</sup>	<b>2.54</b>	1.084×10 <sup>-2</sup>	6.136×10 <sup>-1</sup>	<b>2.18</b>
90%	2.47×10 <sup>-1</sup>	1.89×10 <sup>-1</sup>	<b>1.33</b>	2.582×10 <sup>-1</sup>	1.312×10 <sup>-1</sup>	<b>1.47</b>	2.504×10 <sup>-1</sup>	2.847×10 <sup>-1</sup>	<b>1.15</b>	5.504×10 <sup>-2</sup>	6.161×10 <sup>-1</sup>	<b>1.47</b>	7.329×10 <sup>-2</sup>	5.404×10 <sup>-1</sup>	<b>1.40</b>
80%	4.69×10 <sup>-1</sup>	1.72×10 <sup>-1</sup>	<b>1.09</b>	4.629×10 <sup>-1</sup>	1.123×10 <sup>-1</sup>	<b>1.28</b>	5.018×10 <sup>-1</sup>	2.629×10 <sup>-1</sup>	<b>0.88</b>	1.400×10 <sup>-1</sup>	7.976×10 <sup>-1</sup>	<b>0.95</b>	7.065×10 <sup>-2</sup>	6.798×10 <sup>-1</sup>	<b>1.32</b>
70%	5.85×10 <sup>-1</sup>	1.93×10 <sup>-1</sup>	<b>0.95</b>	5.527×10 <sup>-1</sup>	1.260×10 <sup>-1</sup>	<b>1.16</b>	6.425×10 <sup>-1</sup>	2.815×10 <sup>-1</sup>	<b>0.74</b>	2.455×10 <sup>-1</sup>	9.738×10 <sup>-1</sup>	<b>0.62</b>	5.225×10 <sup>-2</sup>	8.939×10 <sup>-1</sup>	<b>1.33</b>
60%	5.92×10 <sup>-1</sup>	2.49×10 <sup>-1</sup>	<b>0.83</b>	5.664×10 <sup>-1</sup>	1.568×10 <sup>-1</sup>	<b>1.05</b>	7.228×10 <sup>-1</sup>	2.916×10 <sup>-1</sup>	<b>0.68</b>	3.793×10 <sup>-1</sup>	1.169	<b>0.35</b>	3.445×10 <sup>-2</sup>	1.217	<b>1.38</b>
50%	4.89×10 <sup>-1</sup>	4.00×10 <sup>-1</sup>	<b>0.71</b>	5.578×10 <sup>-1</sup>	1.971×10 <sup>-1</sup>	<b>0.96</b>	7.654×10 <sup>-1</sup>	2.750×10 <sup>-1</sup>	<b>0.68</b>	5.565×10 <sup>-1</sup>	1.408	<b>0.11</b>	2.104×10 <sup>-2</sup>	1.710	<b>1.44</b>
40%	3.57×10 <sup>-1</sup>	8.26×10 <sup>-1</sup>	<b>0.53</b>	5.997×10 <sup>-1</sup>	2.204×10 <sup>-1</sup>	<b>0.88</b>	6.284×10 <sup>-1</sup>	2.207×10 <sup>-1</sup>	<b>0.86</b>	8.116×10 <sup>-1</sup>	1.735	<b>-0.15</b>	1.247×10 <sup>-2</sup>	2.431	<b>1.52</b>

293 <sup>a</sup> E-AIM was run using model III and the comprehensive calculation mode including  $\text{NH}_3$  equilibration; no solids considered. It predicts mole-  
 294 fraction-based activity coefficients of  $\text{H}^+$ ;  $\text{pH}_x$  was converted to pH using Eq. (4).  
 295 <sup>b</sup> The AIOMFAC–GLE model was used, including gas–liquid partitioning of  $\text{NH}_3$ ; no solids.  
 296 <sup>c</sup> MOSAIC calculations were run with dynamic gas–particle of  $\text{NH}_3$  considered; no solids.  $\text{pH} \approx \text{pH}_{\pm}$  predicted by use of the listed mean molal ion  
 297 activity coefficients (Eq. 7).  
 298 <sup>d</sup> Models ISORROPIA II and EQUISOLV II approximate  $\text{pH} \approx \text{pH}_{\pm}(\text{H}, \text{X})$  by use of the listed mean molal ion activity coefficients (Eq. 7), with X  
 299 =  $\text{Cl}^-$ ,  $\text{HSO}_4^-$  or  $\text{NO}_3^-$ , as indicated by the  $\gamma_{\pm}(\text{H}^+, \text{X})$  used.

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316 **Table S6.** Observationally-constrained estimates of aerosol acidity. In most cases  $\text{pH}_F$  is reported although there are  
 317 some exceptions (e.g. E-AIM predictions are usually pH).  
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Location	Altitude (m)	Latitude (°N)	Latitude (°E)	Time	Aerosol Size	n	Mean (pH)	$\sigma$ (pH)	Min (pH)	Max (pH)	Method	Reference
<b>Eastern United States</b>												
Chicago, USA		41.93	-87.72	Jan & Jul 2011-2015	PM2.5	46			1.4	3.65	ISORROPIA (forward, aerosol and $\text{NH}_3$ , no $\text{HNO}_3$ input, no organics, internally mixed aerosol)	Battaglia et al., 2017
Chicago, USA rural		41.93	-87.72	Jul 2011-2015	PM2.5				1.6	2.6	ISORROPIA (forward, aerosol and $\text{NH}_3$ , no $\text{HNO}_3$ input, no organics, internally mixed aerosol)	Battaglia et al., 2017
Chicago, USA rural		41.93	-87.72	Jan 2011-2015	PM2.5				3.4	3.65	ISORROPIA (forward, aerosol and $\text{NH}_3$ , no $\text{HNO}_3$ input, no organics, internally mixed aerosol)	Battaglia et al., 2017
Pellston, MI, USA		45.56	-84.86	Jul 2016	0.4-2.5 $\mu\text{m}$		3.5				pH indicator paper/colorimetric image	Craig et al., 2018
Ann Arbor, MI, USA		42.28	-83.73	Aug 2016	0.4-2.5 $\mu\text{m}$		3.5				pH indicator paper/colorimetric image	Craig et al., 2018
Pittsburgh		40.44	-79.98	7-22 Sep 2002	PM1	3599			-0.5	5.5	Aerosol Inorganics Model (AIM II)	Zhang et al., 2007
North East US (Ohio River Valley and the Adirondack region)		40	-83	1979-1980			0.1	0.6				Ferek et al., 1983
Baltimore, USA		39.06	-76.88	Jan & Jul 2011-2015	PM2.5	86			0.7	2.7	ISORROPIA (forward, aerosol and $\text{NH}_3$ , no $\text{HNO}_3$ input, no organics, internally mixed aerosol)	Battaglia et al., 2017
Baltimore, USA		39.06	-76.88	Jan 2011-2015	PM2.5	86			2.3	2.7	ISORROPIA (forward, aerosol and $\text{NH}_3$ , no $\text{HNO}_3$ input, no organics, internally mixed aerosol)	Battaglia et al., 2017
Baltimore, USA		39.06	-76.88	Jul 2011-2015	PM2.5	86			0.7	2.1	ISORROPIA (forward, aerosol and $\text{NH}_3$ , no $\text{HNO}_3$ input, no organics, internally mixed aerosol)	Battaglia et al., 2017
Hampton, VA, C130 flights	up to 5000m	37.03	-76.35	Feb 2015 - Mar 2015	PM1		0.8	1	-0.5	1.9	ISORROPIA metastable, with $\text{HNO}_3$ , no observed $\text{NH}_3$ used, iterated to reach convergence in $\text{NH}_3$	Guo et al., 2016
Yorkville, Georgia, USA		33.93	-85.05	Jun 2012	PM1		1.1	0.6			ISORROPIA (forward, no $\text{NH}_3$ , pH bias correction)	Guo et al., 2015

Location	Altitude (m)	Latitude (°N)	Latitude (°E)	Time	Aerosol Size	n	Mean (pH)	$\sigma$ (pH)	Min (pH)	Max (pH)	Method	Reference
Yorkville, Georgia, USA		33.93	-85.05	Dec 2012	PM1		1.8	1			ISORROPIA (forward, no NH <sub>3</sub> , pH bias correction)	Guo et al., 2015
Yorkville, GA, USA		33.93	-85.05	Sep 2016 - Oct 2016	PM1		2.2	0.6	0.9	3.8	ISORROPIA (forward, metastable, NH <sub>3</sub> , HNO <sub>3</sub> input, internally mixed)	Nah et al., 2018
Atlanta, GA, USA		33.80	-84.40	Mar- Apr 2015	PM1				1	2	ISORROPIA (forward, iterative, no NH <sub>3</sub> , HNO <sub>3</sub> , HCl data, fine PM internally mixed)	Fang et al., 2017
Atlanta, GA, USA		33.80	-84.40	Mar - Apr 2015	coarse		3.5				ISORROPIA (forward external mixed)	Fang et al., 2017
Atlanta, Georgia, USA (GIT)	30-40 m	33.78	-84.40	Jul 2012	PM1		1.1	0.4			ISORROPIA (forward, no NH <sub>3</sub> , pH bias correction)	Guo et al., 2015
JST, Atlanta, Georgia		33.78	-84.42	Aug 2011	PM1		0.55				ISORROPIA (forward, no NH <sub>3</sub> , pH bias correction)	Guo et al., 2015 (from Vasilakos et al., 2018)
JST, Atlanta, Georgia		33.78	-84.42	May 2012	PM1		1.3	0.7			ISORROPIA (forward, no NH <sub>3</sub> , pH bias correction)	Guo et al., 2015
JST, Atlanta, Georgia, USA		33.78	-84.42	Nov 2012	PM1		2.2	0.9			ISORROPIA (forward, no NH <sub>3</sub> , pH bias correction)	Guo et al., 2015
Atlanta, Georgia, USA (RS)		33.78	-84.39	Sep 2012	PM1		1.3	0.7			ISORROPIA (forward, no NH <sub>3</sub> , pH bias correction)	Guo et al., 2015
Centreville, AL, USA	126 m	32.90	-87.25	Jun 1998-Aug 2013	PM2.5		1.19		0	2	ISORROPIA (forward, no NH <sub>3</sub> , 1 unit lower in the original study due to lack of NH <sub>3</sub> measurements)	Weber et al., 2016 corrected by 1 unit due to NH <sub>3</sub>
Centreville, AL, USA	126 m	32.90	-87.25	1 Jun - 15 Jul 2013	PM1		0.94	0.59	-0.9	2.2	ISORROPIA (forward no NH <sub>3</sub> , no correction)	Guo et al., 2015
Centreville, AL, USA		32.90	-87.25	Jun-Jul 2013	PM2.5		1.3	2.1			AIOMFAC (inorganic only, aerosol-only inputs, including activity coefficient)	Pye et al., 2018
Centreville, AL, USA		32.90	-87.25	Jun -Jul 2013	PM2.5		1.5	1.1			AIOMFAC (equilibrium organic-inorganic, aerosol-only inputs, including activity coefficient)	Pye et al., 2018
Centreville, AL, USA		32.90	-87.25	Jun- Jul 2013	PM2.5		1.1	0.7			ISORROPIA (aerosol+gas inputs)	Pye et al., 2018
Barbados, West Indies, USA		13.10	-58.38	Jul - Aug 1989	0.4-0.8 $\mu$ m				-0.7	0.4	aerosol solution pH calculated from aerosol composition, neutralization by NH <sub>3</sub> and CaCO <sub>3</sub>	Zhu et al., 1992

Location	Altitude (m)	Latitude (°N)	Latitude (°E)	Time	Aerosol Size	n	Mean (pH)	$\sigma$ (pH)	Min (pH)	Max (pH)	Method	Reference
Barbados, West Indies, USA		13.10	-58.38	Jul - Aug 1989	0.8-1.7 $\mu\text{m}$				-0.2	11.6	aerosol solution pH calculated from aerosol composition, neutralization by $\text{NH}_3$ and $\text{CaCO}_3$	Zhu et al., 1992
Southeast US (SENEX) aircraft				summer 2013	PM1		1.4	0.4			ISORROPIA	Xu et al., 2015
Southeast US (SOAS & SCAPE)				1 Jun - 15 Jul 2013 & May 2012- Feb 2013	PM1				0	2	ISORROPIA	Xu et al., 2015
<b>North America excluding Eastern US</b>												
Egbert, ON, Canada		44.23	-79.78	20 Jul - 30 Sep 2012	PM2.5		2.1	approx $\pm 1$	2.5	5	E-AIM Model II (sulfate-nitrate-ammonium; AIM-IC measurements; pH=mole fraction activity coeff*mole fraction H*55.508)	Murphy et al., 2017
Harrow, ON, Canada		42.03	-82.89	15 Jun- 15 Jul 2007	PM2.5		1.6	approx $\pm 1$		>2	E-AIM Model II (sulfate-nitrate-ammonium; AIM-IC measurements; pH=mole fraction activity coeff*mole fraction H*55.508)	Murphy et al., 2017
Colorado, BAO-NOAA, USA	1584m	40.05	-105.01	18 Feb - 12 Mar 2011	various sizes < 1 $\mu\text{m}$		about 2.5	about 1.5	about 1	about 4	E-AIM(Inferred From Measured Phase Partitioning based on HCl, $\text{NH}_3$ and $\text{HNO}_3$ , for various sizes	Young et al., 2013
Pasadena, Los Angeles, CA CALNEX, SW USA		34.14	-118.12	15 May - 15 Jun 2010	PM 1		1.9	0.5	1.4	2.5	ISORROPIA forward, with $\text{NH}_3$ and $\text{HNO}_3$	Guo et al., 2017a
Pasadena, Los Angeles, CA CALNEX, SW USA		34.14	-118.12	1-15 Jun 2010	PM 2.5		2.7	0.3	2.2	3	ISORROPIA forward, internally mixed with seasalt	Guo et al., 2017a
Mexico City, Mexico, T1 site		19.71	-98.98	Mar-Apr 2006	PM 1		3.31	1	1.9	7	ISORROPIA forward $\text{NH}_3 >0$ , $\text{HNO}_3 >0$	Hennigan et al., 2015
Mexico City, Mexico, T1 site		19.71	-98.98	Mar-Apr 2006	PM 1		3.24	1	2	5	E-AIM forward $\text{NH}_3 >0$ , $\text{HNO}_3 >0$	Hennigan et al., 2015
Toronto, Canada		43.658	-79.397	2007-2013	PM 2.5	766	2.6		1.48	4.39	E-AIM I (gas + particles $\text{NH}_3$ , $\text{HNO}_3$ )	Tao et al., 2019
Toronto Canada		43.659	-79.395	2014-2016	PM 2.5	301	2.65		1.35	4.02	E-AIM I (gas + particles $\text{NH}_3$ , $\text{HNO}_3$ )	Tao et al., 2019
Ottawa Canada		45.434	-75.676	2007-2016	PM 2.5	851	2.54		1.18	4.67	E-AIM I (gas + particles $\text{NH}_3$ , $\text{HNO}_3$ )	Tao et al., 2019
Simcoe Canada		42.857	-80.27	2007-2016	PM 2.5	713	2.41		0.84	4.38	E-AIM I (gas + particles $\text{NH}_3$ , $\text{HNO}_3$ )	Tao et al., 2019
Montreal Canada		45.652	73.574	2007-2016	PM 2.5	840	2.35		0.51	4.72	E-AIM I (gas + particles $\text{NH}_3$ , $\text{HNO}_3$ )	Tao et al., 2019

Location	Altitude (m)	Latitude (°N)	Latitude (°E)	Time	Aerosol Size	n	Mean (pH)	$\sigma$ (pH)	Min (pH)	Max (pH)	Method	Reference
Windsor Canada		42.293	-83.073	2007-2010	PM 2.5	256	2.12		0.94	3.2	E-AIM I (gas + particles NH <sub>3</sub> , HNO <sub>3</sub> )	Tao et al., 2019
Windsor Canada		42.293	-83.073	2012-2016	PM 2.5	456	2.35		0.4	3.98	E-AIM I (gas + particles NH <sub>3</sub> , HNO <sub>3</sub> )	Tao et al., 2019
St Anicet Canada		45.121	-74.288	2007-2016	PM 2.5	742	2.51		0.9	4.65	E-AIM II (gas + particles NH <sub>3</sub> , HNO <sub>3</sub> )	Tao et al., 2019
<b>North Pacific</b>												
Oahu, Hawaii, USA		21.366 67	- 157.716 7	4-29 Sep 2000			4.6		2.6	5.3	MOCCA model & measurements	Pszenny et al., 2004
<b>Mainland China</b>												
Beijing, China (PKU)		39.99	116.30	Nov 2015 - Dec 2016	PM2.5		4.2		3	4.9	ISORROPIA forward with NH <sub>3</sub> , HNO <sub>3</sub> , HCl tested with AIM, good agreement with a difference of 0.3 units	Liu et al., 2017
Guangzhou, China	15 m	23.13	113.26	1 Jul - 31 Jul 2013	PM2.5		2.5	0.3	1.4	3.1	E-AIM IV forward with NH <sub>3</sub> , HNO <sub>3</sub> , HCl	Jia et al., 2018
Guangzhou, China	15 m	23.13	113.26	1 Jul- 31 Jul 2013	PM2.5		2.8	0.4	1.55	3.5	ISORROPIA forward mode, no NVC, with NH <sub>3</sub> , HNO <sub>3</sub> , HCl	Jia et al., 2018
Guangzhou, China	15 m	23.13	113.26	1 Jul- 31 Jul 2013	PM2.5		2.6	0.3	1.5	3.1	AIOMFAC	Jia et al., 2018
Beijing	49 m	39.97	116.37	17 Nov- 12 Dec 2014	PM2.5		4.6	0.5	4	5.1	ISORROPIA forward, Ca <sup>2+</sup> and Mg <sup>2+</sup> were not measured, pH values would be increased by 0.1 unit	Song et al., 2018
Beijing	49 m	39.97	116.37	17 Nov- 12 Dec 2014	PM2.5		4	0.4	3.6	4.4	E-AIM forward, Ca <sup>2+</sup> and Mg <sup>2+</sup> were not measured, pH values would be increased by 0.1 unit	Song et al., 2018
Beijing	8m	39.97	116.37	24 Jan - 1 Feb 2013	PM2.1		1.1	0.05	1.1	1.2	AIM-IV, no gases	Tian et al., 2018
Chengdu	20 m	30.66	104.02	5 Jan - 24 Jan 2011 <b>night</b>	PM2.1		-1.2	1.1	-2.3	0.3	E-AIM Model II, no gases	Cheng et al., 2015
Chengdu	20 m	30.66	104.02	5 Jan - 24 Jan 2011 <b>day</b>	PM2.1		0.7	1	-0.2	2.1	E-AIM Model II, no gases	Cheng et al., 2015
Beijing	20 m	40.41	116.68	Oct 2014- Jan 2015	PM2.5		4.70	1.1	3.4	7.6	ISORROPIA (metastable)	He et al., 2018
Beijing	20 m	39.99	116.31	1 Jan- 31 Dec 2014	PM2.5		3.02	1.62	3	4.9	ISORROPIA (forward mode, metastable)	Tan et al., 2018
Beijing	20 m	39.99	116.31	winter 2014	PM2.5		4.11	1.37			ISORROPIA (forward mode, metastable)	Tan et al., 2018
Beijing	20 m	39.99	116.31	fall 2014	PM2.5		3.13	1.2			ISORROPIA (forward mode, metastable)	Tan et al., 2018

Location	Altitude (m)	Latitude (°N)	Latitude (°E)	Time	Aerosol Size	n	Mean (pH)	$\sigma$ (pH)	Min (pH)	Max (pH)	Method	Reference
Beijing	20 m	39.99	116.31	spring 2014	PM2.5		2.12	0.72			ISORROPIA (forward mode, metastable)	Tan et al., 2018
Beijing	20 m	39.99	116.31	summer 2014	PM2.5		1.82	0.53			ISORROPIA (forward mode, metastable)	Tan et al., 2018
Tianjin	22 m	39.11	117.16	25 Dec 2014-2 Jun 2015	PM2.5		4.9	1.4	0.33	13.6	ISORROPIA (forward, metastable)	Shi et al., 2017
Beijing, China		39.99	116.30		PM 1		4.5				ISORROPIA forward, metastable with NH <sub>3</sub> & HNO <sub>3</sub>	Guo et al., 2017b
X'ian, China		34.23	108.89		PM2.5		5		4.6	5.4	ISORROPIA forward, metastable with NH <sub>3</sub> & HNO <sub>3</sub>	Guo et al., 2017b
Beijing		39.98333	116.28	winter (Feb 2017)	PM2.5		4.5	0.7			ISORROPIA forward	Ding et al., 2019
Beijing		39.98333	116.28	spring (Apr - May 2016)	PM2.5		4.4	1.2			ISORROPIA forward	Ding et al., 2019
Beijing		39.98333	116.28	summer (Jul - Aug 2017)	PM2.5		3.8	1.2			ISORROPIA forward	Ding et al., 2019
Beijing		39.98333	116.28	fall (Sep - Oct 2017).	PM2.5		4.3	0.8			ISORROPIA forward	Ding et al., 2019
Guangzhou, China		23.13	113.26	Jul -Sep 2013	PM2.5		2.4	0.3	1.5	3.4	E-AIM-III, open system, with NH <sub>3</sub> , HNO <sub>3</sub> , HCl	Jia et al., 2018
Hohhot, Inner Mongolia, China		40.48	111.41	summer 2014	PM2.5		5				ISORROPIA (forward metastable, no NH <sub>3</sub> )	Wang et al., 2019
Hohhot, Inner Mongolia, China		40.48	111.41	autumn 2014	PM2.5		5.3				ISORROPIA (forward metastable, no NH <sub>3</sub> )	Wang et al., 2019
Hohhot, Inner Mongolia, China		40.48	111.41	winter 2015	PM2.5		5.7				ISORROPIA (forward metastable, no NH <sub>3</sub> )	Wang et al., 2019
Hohhot, Inner Mongolia, China		40.48	111.41	spring 2015	PM2.5		6.1				ISORROPIA (forward metastable, no NH <sub>3</sub> )	Wang et al., 2019
Hohhot, Inner Mongolia, China		40.48	111.41	annual 2014-2015	PM2.5		5.6		1.1	8.4	ISORROPIA (forward metastable, no NH <sub>3</sub> )	Wang et al., 2019
<b>ASIA (excluding mainland China)</b>												
Singapore	20 m	1.30	103.78	Sep - Nov 2011	PM2.5		0.6	0.4	-0.2	1.5	AIM-IV	Behera et al., 2013
Hong Kong (HKUST)		22.34	114.26	24 Jul 1997 - 29 May 1998	PM2.5		0.3	1.3	about -0.9	1.5	AIM-II (for RH $\geq$ 70% range only)	Yao et al., 2007
Hong Kong (HKUST)		22.34	114.26	18 Nov 1996 - 5 Nov 1997	PM2.5		-1	1	-2.5	about 0.4	AIM-II (for RH <70% range only)	Yao et al., 2007
Hong Kong (three sites)		22.34	114.26		PM2.5	25	-0.4	1.5	-2.2	1.1	AIM-II (range only)	Yao et al., 2006, data from Pathak et al., 2003
Hong Kong (three sites)		22.34	114.26		PM2.5	25	3.9		-1.2	7.5	ISORROPIA (gases + aerosols)	Yao et al., 2006

Location	Altitude (m)	Latitude (°N)	Latitude (°E)	Time	Aerosol Size	n	Mean (pH)	$\sigma$ (pH)	Min (pH)	Max (pH)	Method	Reference
Hong Kong (three sites)		22.34	114.26		PM2.5	25	2.3		-1.3	7.3	SCAPE2	Yao et al., 2006
<b>Middle East</b>												
Israel (Tel Aviv)		32.11	34.86	summer 1988-1989			2	2	1.5	2.5	Haze	Ganor et al., 1993
<b>Europe</b>												
Po Valley Italy		45.40	12.20	Mar 2009 - Jan 2010	PM2.5	267	3.1	0.6			E-AIM thermodynamic model 4 (average for three sites, four seasons)	Squizzato et al., 2013
Po Valley Italy		45.40	12.20	spring 2009	PM2.5	73	3.6	0.7			E-AIM thermodynamic model 4 (average for three sites)	Squizzato et al., 2013
Po Valley Italy		45.40	12.20	summer 2009	PM2.5	69	2.3	0.7			E-AIM thermodynamic model 4 (average for three sites)	Squizzato et al., 2013
Po Valley Italy		45.40	12.20	fall 2009	PM2.5	62	3	0.9			E-AIM thermodynamic model 4 (average for three sites)	Squizzato et al., 2013
Po Valley Italy		45.40	12.20	winter 2009-2010	PM2.5	63	3.4	0.5			E-AIM thermodynamic model 4 (average for three sites)	Squizzato et al., 2013
Finokalia, Crete	250 m asl	35.33	25.67	Jun - Nov 2012	PM1	37	1.3	1.1	-0.97	3.75	ISORROPIA (forward, no NH <sub>3</sub> )	Bougiatioti et al., 2016
Finokalia, Crete-Biomass burning influenced	250 m asl	35.33	25.67	Jun - Nov 2012	PM1	7	2.77	0.88			ISORROPIA (biomass burning influenced air, no NH <sub>3</sub> )	Bougiatioti et al., 2016
Cabauw, Netherlands		51.97	4.93	Jul 2012 - Jun 2013	PM 2.5		3.7	0.6			ISORROPIA (Forward mode, gas+aerosol)	Guo et al., 2018
Cabauw, Netherlands		51.97	4.93	Jun - Aug 2013	PM 2.5		3.3	0.5			ISORROPIA (Forward mode, gas+aerosol)	Guo et al., 2018
Cabauw, Netherlands		51.97	4.93	Dec - Feb 2012	PM 2.5		3.9	0.4			ISORROPIA (Forward mode, gas+aerosol)	Guo et al., 2018
<b>South Hemisphere</b>												
Sao Paulo, Brazil		-23.55	-46.63	8 Aug - 5 Sep 2012	size resolved PM				4.1	5.4	E-AIM	Vieira-Filho et al., 2016
Pacific Ocean/South of Australia-ACE1		-47.5	147.5	18 Nov- 11 Dec 1995	fine	19	1		0	2	EQUISOLV	Frilind and Jacobson, 2000
South Ocean Weddel, PEGASO, sea-ice influence zone		-61	-45	9 Jan-24 Jan 2015	PM2.5	3	1.36667	0.75056	0.5	1.8	ISORROPIA (forward mode, no NH <sub>3</sub> )	Dall'Osto et al., 2019
South Ocean, Weddel, PEGASO		-64	-65	24 Jan-9 Feb 2015	PM2.5	3	3.76667	0.41633	3.3	4.1	ISORROPIA (forward mode, no NH <sub>3</sub> )	Dall'Osto et al., 2019

319 **Table S7.** Published aerosol acidity values not used in this study.

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Location	Altitude (m)	Latitude (°N)	Latitude (°E)	Time	Aerosol Size	n	Mean (pH)	$\sigma$ (pH)	Min (pH)	Max (pH)	Method	Reference
Beijing		40.41	116.68	Oct 2014- Jan 2015	PM2.5		7.6	0.1			ISORROPIA (stable state)	He et al., 2018
Xi'an		34.23	108.89	17 Nov - 12 Dec 2012	PM2.5		6.7	1.3	4.14	11	ISORROPIA (forward, gas aerosols)	Wang et al., 2016
Beijing		39.99	116.30	21 Jan - 4 Feb 2015	PM2.5		7.63	0.03	7.6	7.7	ISORROPIA (forward, gas aerosols)	Wang et al., 2016
Hong Kong (TST site)		22.26	114.07	5 Dec - 16 Dec 2000	PM2.5	10					AIM2 (only free H+ is available)	Pathak et al., 2004
Hong Kong (HKUST site)		22.33	114.26	12 Mar - 17 Mar 2002	PM2.5	6					AIM2 (only free H+ is available)	Pathak et al., 2004
Jinan	20 m	36.65	117.01	Apr, Aug, Oct, Dec 2006 & Jan 2007	PM1.8		-1.2		-3	1.5	AIM-II, no gases, no organic acids	Cheng et al., 2011
Jinan	20 m	36.65	117.01	Apr, Aug, Oct, Dec 2006 & Jan 2007	PM1.8		-1.1		-2.5	1	AIM-II, no gases, with organic acids	Cheng et al., 2011
Beijing	285 m asl	40.35	116.30	29 Jun - 2 Aug 2005	PM2.5	25	-0.52	0.62			E-AIM Model II only aerosol input	Pathak et al., 2009
Shanghai	15 m	31.45	121.10	5 May - 15 Jun 2005	PM2.5	40	-0.77	0.67			E-AIM Model II only aerosol input	Pathak et al., 2009
Lanzhou		36.13	103.68	18 Jun - 17 Jul 2006	PM2.5	25	-0.38	0.64			E-AIM Model II only aerosol input	Pathak et al., 2009
Guangzhou	17 m	22.69	113.56	15 May - 27 May 2004	PM2.5	13	0.61	0.71			E-AIM Model II only aerosol input	Pathak et al., 2009
Beijing		40.32	116.32	28 Jan 2005- 28 Apr 2006	PM2.5		0.65	1.08	about -0.6	about 3	E-AIM Model II (two sites averaged), only aerosol input	He et al., 2012
Chongqing		29.57	106.53	28 Jan 2005- 28 Apr 2006	PM2.5		1.45	0.77	about 0.5	about 4	E-AIM Model II (three sites averaged), only aerosol input	He et al., 2012
Beijing	10 m	40.00	116.33	1 Jan - 31 Jan 2013	PM2.5		5.8	0.4	5.4	6.2	ISORROPIA mean of forward and reverse, NH <sub>3</sub> estimated from NO <sub>x</sub>	Cheng et al., 2016 in Song et al., 2018
Hong Kong	20 m	22.34	114.26	21 Oct - 25 Oct 2008	PM2.5	127	0.59	0.43	-0.8	2.4	AIM-III (aerosol only input)	Xue et al., 2011
Hong Kong	20 m	22.34	114.26	6 Nov - 13 Nov 2008	PM2.5	180	-0.45	0.59	-2	0.4	AIM-III (aerosol only input)	Xue et al., 2011
Hong Kong	20 m	22.34	114.26	29 Jun - 3 Jul 2009	PM2.5	213	-0.08	0.81	-1.6	3.2	AIM-III (aerosol only input)	Xue et al., 2011

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323 **Table S8.** Observed cloud acidity (pH) – sorted by region.

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325 (a) Africa

Location	Altitude (m)	Latitude (°N)	Longitude (°E)	Period (mo/yr)	N	Mean (pH)	Median (pH)	Min (pH)	Max (pH)	Reference
<b>Congo</b>										
Mayombé Massif, Dimonika	290	-4	12.5	Nov-Sept 1986-1987	14	4.9				Lacaux et al., 1992
<b>Egypt</b>										
Delta Barrage, Egypt	18	30.2	31.12	2015-2016		7.6				Salem et al., 2017
<b>Namibia</b>										
Namib Desert, Namibia	420	-22.74	15.89	Apr-Oct 1994-1995	7	6.2	6.4	5.6	6.6	Eckardt and Schemenauer, 1998
<b>South Africa</b>										
Cape Columbine, South Africa		-32.83	17.85	Aug 1997	1	6.5				Olivier, 2002
Cape Columbine, South Africa		-32.83	17.85	May 1998	1	7.3				Olivier, 2002
Lepelfontein, South Africa	200	-31.05	17.85	Sept 2000	1	7.2				Olivier and De Rautenbach, 2002
Soutpansberg, South Africa	1004	-22.93	30.36	Aug 2001	1	5.9				Olivier and De Rautenbach, 2002
<b>Morocco</b>										
Boutmezguida, Anti-Atlas Mts., Morocco	1225	29.2	-10.02	Nov-June 2013-2015		7.3		7	8.5	Dodson and Bargach, 2015; Schunk et al., 2018

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327 (b) Antarctica

Location	Altitude (m)	Latitude (°N)	Longitude (°E)	Period (mo/yr)	N	Mean (pH)	Median (pH)	Min (pH)	Max (pH)	Reference
Antarctic	600-1490	-77.82	166.69	Dec 1982	7	5.3		4.9	6.2	Saxena et al., 1985

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329 (c) Asia

Location	Altitude (m)	Latitude (°N)	Longitude (°E)	Period (mo/yr)	N	Mean (pH)	Median (pH)	Min (pH)	Max (pH)	Reference
<b>Bangladesh</b>										
Bangladesh, coastal Bhola		22.17	90.75	2015-2016	15	7.1		6.9	7.3	Ahmed et al., 2018
<b>Mainland China and Hong Kong</b>										
Anning Industrial Zone, China		24.92	102.46	Dec 1988	19	6.12		5.4	7.5	Huang et al., 1992
Beijing, China		39.93	166.28	1998-1999	3	5.3				Jiang et al., 2010
Beijing, China		39.93	166.28	2005-2006	2	6.2				Jiang et al., 2010
Changsha, China		28.22	112.96	1993	13	4.9		3.4	5.6	Shen et al., 1996
Chengdu, China				Sept 1989	48	3.8	4.1	2.9	6.8	Lei et al., 1997
Chengdu, China		32.2	118.71	1989	58	4.6		3.2	6.8	Shen et al., 1996
Chongqing, China		29.43	106.89	Sept-Oct 1985	18	5.85				Lei et al., 1997
Chongqing, China		29.43	106.89	1985	16	6.2		4.9	7.8	Shen et al., 1996
Chongqing, China		29.43	106.89	Oct 1989	39	3.9	4.6	3	6	Lei et al., 1997
Chongqing, China		29.43	106.89	1989	46	4.6		3.2	6.0	Shen et al., 1996

Location	Altitude (m)	Latitude (°N)	Longitude (°E)	Period (mo/yr)	N	Mean (pH)	Median (pH)	Min (pH)	Max (pH)	Reference
Chongqing, China		29.43	106.89	1984-1990	182	4.4		3.0	8	Li et al., 1996; Li and Peng, 1994
Guangzhou, China		23.15	113.29	Mar 1988	29	3.8	3.9	3.4	4.2	Lei et al., 1997
Guangzhou, China		23.15	113.29	1988	47	3.9		3.4	6.3	Shen et al., 1996
Guangzhou, China		23.15	113.29	Oct 1989	41	4.1	4.6	3.4	6	Lei et al., 1997
Guangzhou, China	82	23.13	113.30	Feb-Mar 2005	3	5.7				Wu et al., 2009
Guilin, China		25.24	110.18	Mar-Apr 1988	13	4.1	4.2	3.6	5.7	Lei et al., 1997
Guilin, China		25.24	110.18	1988	20	3.9		3.3	5.3	Shen et al., 1996
Guiyang, China		26.65	106.63	1989	48	4.6		3.4	6.0	Shen et al., 1996
Jinan, China		36.67	117.05	Jan 2013		2.9		2.62	4.2	Wang et al., 2014
Jingdong County, China	2476	24.54	101.03	2015-2016	117		4.1	3.5	6.9	Nieberding et al., 2018
Jinghong, China	582	22.01	100.80	Dec 1997	3	6.3		5.8	6.8	Zhu et al., 2000
Lushan, China	~1250	29.58	116.02	May 1987	60	5.1		4.9	5.4	Ding et al., 1991
Mengyangzhen, China	771	22.09	100.90	Dec 1997	8	8.3		7.9	9.2	Zhu et al., 2000
Mount Heng, Hunan, China	1269	27.3	112.70	Mar-May 2009	194	3.8		2.9	6.9	Sun et al., 2010
Mt. Lu, Jiujiang city, China	1165	29.58	116.00	Aug-Sept 2011	11	3.5		3.2	3.9	Li et al., 2013
Mt. Tai, China	1534	36.3	117.22	Mar-Apr 2007		4.6	3.7	2.6	7.6	Wang et al., 2011
Mt. Tai, China	1545	36.3	117.22	2007-2008	482	4.6	4.3	2.6	7.6	Guo et al., 2012; Liu et al., 2012; Shen et al., 2012
Mt. Tai, China	1545	36.3	117.22	July-Oct 2014	39	5.9		3.8	6.9	Li et al., 2017
Mt. Tai, China	1534	36.3	117.22	June-Aug 2015	17	4.9		3.8	6.3	Zhu et al., 2018
Mt. Tai Mao Shan, China	957	22.4	114.27	Oct-Nov 2016		3.6		3.0	5.9	Li et al. 2019
Nangchang, China		28.68	115.84	1993	51	4.7		3.4	6.5	Shen et al., 1996
Nanjing, China		32.20	118.73	Dec 2001	13	5.6		4.3	7.3	Li et al., 2008
Nanjing, China	22	32.2	118.71	Dec 2006	11	5.7		4.1	7.6	Tang et al., 2008
Nanjing, China	22	32.2	118.71	2006-2007	37	5.9		4.1	7.3	Yang et al., 2012
Nanling Dayaoshan Mountain, China	815	25.083	113.1	Jan 1999	21	6.1				Wu et al., 2004
Nanling Dayaoshan Mountain, China	815	25.083	113.1	Feb-Mar 2001	36	5.2				Wu et al., 2004
Shanghai, China		31.26	121.45	June-Jul 1986	22	5.5	6.2	4.5	7.4	Lei et al., 1997
Shanghai, China		31.26	121.50	1992-1993	80	6.0		4.5	7.8	Minghua and Demin, 1999
Shanghai, China		31.20	121.50	1989-1991	28	5.2		4.3	6.5	Bao et al., 1995
Shanghai, China		31.3	121.48	2009-2010	26	6.0		4.7	6.6	Li et al., 2011
Urumqi, China		43.41	87.27	Mar-May 2003	19	6.35				Xu et al., 2011
Xiamen, China		24.49	118.10	Mar-Apr 1993	5	3.6		2.9	4.5	Liu et al., 1996
Zhanjiang, Donghai Island, China		21.28	110.2	Mar-Apr 2010	19	5.2		4.8	6.1	Xu et al., 2011
Zhoushan, China		30.00	122.21	May 1987	31	6.0		3.8	7.3	Mo et al., 1989

Location	Altitude (m)	Latitude (°N)	Longitude (°E)	Period (mo/yr)	N	Mean (pH)	Median (pH)	Min (pH)	Max (pH)	Reference
<b>India</b>										
Agra, India	169	27.17	78.08	1998-2000	37	7.2		7	7.6	Lakhani et al., 2007
Akaltara, India	283	22.02	82.43	Feb 2011	1	7.5				Ambade, 2014
Bilaspur, India	346	22.09	82.15	Feb 2011	1	7.2				Ambade, 2014
Delhi, India	217	28.62	77.2	Winter 2001-2003	43	6.7		6.6	6.7	Ali et al., 2004
Dongargarh, India	335	21.02	80.08	Feb 2011	1	7.5				Ambade, 2014
Kanpur, India	142	26.5	80.3	Jan-Feb 2010		7.2		6.0	8.1	Kaul et al., 2011
Kanpur, India		26.46	80.33	2012-2014	66	5.4		5.1	7.3	Chakraborty et al., 2016
Korba, India	252	22.35	82.68	Jan-Feb 2011	4	5.6		5	6.4	Ambade, 2014
New Delhi, India	218	28.35	77.12	Winter 2014-2015	24	5.5		5.1	7.0	Nath and Yadav, 2018
Pune region, India	559	18.52	73.86	1983-1985	47	6.6		6.3	7.6	Khemani et al., 1987
Raipur, India	298	21.23	81.63	2010-2011	14	6.8		6.3	7.4	Ambade, 2014
Rajnandgaon, India	307	21.1	81.03	Feb 2011	1	7.3				Ambade, 2014
Sinhagad, India	1450	18.35	73.75	2007-2010	123	6		4.7	7.4	Budhavant et al., 2014
<b>Japan</b>										
Bijodaira, Mt Tateyama, Japan	977	36.58	137.46	Sept-Oct 2009		4.4		3.9	6	Watanabe et al., 2011
Lake Mashu, Japan	542	43.55	144.5	2006-2012	258	4.6				Yamaguchi et al., 2015
Lake Mashu, Japan	542	43.55	144.5	Jul-Oct 2006		4.5				Yamaguchi et al., 2015
Lake Mashu, Japan	542	43.55	144.5	Jul-Oct 2007		4.6				Yamaguchi et al., 2015
Lake Mashu, Japan	542	43.55	144.5	Jul-Oct 2008		4.3				Yamaguchi et al., 2015
Lake Mashu, Japan	542	43.55	144.5	Jul-Oct 2009		4.5				Yamaguchi et al., 2015
Lake Mashu, Japan	542	43.55	144.5	Jul-Oct 2010		5.1				Yamaguchi et al., 2015
Lake Mashu, Japan	542	43.55	144.5	Jul-Oct 2011		4.8				Yamaguchi et al., 2015
Lake Mashu, Japan	542	43.55	144.5	Jul-Oct 2012		4.6				Yamaguchi et al., 2015
Midagahara, Mt Tateyama, Japan	1930	36.57	137.56	Sept-Oct 2009		4.7		3.8	6.2	Watanabe et al., 2011
Mt. Awaga, Aogakicho, Japan	962	35.33	135.03	May-Nov 1999	14	4.1		4	4.5	Aikawa et al., 2006
Mt. Fuji, Japan	3776	35.4	138.7	Sept 2002	34	4.6		4	6.8	Watanabe et al., 2006
Mt. Mokko, Iwate, Japan	1465	39.93	140.85	Aug-Sept 1997	16	4.1		3.6		Ogawa et al., 1999
Mt. Mokko, Iwate, Japan	1465	39.93	140.83	Sept 1998	62	4.4				Adzuhata et al., 2001b
Mt. Mokko, Iwate, Japan	1465	39.93	140.83	June-Sept 1999	20	4.6				Adzuhata et al., 2001a
Mt. Norikura, Japan	3026	36.34	137.52	July 1963	10	3.8		3.4	4.3	Okita, 1968
Mt. Norikura, Japan	3026	36.34	137.52	Aug 1989	2	3.7		3.6	3.8	Qian et al., 1992
Mt. Norikura, Japan	3026	36.34	137.52	July 1990	12	3.8		3.6	4.2	Qian et al., 1992
Mt. Norikura, Japan	2770	36.34	137.52	Aug 1991	55	3.6		3.3	4.3	Minami and Ishizaka, 1996

Location	Altitude (m)	Latitude (°N)	Longitude (°E)	Period (mo/yr)	N	Mean (pH)	Median (pH)	Min (pH)	Max (pH)	Reference
Mt. Norikura, Japan	2770	36.34	137.52	July & Sept 1994	68	4		3.7	6.7	Watanabe et al., 1999
Mt. Norikura, Japan	2770	36.34	137.52	Aug 1995	7	3.3		3.1	3.5	Watanabe et al., 1999
Mt. Norikura, Japan	2770	36.34	137.52	Oct 1996	32	3.7		3.4	5.1	Watanabe et al., 1999
Mt. Oyama, Japan	700	35.56	139.21	1991-1992	1169	4.0	4.3	2.6	6.8	Hosono et al., 1994
Mt. Oyama, Japan	680	35.56	139.21	1995	175	3.7		2.6	6.1	Igawa et al., 1998
Mt. Oyama, Japan	680	35.56	139.21	Aug-Sept 1998		3.3		3.1	5.6	Watanabe et al., 2001a
Mt. Rokko, Kobe City, Japan	931	34.79	135.26	1997-1998	55	3.8		2	5.8	Aikawa et al., 2001
Mt. Rokko, Kobe City, Japan	931	34.79	135.26	June-Nov 1999	14	3.7		3.3	3.9	Aikawa et al., 2006
Mt. Rokko, Kobe City, Japan	931	34.79	135.26	1997-2001	403	3.8	3.8	3.2	4.9	Aikawa et al., 2005
Mt. Tsukuba, Japan	876	36.39	140.12	Nov 1963	5	5.9		5.6	6.5	Okita, 1968
Murododaria, Mt Tateyama, Japan	2450	36.61	137.63	July-Aug 2004	11	4.5		4	5.4	Watanabe et al., 2010; Watanabe et al., 2011
Murododaria, Mt Tateyama, Japan	2450	36.61	137.63	Autumn 2005	14	3.8		3.5	5.5	Watanabe et al., 2010; Watanabe et al., 2011
Murododaria, Mt Tateyama, Japan	2450	36.61	137.63	Sept-Oct 2006	7	4.8		4.5	6.3	Watanabe et al., 2010; Watanabe et al., 2011
Murododaria, Mt Tateyama, Japan	2450	36.61	137.63	Sept-Oct 2007		4.6		4.3	5.5	Watanabe et al., 2011
Murododaria, Mt Tateyama, Japan	2450	36.61	137.63	Sept-Oct 2008		4.0		3.7	4.7	Watanabe et al., 2011
Murododaria, Mt Tateyama, Japan	2450	36.61	137.63	Sept-Oct 2009		4.2		3.8	5.7	Watanabe et al., 2011
Shiobara, Japan		36.97	139.82	July 1963	2	5.9		5.9	5.9	Okita, 1968
<b>Pakistan</b>										
Kala Bagh, Nathiagali, Pakistan		34.3	73.2	July 1996	18	6.3		5.3	6.8	Ghauri et al., 2001
<b>South Korea</b>										
Chongwon, South Korea	39	36.63	127.49	Sept 1994	32	4.6		4.2	6.4	Chung et al., 1999
Chongwon, South Korea	39	36.63	127.49	Apr-Nov 1995	36	5.3		4.9	5.8	Chung et al., 1999
Chongwon, South Korea	39	36.63	127.49	Jan-Oct 1996	45	5.5		4.4	5.8	Chung et al., 1999
Daekwanreung, South Korea	840	37.68	128.45	2002-2003	203	5.2		3.6	6.8	Kim et al., 2006
Mt. Sobaek, South Korea	1340	36.93	128.43	Aug 1995	27	4.4				Nam et al., 2001
<b>Taiwan</b>										
Chilan Mountain, Taiwan	1650	24.59	121.5	Apr-May 2011	36		4.5	3.7	5.2	Simon et al., 2016
Kinmen Weather Station, Taiwan	48	24.41	118.29	Mar-Apr 2014	15		3.0	2.3	3.4	Simon et al., 2016
Lulin Atmospheric Background Station, Taiwan	2826	23.47	120.87	Apr-May 2011	14		3.9	3.4	4.5	Simon et al., 2016
Mt. Bamboo, Taiwan	1050	25.19	121.54	Jan 2011	291		4.2	3.2	6.4	Klemm et al., 2015
Xitou Flux Tower, Taiwan	1150	23.66	120.8	Sept-Nov 2013	69		4.1	3.2	6.0	Simon et al., 2016

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331 (d) Australia

Location	Altitude (m)	Latitude (°N)	Longitude (°E)	Period (mo/yr)	N	Mean (pH)	Median (pH)	Min (pH)	Max (pH)	Reference
Barrington Tops	400	-31.92	151.58	Jan-Apr 1989		5.6				Post et al., 1991
Craigieburn	1600	-37.56	144.93	1985-1986	3	6.4				Verhoeven et al., 1987
Dorrigo	900	-30.33	152.67	Jan-Apr 1989		5.4				Post et al., 1991
Sydney	1500-1800	-34.11	151.28	July-Aug 1976	28	5.9		4.6	6.6	Scott, 1978
Tasmania		-40.68	114.68	1981 & 1983	55	5.4		4.1	7.0	Gillett and Ayers, 1989

332

333 (e) Central and South America

Location	Altitude (m)	Latitude (°N)	Longitude (°E)	Period (mo/yr)	N	Mean (pH)	Median (pH)	Min (pH)	Max (pH)	Reference
<b>Brazil</b>										
São Paulo City, Brazil		-23.56	-46.66	2009	8	5.9		5.0	7.7	Vasconcellos et al., 2018
<b>Chile</b>										
El Tofo, Chile	780	-29.43	-71.25	1987	2	5.2		5.0	5.8	Schemenauer and Cereceda, 1992b
El Tofo, Chile	780	-29.43	-71.25	1987	5	3.6		3.5	5.6	Schemenauer and Cereceda, 1992b
El Tofo, Chile	780	-29.43	-71.25	1987	8	4.5		4.0	6.7	Schemenauer and Cereceda, 1992b
Torres del Paine (+Punta Arenas)	50-400	-51.17	-71.97	1987-1994	22	4.8	5.2	4.2	6.3	Weathers and Likens, 1996
<b>Venezuela</b>										
Altos de Pipe, Miranda State	1747	10.33	-66.92	June-Aug 1989	6	4.9				Gordon et al., 1994
Altos de Pipe, Miranda State	1747	10.33	-66.92	Jun, Oct-Dec 1989	13	5.4		4.8		Sanhueza et al., 1992
Altos de Pipe, Miranda State	1747	10.33	-66.92	Mar-May 1990	10	5.0				Gordon et al., 1994
Pico del Avila, Distrito Federal	2150	10.54	-66.88	June-Aug 1989	19	5.0				Gordon et al., 1994
Pico del Avila, Distrito Federal	2150	10.54	-66.88	Mar-May 1990	51	4.6				Gordon et al., 1994
<b>Costa Rica</b>										
Monteverde, Costa Rica	1470	10.3	-84.4	1988-1992	34	3.9		3.3	5.6	Clark et al., 1998
<b>Ecuador</b>										
El Tiro, Ecuador	2825	-4.00	-79.16	2003-2004	59		4.6	3.9	5.6	Beiderwieden et al., 2005
El Tiro, Ecuador*	2870	-4.00	-79.16	2005-2009		4.7				Giannoni et al., 2013
Cerro del Consuelo, Ecuador*	3180	-4.00	-79.16	2004-2009		5.4				Giannoni et al., 2016
ECSF, Ecuador*	1960	-4.00	-79.16	2005-2009		4.9				Giannoni et al., 2013
TS1, Ecuador*	2660	-4.00	-79.16	2005-2009		5.3				Giannoni et al., 2013
Antenas*	3180	-4.00	-79.16	2005-2009		5.5				Giannoni et al., 2013

334 \*These sampling locations are close to each other, thus the average pH value was calculated and shown in Figure

335 5.2.

336

337 (f) Europe

Location	Altitude (m)	Latitude (°N)	Longitude (°E)	Period (mo/yr)	N	Mean (pH)	Median (pH)	Min (pH)	Max (pH)	Reference
<b>Austria</b>										
Mt. Sonnblick, Austria	3106	47.05	12.96	May & Nov 1991	15	4.5	4.5	3.8	5.2	Brantner et al., 1994
Mt. Sonnblick, Austria	3106	47.05	12.96	Sept 1996		4.7				Hitzenberger et al., 2000
Mt. Sonnblick, Austria	3106	47.05	12.96	Apr-May 1997		5.7				Hitzenberger et al., 2000

Location	Altitude (m)	Latitude (°N)	Longitude (°E)	Period (mo/yr)	N	Mean (pH)	Median (pH)	Min (pH)	Max (pH)	Reference
Rax, Austria	1644	47.7	15.75	Ap1999 & Mar 2000	12	4.1	3.9	3.36	5.26	Löflund et al., 2002
<b>Czech Rep.</b>										
Beskydy Mts., Czech Rep.	1324	49.53	18.43	May- Apr 2003-2004	48	5.2		3.8	6.7	Skybova, 2006
Cervenhorske sedlo, Czech Rep.	1013	50.12	17.16	1999	12	4.7	4.4	3.9	5.9	Zapletal et al., 2007
Cervenhorske sedlo, Czech Rep.	1013	50.12	17.16	2000	12	5.2	5.8	3.1	6.5	Zapletal et al., 2007
Cervenhorske sedlo, Czech Rep.	1013	50.12	17.16	2001	12	4.8	4.5	3.9	6.0	Zapletal et al., 2007
Cervenhorske sedlo, Czech Rep.	1013	50.12	17.16	2002	12	5.2	4.9	3.9	7.0	Zapletal et al., 2007
Flaje, Krušné Hory Mts., Czech Rep.	740	50.68	13.6	Mar-Mar1995-1996	27	3.0		2.3	4.1	Bridges et al., 2002
Mt. Churanov, Czech Rep.,	1122	49.06	13.61	Mar-Nov 1999-2000	23	4.6	4.4	3.4	6.6	Fisak et al., 2002
Mt. Milesovka, Czech Rep.,	837	50.56	13.9	Sept-Nov1999-2000	141	4.6	4.5	3.4	6.1	Fisak et al., 2002
Mt. Milesovka, Czech Rep.,	837	50.56	13.9	May-June 2006	5	4.1	4.2	3.8	4.7	Fisak et al., 2009; Fisak et al., 2009
Sumava Mts., Czech Rep.	1123	49.06	13.61	Oct-Oct 1989-1992	40	4.2		2.9	7.0	Elias and Tesar, 1994
Sumava Mts., Czech Rep.	1123	49.06	13.61	Feb-Oct 1988-1991	30	3.2	3.67	2.9	6.4	Elias et al., 1995
<b>France</b>										
Mt. Le Donon, Vosges, France	750	48.5	7.2	Mar 1990	4	3.3		2.8	4.7	Lammel and Metzsig, 1991
Near Paris, France	68	48.7	2.2	2012-2013	9	5.2		3.7	6.2	Degeffie et al., 2015
Puy de Dôme, France	1465	48	2	2001-2011	143	5.5	5.6	3.1	7.6	Deguillaume et al., 2013
Puy de Dôme, France	1465	48	2	2010-2013	23	5.6	5.5	3.9	7.1	Wirgot et al., 2017
Strasbourg, France	145	48.58	7.77	Feb-Nov 1991	31	3.8	3.7	2.8	5.8	Millet et al., 1997
Strasbourg, France	145	48.58	7.77	Jan-Dec 1992	21	3.9	3.8	2.3	6.2	Millet et al., 1996; Millet et al., 1997
Strasbourg, France	145	48.58	7.77	Oct-Jan 1993-1994	7	5.2	5.5	2.4	6.3	Millet et al., 1997
Strasbourg, France	145	48.58	7.77	1991-1999	54	4.6	4.7	2.3	6.6	Herckes et al., 2002
<b>Germany</b>										
Bavarian Alps, Germany	1780	47.52	11.15	Mar-May 1985-1986	104	4.5	4.9	3.9 <sup>*</sup>	6.0 <sup>#</sup>	Munzert, 1988
Bayreuth, Germany	≈350	≈49.95	≈11.58	Sept-Oct 1987	2	5.0		4.7	5.2	Trautner et al., 1989
Collmburg, Germany	316	51.3	13.01	~1960	9	4.2				Mrose, 1966
Kap Arkona, Germany	46	54.67	13.44	~1960	42	3.8				Mrose, 1966
Lugstein, Ore Mt., Germany	880	50.7	13.75	Dec-May 1997-1998	27	4		3.3	5.4	Lange et al., 2003
Mt. Brocken, Germany	1142	51.79	10.67	~1960	19	5.1				Mrose, 1966
Mt. Brocken, Germany	1142	51.79	10.67	1992	35	4.4				Acker et al., 1998a
Mt. Brocken, Germany	1142	51.79	10.67	1993		4.2				Acker et al., 1998a
Mt. Brocken, Germany	1142	51.79	10.67	1994		3.9				Acker et al., 1998a
Mt. Brocken, Germany	1142	51.79	10.67	1995		4.0				Acker et al., 1998a
Mt. Brocken, Germany	1142	51.79	10.67	1996		4.0		2.5	>8	Acker et al., 1998a; Acker et al., 1998b

Location	Altitude (m)	Latitude (°N)	Longitude (°E)	Period (mo/yr)	N	Mean (pH)	Median (pH)	Min (pH)	Max (pH)	Reference
Mt. Brocken, Germany	1142	51.79	10.67	1997		4.3	4.3	3.4	6.8	Möller, 2007; Plessow et al., 2001
Mt. Brocken, Germany	1142	51.79	10.67	1998		4.1				Möller, 2007
Mt. Brocken, Germany	1142	51.79	10.67	1999		4.3				Möller, 2007
Mt. Brocken, Germany	1142	51.79	10.67	2000		4.2				Möller, 2007
Mt. Kleiner Feldberg, Germany	825	50.22	8.44	-		2.8				Schrimpf, 1983
Mt. Kleiner Feldberg, Germany	825	50.22	8.44	1983-1986	250	3.8		2.4	7	Günther Schmitt, 1986
Mt. Kleiner Feldberg, Germany	825	50.22	8.44	Oct-Nov 1990		4.3		3.5	5.9	Wobrock et al., 1994
Mt. Kleiner Feldberg, Germany	825	50.22	8.44	Oct-Nov 1995	114	4.1	4.1	2.7	5	Deutsch et al., 2001
Mt. Kleiner Feldberg, Germany	825	50.22	8.44	Apr-May 1997	25	4.6	4.6	4.1	4.9	Deutsch et al., 2001
Mt. Ochsenkopf, Germany	1024	50.03	11.81	May-June 1985	14	3.7				Verhoeven et al., 1987
Mt. Ochsenkopf, Germany	1024	50.03	11.81	Nov 1987	1	3.4				Trautner et al., 1989
Mt. Schmücke, Germany	937	50.65	10.77	Oct & Nov 2001/2002	22	4.5		4	5.1	Brüggemann et al., 2005
Mt. Schmücke, Germany	937	50.65	10.77	2010	60	4.3	4.6	3.6	5.3	van Pinxteren et al., 2016
Mt. Schöllkopf, Black Forest, Germany	540	48.43	8.39	Dec 1988	5	4.1	4.1	4	4.9	Lammel and Metzsig, 1991
Mt. Waldstein, Germany	776	50.14	11.87	Summer 1997	56	4.3	4.3	3.3	5.7	Wrzesinsky and Klemm, 2000
Mt. Waldstein, Germany	776	50.14	11.87	June-Dec 2000	56	4.1				Thalmann et al., 2002
Mt. Waldstein, Germany	776	50.14	11.87	Apr-Mar 2001-2002	247		4.3	3.3	5.4	Klemm and Wrzesinsky, 2007
Ochsenkopf, Germany	1024	50.0314	11.81	May-June, 1985		3.7				Verhoeven et al., 1987
Taunus Mts, Germany	800	50.22	8.41	Autumn, 1983-1986		3.8		2.3	7.9	Schmitt, 1989
Zinnwald, Germany	877	50.73	13.76	Dec-May 1997-1998	51	4		3.3	4.5	Zimmermann and Zimmermann, 2002
<b>Italy</b>										
Po-Valley, S. Pietro Capofiume, Italy	10	44.65	11.62	Winter 1980-1982	36	4.9	5	2.4	7.5	Fuzzi et al., 1983
Po-Valley, S. Pietro Capofiume, Italy	10	44.65	11.62	Feb & Nov 1984	5	3.8	3.6	3.5	4.3	Fuzzi et al., 1985
Po-Valley, S. Pietro Capofiume, Italy	10	44.65	11.62	Nov 1985	63	3.5		2.5	6.7	Fuzzi, 1988
Po-Valley, S. Pietro Capofiume, Italy	10	44.65	11.62	Nov 1985/1986	10	5.1	5.1	4	6.3	Winiwarter et al., 1988
Po-Valley, S. Pietro Capofiume, Italy	10	44.65	11.62	Feb-Mar 1989	62	5.4	5.2	3.4	7.1	Facchini et al., 1990
Po-Valley, S. Pietro Capofiume, Italy	10	44.65	11.62	Nov-Apr 1989-1990	182	3.5	5.4	3.2	6.9	Fuzzi et al., 1992a; Fuzzi et al., 1992b
Po-Valley, S. Pietro Capofiume, Italy	10	44.65	11.62	Nov-Mar 1992-1993			5.4	3.1	7.0	Fuzzi et al., 1996
Po-Valley, S. Pietro Capofiume, Italy	10	44.65	11.62	Feb-Mar 1994	7	5.4	5.5	2.6	7.0	Fuzzi et al., 1997
Po-Valley, S. Pietro Capofiume, Italy	10	44.65	11.62	1996-1997	17	4.6	4.6	3.1	6.4	Facchini et al., 1999
Po-Valley, S. Pietro Capofiume, Italy	10	44.65	11.62	1990-2000		5.5				Giulianelli et al., 2014
Po-Valley, S. Pietro Capofiume, Italy	10	44.65	11.62	2000-2010		6.5				Giulianelli et al., 2014



Location	Altitude (m)	Latitude (°N)	Longitude (°E)	Period (mo/yr)	N	Mean (pH)	Median (pH)	Min (pH)	Max (pH)	Reference
Po-Valley, S. Pietro Capofiume, Italy	10	44.65	11.62	Nov2013	2		4.6	3.3	5.8	Brege et al., 2018
Vallombrosa, Italy	950	43	11	Nov-May 1992-1995	20	4.2	3.8	3.2	6.2	Cini et al., 2002
<b>Norway</b>										
Bakka, Norway	27	60.77	5	Fall 2011		5.0				Wang et al., 2015
Hakadal, Norway	170	60.12	10.83	Fall 2011		4.7		4.4	5.5	Wang et al., 2015
Sundsbo, Norway	28	60.77	5.2	Fall 2011		5.0				Wang et al., 2015
<b>Poland</b>										
Mt. Sznica, Poland	1332	50.79	15.51	Winter 1993		3.8				Błaś et al., 2008
Mt. Sznica, Poland	1332	50.79	15.51	Summer 1995		4.1	4.4	3.1	6.7	Błaś et al., 2008; Kmiec et al., 1997
Mt. Sznica, Poland	1332	50.79	15.51	Dec-Dec 2005-2006	55	4.6		3.5	7.4	Błaś et al., 2010
Zakopane, Poland	911	49.28	17.97	Dec-Dec 2005-2006	4	5.1		4.6	5.8	Błaś et al., 2010
<b>Spain</b>										
Valencia region, Spain	≈842	≈39.3	≈-0.38	Apr-Dec 2008	71	6.6		5.3	7.5	Corell, 2010
Xistral Mts., Spain	700	43.54	-7.5	Sept-Apr 2011-2012	14	4.5		3.8	5.2	Fernández-González et al., 2014
<b>Sweden</b>										
Areskutan, Sweden	1250	63.43	13.09	Summer 1983-1984	125	4.4				Ogren and Rodhe, 1986
<b>Switzerland</b>										
Bern-Belpmoos, Switzerland	515	46.9	7.51	Oct-Mar 1983-1985	40	4.4	5.6	3.1	6.7	Fuhrer, 1986
Dübendorf, Switzerland		47.40	8.61	Dec 1985	21	2.7	4.2	1.9	6.0	Johnson et al., 1987
Eawag, Dübendorf, Switzerland	440	47.40	8.61	Nov-Dec 1986 & Dec 1987	20	3.3		2.1	6	Capel et al., 1990
Eawag, Dübendorf, Switzerland	440	47.40	8.61	Oct 1989 – Jan 1990	24	3.5		2.5	6.3	Xue et al., 1991; Zuo, 1994
Eawag, Dübendorf, Switzerland	440	47.40	8.61	1991-1992		4.1		3.28	6.17	Kotronarou et al., 1993
Jungfrauoch, Switzerland	3450	46.55	7.98	Oct-Nov 1993	72	4.9				Baltensperger et al., 1998
Mt. Lägeren, Switzerland	682	47.48	8.36	Sept-Dec 1986-1987	97	4.8	4.6	2.7	7.1	Joos and Baltensperger, 1991
Mt. Lägeren, Switzerland	682	47.48	8.36	Summer 2006-2007		7	7.3	5.6	7.5	Michna et al., 2015
Mt. Lägeren, Switzerland	682	47.48	8.36	2007		7.2	7.3	7	8.1	Michna et al., 2015
Mt. Lägeren, Switzerland	682	47.48	8.36	May-Apr 2001-2002		4.3	3.9	3.7	5.8	Burkard et al., 2003
Mt. Rigi, Switzerland	1620	47.06	8.49	May-Dec 1990 & Apr 1991	38	4.6		3.0	6.9	Collett Jr et al., 1993
Niesen Kulm, Switzerland	2330	46.65	7.01	2006-2007		6.4	6.8	6	7.7	Michna et al., 2015
Niesen Schwandegg, Switzerland	1650	46.64	7.67	2006-2007		6.6	6.8	5.8	7.6	Michna et al., 2015
Seeboden, Switzerland	1030	47.07	8.47	1990-1991		5.3		3.8	6.9	Collett Jr et al., 1993
<b>UK</b>										
Dunslair Heights, UK	602	55.68	-3.13	1993-1994		4.1				Fowler et al., 1995
Great Dun Fell, UK	850	54.69	-2.45	1988		3.7	3.7	3.2	4.2	Radojevic et al., 1990
Great Dun Fell, UK	847	54.69	-2.45	Apr-May 1993		4.1	4.2	3.1	5.8	Laj et al., 1997; Sedlak et al., 1997
Holmes Moss, UK	550	53.53	-1.86	1993-1994		4.2				Fowler et al., 1995

Location	Altitude (m)	Latitude (°N)	Longitude (°E)	Period (mo/yr)	N	Mean (pH)	Median (pH)	Min (pH)	Max (pH)	Reference
Plynlimon, UK	390	52	-4.6	1995		5.1		2.9	6.9	Wilkinson et al., 1997

338 Remarks: \*10% percentile, #90% percentile, ~estimated latitude/longitude/height a.m.s.l.

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340 (g) Island/Marine

Location	Altitude (m)	Latitude (°N)	Longitude (°E)	Period (mo/yr)	N	Mean (pH)	Median (pH)	Min (pH)	Max (pH)	Reference
Baengnyeong Island, Yellow Sea	100	37.97	124.62	June-July 2014	11	3.9		3.5	5	Boris et al., 2016
Bering Sea		58.22	-178.29	1998	1	5.5				Sasakawa and Uematsu, 2005
East Peak, Luquillo, Puerto Rico	1051	18.27	-65.75	2004-2005	8	5.8		4.8	6.4	Gioda et al., 2009
East Peak, Luquillo, Puerto Rico	1051	18.27	-65.75	2004-2007	45	5.5		3.5	6.3	Gioda et al., 2013; Gioda et al., 2011
East Peak, Luquillo, Puerto Rico	1051	18.27	-65.75	June-Aug 2007	9	5.6		5.0	7.4	Reyes-Rodríguez et al., 2009
East Peak, Luquillo, Puerto Rico	1051	18.27	-65.75	2010-2012	94	4.9		4.0	7.3	Valle-Díaz et al., 2016
Eastern Pacific Ocean		30.5	-121.5	July 2001	50		4.0	3.3	4.8	Straub et al., 2007
Hilo City, Hawaii		19.74	-155.04	June 1980	6	4.5		4.2	4.7	Parungo et al., 1982
Luquillo Mountains, Puerto Rico	1020	18.32	-65.75	Nov-Dec 1967	7	5.1		4.9	5.4	Lazrus et al., 1970
Luquillo Mountains, Puerto Rico	1050	18.3	-65.78	1986-1987	12	4.6				Asbury et al., 1994
Northeastern North Pacific		49.58	-145	1999	1	4.1				Sasakawa and Uematsu, 2005
Northwestern North Pacific		37.85	142.40	Dec 1996	4	4.1		3.8	5.2	Watanabe et al., 2001b
Northwestern North Pacific		37.85	142.40	July 1998	13	4.2				Sasakawa and Uematsu, 2002
Northwestern North Pacific		44	155	1999	1	3.9				Sasakawa and Uematsu, 2005
Northwestern North Pacific		44	155	2000	21	4.2				Sasakawa and Uematsu, 2005
Northwestern North Pacific		44	155	2001	30	3.7				Sasakawa and Uematsu, 2005
Pico del Ingles, Tenerife, Canary Islands	992	28.5	-16.3	1995 & 1996		3.3		3	4.4	Borys et al., 1998
Puerto Rico	1020	18.32	-65.75	May-Nov 1984-1985		4.7	5.0	4.1	6.9	Weathers et al., 1988
Sea of Japan		39.54	134.28	Feb 1997	1	3.8				Watanabe et al., 2001b
Sea of Japan		39.54	134.28	July 1998	2	2.8				Sasakawa and Uematsu, 2002
Sea of Okhotsk		47.68	145.93	July 1998	5	3				Sasakawa and Uematsu, 2002
Sea of Okhotsk		47.68	145.93	2000	4	4				Sasakawa and Uematsu, 2005
Southeast Pacific		-24	-78	Oct-Nov 2008	72	4.3		2.9	7.2	Benedict et al., 2012
Tenerife, Canary Islands		28.34	-16.64	July 1997	2	3.6		3.7	3.4	Zhang and Anastasio, 2001
Tenerife, Canary Islands		28.34	-16.64	July 1997	1	3.4				Anastasio and McGregor, 2001

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## 345 (h) Middle East

Location	Altitude (m)	Latitude (°N)	Longitude (°E)	Period (mo/yr)	N	Mean (pH)	Median (pH)	Min (pH)	Max (pH)	Reference
Dhofar, Oman	900	17.26	54.28	July 1990	7	6.4		7.0	7.9	Schemenauer and Cereceda, 1992a
Mt. Carmel, Israel	1120	32.74	35.05	1988	1	2				Ganor et al., 1993
Mt. Meron, Israel	341	33.00	35.41	1992	1	2				Ganor et al., 1993
Tel Aviv University		32.11	34.80	1987 & 1989	3	2.1				Ganor et al., 1993

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## 347 (i) North America

Location	Altitude (m)	Latitude (°N)	Longitude (°E)	Period (mo/yr)	N	Mean (pH)	Median (pH)	Min (pH)	Max (pH)	Reference
<b>USA</b>										
Albany, NY		42.66	-73.77	Oct 1982	24	5.6		4.3	6.4	Fuzzi et al., 1984
Angiola, CA	60	35.58	-119.53	2000-2001	36		6.7	5.9	8.0	Herckes et al., 2007
Angora Peak, OR	820	45.79	-123.92	July 1993	6	4.6		4.2	5.0	Rao and Collett, 1995
Appledore Island		42.99	-70.61	1987	4	3.5		3.1	6.5	Jagels et al., 1989
Bakersfield FACT, CA		35.35	-119.09	Jan 1994	2	6.4		6.0	6.6	Collett Jr et al., 1999
Bakersfield, CA		35.37	-119	1982-1983	108	2.9		2.6	7	Jacob et al., 1984
Bakersfield, CA	76	35.37	-119	1983-1984	16	5.7		5.1	6.9	Jacob et al., 1986a
Bakersfield, CA	76	35.37	-119	1984-1985	35	4.0		2.9	7.6	Jacob et al., 1986b
Bakersfield, CA		35.37	-119	Jan 1993	5	6.7		6.4	7.1	Erel et al., 1993
Bakersfield, CA	130	35.37	-119.03	Jan 1994	4	6.7		6.5	6.9	Rao and Collett, 1995
Bakersfield, CA	130	35.37	-119.03	Feb 1995	5	5.5		4.9	6.9	Siefert et al., 1998
Bakersfield, CA	130	35.37	-119.03	Dec 1995	4	6.7		6.3	7.0	Collett Jr et al., 1999
Bar Harbor, ME	10	44.38	-68.23	Aug 1984	3	3.0		2.9	3	Weathers et al., 1986
Bar Harbor, ME	5	44.4	-68.23	1984-1985	15	3.2	3.6	2.4	5.4	Weathers et al., 1988
Bar Harbor, ME	< 10	44.4	-68.23	1984-1985	16	3.2	3.7	2.4	5.4	Kimball et al., 1988
Baton Rouge, LA		30.82	-90.79	2002-2004	15	6.1		4.7	6.7	Raja et al., 2005
Baton Rouge, LA		30.82	-90.79	2004-2005	21	5		2.7	6.4	Raja et al., 2008
Brooklyn, NY		40.68	-73.94	1954	11	4.7		3.5	6.3	Houghton, 1955
Buttonwillow, CA	24	35.40	-119.47	1983-1984	7	5.3		5.0	6.8	Jacob et al., 1986a
Buttonwillow, CA	24	35.40	-119.47	Jan 1985	11	5.9		5.3	5.9	Jacob et al., 1986b
Camels Hump mountain, Vermont		44.86	-72.59	Mar-Sept 1991	47	3.2		2.9	4.5	Hemmerlein and Perkins, 1993
Cape Elizabeth		43.56	-70.20	1987	3	2.8		2.5	3.6	Jagels et al., 1989
Casitas Pass, CA		34.39	-119.42	June 1985	42	4.0		3.3	4.9	Munger, 1989
Casitas Pass, CA	290	34.39	-119.42	June 2015	20	5.9		5.3	6.7	Boris et al., 2018
Catoctin Mountain, MD	860	40.22	-77.42	Jan-May 1987	9	5.3				Anderson and Landsberg, 1979
Charleston, SC	~520	32.78	-79.94	Feb 1982	27	3.6				Daum et al., 1984
Cheeka Peak Observatory, WA	460	48.35	-124.67	May 1993	102	4.2				Vong et al., 1997
Clingman's Dome, TN	2014	35.56	-83.50	June-Oct 1994	9	4.4		3.8	6.1	Anderson et al., 1999; Baumgardner Jr et al., 2003
Clingman's Dome, TN	2014	35.56	-83.50	June-Oct 1995	136	3.9		2.7	6.3	Anderson et al., 1999; Baumgardner Jr et al., 2003
Clingman's Dome, TN	2014	35.56	-83.50	June-Oct 1996	103	3.7		3.1	4.9	Anderson et al., 1999; Baumgardner Jr et al., 2003
Clingman's Dome, TN	2014	35.56	-83.50	June-Oct 1997	318	3.8		2.8	5.9	Anderson et al., 1999; Baumgardner Jr et al., 2003
Clingman's Dome, TN	2014	35.56	-83.50	June-Oct 1998	268	3.6		2.8	5.8	Anderson et al., 1999; Baumgardner Jr et al., 2003
Clingman's Dome, TN	2014	35.56	-83.50	June-Sept 1999	173	3.7		2.7	5.5	Anderson et al., 1999; Baumgardner Jr et al., 2003
Clingman's Peak, NC	1987	35.61	-83.50	1986	48	3.7				Dasch, 1988

Location	Altitude (m)	Latitude (°N)	Longitude (°E)	Period (mo/yr)	N	Mean (pH)	Median (pH)	Min (pH)	Max (pH)	Reference
Columbus, OH		39.96	-82.99	May 1990	17	3.8		3.5	4.1	Burkhard et al., 1994
Corona del Mar, CA		33.60	-117.87	Dec 1982	1	2.2				Jacob et al., 1985
Corvallis, OR		44.5661	-123.26	1988-1989	20	5.6	5.7	4.5	6.8	Muir, 1991
Damariscove Island, ME	<10	43.77	-69.62	July-Oct 1985	4	3.4	4.1	2.9	4.2	Kimball et al., 1988
Damariscove Island		43.7579	-69.62	1987	5	2.7		2.4	3.7	Jagels et al., 1989
Davis, CA		38.32	-121.47	Jan-Feb 1991	6	6.3		5.8	7.3	Sagebiel and Seiber, 1993
Davis, CA		38.32	-121.47	Dec 1998	9	6.1		5.5	6.5	Reilly et al., 2001
Davis, CA		38.32	-121.47	Jan 1999	43	6.3		5.3	6.8	Reilly et al., 2001
Davis, CA	50	38.5	-121.75	1998-1999	51	6.3		5.7	6.9	Moore et al., 2004
Davis, CA		38.54	-121.76	1997-2001	16	6.5	6.7	5.6	7.5	Zhang and Anastasio, 2001
Davis, CA		38.54	-121.63	Jan 2011	11		6.8	6.4	7.2	Ehrenhauser et al., 2012
Del Mar, CA		32.96	-117.27	Jan 1983	5	2.9				Jacob et al., 1985
Delaware Bay		39.31	-75.23	Mar 1992	2	3.1		2.9	3.8	Erel et al., 1993
Douglas Island	800	58.27	-134.5	1984-1985	20	4.5	4.8			Bormann et al., 1989
East coast		34.04	-77.89	Jan-Mar 1986	11	3.9				Barth et al., 1989
Eastern+Western Washington		48.41	-119.68	1982-1983	31	3.9		3.3	5.2	Hegg et al., 1984a; Hegg et al., 1984b
Etiwanda		34.13	-117.52	May 1982	1	2.4				Richards et al., 1983
Fontana		34.10	-117.44	May 1982	3	2.8		2.7	3.4	Richards et al., 1983
Fresh Water Bay, AK (Douglas Island, AK)	25	57.88	-135.17	1984-1985	18	4.6	4.4	3.8	5.3	Weathers et al., 1988
Fresno, CA		36.74	-119.78	Dec 1995	2	6.1		5.9	6.4	Collett Jr et al., 1999
Fresno, CA		36.74	-119.78	Jan 1996	1	6		6		Collett Jr et al., 1999
Fresno, CA		36.83	-119.75	Jan 2010	11		6.7	6.4	7.3	Ehrenhauser et al., 2012
Henninger Flats, CA	780	34.18	-118.1	June 1982	42		2.9	5.4	5.7	Waldman et al., 1985
Henninger Flats, CA	780	34.18	-118.1	May-June 1982	86		3.0	5.4	5.7	Waldman et al., 1985
Henninger Flats, CA	780	34.18	-118.1	May-June 1991	21	2.8		2.5	3.9	Erel et al., 1993
Henninger Flats, CA	780	34.18	-118.1	June-July 1987	76	3.3		2.6	4.8	William Munger et al., 1990
Houston, TX		29.78	-95.12	Feb 2006	11	4.3		3.2	7.2	Suresh Raja et al., 2008
Hubbard Brook, NH	765	43.93	-71.75	Aug 1984	1	3.0				Weathers et al., 1986
Hubbard Brook, NH	765	43.93	-71.75	1984-1985	10	3.9		3.2	4.6	Weathers et al., 1988
Huntington Beach		33.66	-117.99	May 1982	4	3.3		3.1	3.6	Richards et al., 1983
Indianapolis, IN		39.84	-86.17	1985-1986	10	3.4		2.9	4.1	Muir et al., 1986
Isle au Haut, ME	<10	44.03	-68.38	Sept 1985	2	3.6	3.6	3.4	3.8	Kimball et al., 1988
Isle au Haut, ME	<10	44.03	-68.38	1987	11	3.2		2.7	4.0	Jagels et al., 1989
Kearney Agricultural Center, CA		36.60	-119.51	Jan 1991	4	7.0		6.5	7.8	Sagebiel and Seiber, 1993
Kent Island		39.22	-73.34	1954	19	7.2		7	7.4	Houghton, 1955
Kent Island		39.22	-73.34	1987	15	3.5		3.0	5.8	Jagels et al., 1989
Kern Wildlife Refuge, CA		35.76	-119.58	Dec 1995	2	7.0		6.8	7.4	Collett Jr et al., 1999
Kern Wildlife Refuge, CA		35.76	-119.58	Jan 1996	5	7.0		6.9	7.1	Rao and Collett, 1998
La Jolla Peak, CA	475	34.12	-119.05	July 1993	10	3.4		3.1	3.9	Rao and Collett, 1995
La Jolla Peak, CA	475	34.12	-119.05	June 1994	13	3.2		2.8	3.7	Rao and Collett, 1998
Lakes-of-the-clouds, NH	1534	44.27	-71.32	1984-1985	35	3.7	4.0	2.9	4.8	Weathers et al., 1988
Lakes-of-the-clouds, NH	1534	44.27	-71.32	1984-1985	32	3.7	4.0	2.9	4.7	Kimball et al., 1988
Lakes-of-the-clouds, NH	1534	44.27	-71.32	1984-2010	1216		4.1			Murray et al., 2013

Location	Altitude (m)	Latitude (°N)	Longitude (°E)	Period (mo/yr)	N	Mean (pH)	Median (pH)	Min (pH)	Max (pH)	Reference
Lawrence Berkeley Laboratory	246	37.90	-122.2	July-Sept 1986	13	4		3.4	6.2	Gundel et al., 1994
Lennox, CA		33.94	-118.36	Dec 1981	11	2.8		2.7	3.0	William Munger et al., 1983
Lennox, CA		33.94	-118.36	Jan 1983	5	3.6				William Munger et al., 1983
Lennox, CA		33.94	-118.36	Jan 1981	16	3.0		2.7	3.6	Jacob et al., 1985
Loft Mountain, VA	990	38.25	-78.67	Aug 1984	1	3.1				Weathers et al., 1986
Loft Mountain, VA	500	38.25	-78.67	1984-1985	20	3.6	3.8	3.0	5.5	Weathers et al., 1988
Long Beach, CA		33.77	-118.2	Jan 1983	2	4.9				Jacob et al., 1985
Long Beach, CA		33.77	-118.2	May 1992	1	3.0				Richards et al., 1983
Los Angeles		34.06	-118.25	Jan 1980	10	5.3		4.6	4.8	Hegg and Hobbs, 1981
Los Angeles Basin	600-700	33.8	-118	May 1982		3		2.3	3.6	Richards, 1995
Los Angeles Basin	600-700	33.8	-118	June 1984		3		2.8	3.6	Richards, 1995
Los Angeles Basin	600-700	33.8	-118	May-June 1985		3.4		2.9	3.8	Richards, 1995
Los Angeles, 16 sites		34.06	-118.25	Winter 1980-1982	10		3.3	2.7	7.1	Brewer et al., 1983
Lower Kaweah, CA (SNP) <sup>1</sup>	1856	36.57	-118.77	1985-1986	12	5.0		4.4	5.7	Collett Jr et al., 1989
Lower Kaweah, CA (SNP) <sup>1</sup>	1856	36.57	-118.77	1987-1988	70	4.8		3.9	6.5	Collett Jr et al., 1990b
Lower Kaweah, CA (SNP) <sup>1</sup>	1856	36.57	-118.77	Apr 1988	5	4.5		4.4	4.6	Collett Jr et al., 1990a
Marys Peak, OR	1245	44.5	-123.57	June-Nov 1985	14	4.7	5.2			Bormann et al., 1989
Marys Peak, OR	1249	44.43	-123.63	1984-1985	12	4.6	4.7	4.1	5.5	Weathers et al., 1988
McKittrick, CA	262	35.31	-119.62	1983-1984	58	4.0		2.7	5.2	Jacob et al., 1986a
McKittrick, CA	262	35.31	-119.62	Jan 1984	24	4.2		3.7	5.0	Jacob et al., 1986b
Mohonk Mountain, NY	467	41.78	-74.4	Aug 1984	1	2.8				Weathers et al., 1986
Mohonk Mountain, NY	467	41.78	-74.4	1984-1985	23	3.5	3.6	2.8	4.4	Weathers et al., 1988
Monterey, CA		36.6	-121.89	Sept 1987	5	5.6		5.1	6.8	Schomburg et al., 1991
Moosilauke, NH	962	44.02	-71.83	1986-1988	200	3.7				Li and Aneja, 1992; Mohnen and Vong, 1993; Vong and Guttorp, 1991
Moro Rock, CA (SNP) <sup>1</sup>	1965	36.55	-118.77	Apr 1988	9	4.5		4.3	4.8	Collett Jr et al., 1990a
Morro Bay, CA		35.37	-120.85	July 1982	2	6.2				Jacob et al., 1985
Mount Gibbes, (Mt. Mitchell, NC)	2006	35.76	-82.29	Summer 1986	132	3.8		2.7	5.4	Aneja et al., 1992
Mount Gibbes, (Mt. Mitchell, NC)	2006	35.76	-82.29	Autumn 1986	15	3.9		3.3	4.2	Aneja et al., 1992
Mount Gibbes, (Mt. Mitchell, NC)	2006	35.76	-82.29	Summer 1987	39	4.0		3.5	4.9	Aneja et al., 1992
Mount Gibbes, (Mt. Mitchell, NC)	2006	35.76	-82.29	Autumn 1987	13	4.5		4.0	6.7	Aneja et al., 1992
Mount Gibbes, (Mt. Mitchell, NC)	2006	35.76	-82.29	Summer 1988	53	3.4		2.8	4.5	Aneja et al., 1992
Mount Gibbes, (Mt. Mitchell, NC)	2006	35.76	-82.29	Autumn 1988	46	3.9		3.1	5.3	Aneja et al., 1992
Mount Gibbes, (Mt. Mitchell, NC)	2006	35.76	-82.29	June 1993	15	3.1		2.5	3.8	DeFelice, 1997
Mount Gibbes, (Mt. Mitchell, NC)	2006	35.76	-82.29	June-Aug 1996	5	4.0		3.8	4.2	Menon et al., 2000
Mount Lafayette, NH	1220	44.16	-71.64	Aug 1984	1	3.0				Weathers et al., 1986
Mount Washington, NH		44.41	-71.31	1954	35	4.5		3	5.9	Houghton, 1955
Mount Washington, NH	1524	44.41	-71.31	Aug 1984	1	3.0				Weathers et al., 1986
Mt. Desert Rock, ME	<10	43.97	-68.13	July-Sept 1985	5	3.6	3.6	3.3	5.0	Kimball et al., 1988

Location	Altitude (m)	Latitude (°N)	Longitude (°E)	Period (mo/yr)	N	Mean (pH)	Median (pH)	Min (pH)	Max (pH)	Reference
Mt. Elden, AZ	2834	35.24	-111.6	2005-2007	8		6.3	5.1	6.6	Hutchings et al., 2009
Mt. Mitchell, NC	1950	35.74	-82.29	1986-1988	477	3.6				Mohnen and Vong, 1993; Vong and Guttorp, 1991
Mt. Mitchell, NC	2038	35.74	-82.29	May-Sept 1986	149	3.5		2.7	4.8	Lin and Saxena, 1991; Saxena and Lin, 1990
Mt. Mitchell, NC	2038	35.74	-82.29	May-Sept 1987	86	3.5		2.8	5.9	Lin and Saxena, 1991; Saxena and Lin, 1990
Mt. Mitchell, NC	2038	35.73	-82.27	July-Oct 1987	495	3.3		2.9	4.8	Kim and Aneja, 1992
Mt. Mitchell, NC	2038	35.73	-82.27	June-Sept 1988	978	3.4		2.4	5.6	Kim and Aneja, 1992
Mt. Mitchell, NC	2038	35.73	-82.27	May-Aug 1989	413	3.6		2.8	4.9	Kim and Aneja, 1992
Mt. Mitchell, NC	1980	35.77	-82.27	Aug 1993	16	2.9		2.7	3.3	Rao and Collett, 1995
Mt. Mitchell, NC (Polluted)	2038	35.74	-82.29	1993-1994	14	3.2		3.1	3.3	Deininger and Saxena, 1997
Mt. Mitchell, NC (Black Mountain)	2006	35.73	-82.27	Summer 1987	139	3.3		2.9	4.5	Aneja et al., 1990
Mt. Mitchell, NC (Black Mountain)	2006	35.73	-82.27	Fall 1987	47	4.1		3.6	4.5	Aneja et al., 1990
Mt. Mitchell, NC (Continental)	2038	35.74	-82.29	1993-1994	8	3.5		3.1	4.2	Deininger and Saxena, 1997
Mt. Mitchell, NC (Marine)	2038	35.74	-82.29	1993-1994	11	3.3		3.0	3.8	Deininger and Saxena, 1997
Muskegon, MI	670, 1525	43.84	-86.18	Mar 1977	2	3.8				Scott and Laulainen, 1979
Olidale, CA		35.42	-119.02	Jan 1982	3	3.2		2.1	3.1	Munger et al., 1983
Parlier, CA		36.61	-119.53	Jan 1986	5	5.7		5.4	7	Glotfelty et al., 1990
Pasadena, CA		34.14	-118.13	Dec 1981	8	3.2		2.9	5.3	Munger et al., 1983
Pasadena, CA		34.14	-118.13	Jan 1982	1	2.3				Munger et al., 1983
Pomona-Corona, CA		33.88	-117.57	May 1992	2	2.6				Richards et al., 1983
Pt. Reyes, CA		38.05	-113	Aug 1982	17	4.1				Jacob et al., 1985
Puget Sound Basin, WA		47.75	-112.48	Jan-May 1984	17	4.1		3.7	5.3	Hegg and Hobbs, 1986
Redwood National Park, CA	287	41.25	-124.03	1984-1985	13	4.3	4.4	3.7	5.2	Weathers et al., 1988
Riverside, CA	350	33.97	-117.32	Jan-Mar 1986	16	3.3		2.3	5.7	Munger et al., 1990
Roque Island, ME	<10	44.58	-67.53	July-Sept 1985	3	3.8	3.7	3.6	4.9	Kimball et al., 1988
Roque Island, ME		44.58	-67.53	1987	5	3.5		3.3	3.7	Jagels et al., 1989
Rye Harbor, NH		43	-70.75	Mar-Sept 1990	56	3.4		2.6	4.8	Klemm et al., 1994; Klemm et al., 1992
San Joaquin Valley, CA		36.74	-119.79	1983-1984	13	6.1		5.2	6.8	Miller et al., 1987
San Joaquin Valley, CA (BF, KWR, Fresno)		36.74	-119.79	1995-1996	59		6.5	5.0	7.4	Collett Jr et al., 1998
San Marcos Pass, CA		34.51	-119.82	Aug 1983	14	4.5				Jacob et al., 1985
San Nicholas Island, CA		33.25	-119.5	Aug 1982	7	3.9				Jacob et al., 1985
San Pedro Hill, CA	450	33.74	-118.41	June-July 1987	242	3.3		2.4	5.0	Munger et al., 1990
San Pedro Hill, CA	450	33.74	-118.41	June-July 1991	21	2.8		2.2	4.2	Erel et al., 1993
San Pedro Hill, CA	450	33.74	-118.41	June 1992	3	4.3		4.2	4.4	Erel et al., 1993
San Pedro Hill, CA	450	33.74	-118.41	1994	6	3.8		3.2	4.1	Siefert et al., 1998
Seal Beach, CA		33.74	-118.1	Nov 1981	2	3		3.0	3.0	Richards et al., 1983
Shenandoah National Park, VA	1037	38.62	-78.35	Sept 1990	2	3.6		3.3	4.8	Keene et al., 1995

Location	Altitude (m)	Latitude (°N)	Longitude (°E)	Period (mo/yr)	N	Mean (pH)	Median (pH)	Min (pH)	Max (pH)	Reference
Shenandoah, VA	1040	38.2	-78.33	1986-1988	55	3.7				Li and Aneja, 1992; Mohnen and Vong, 1993; Vong and Guttorp, 1991
Mt. Sutro, CA		37.78	-122.41	Aug 1982	1	4.0				Jacob et al., 1985
Steamboat Springs, CO	3220	40.46	-106.74	Jan 1997	27	4.3		3.9	5.1	Collett Jr et al., 2002
Steamboat Springs, CO	3220	40.46	-106.74	Jan 1997	40	5.1		3.9	6.5	Xu et al., 1999
Sugarloaf Mountain, ME	1280	45.03	-70.32	Aug-Oct 1985	14	3.4	3.9	2.9	4.9	Kimball et al., 1988
Susquehanna University, PA	-	40.79	76.88	2007-2015	146	4.7	6.5	3.1	7.4	Straub, 2017; Straub et al., 2012
Tennessee Vally, Whitetop Mountain, VA	1682	36.52	-82.08	Apr-Oct 1986	14	3.6		2.9	4.6	Joslin et al., 1988
Turtleback Dome, CA (YNP) <sup>2</sup>	1590	37.72	-119.71	Apr-Nov 1988	43	4.5		3.8	5.2	Collett Jr et al., 1990b
University of California at Davis		38.54	-121.76	Jan-Feb 1997	2	6.8				Anastasio and McGregor, 2001
University of California at Davis		38.54	-121.76	Jan-Feb 1998	5	6.5				Anastasio and McGregor, 2001
University of California at Davis		38.54	-121.76	Jan 1999	1	6				Anastasio and McGregor, 2001
University of Michigan, Biological Station, MI	2452	45.67	-84.47	Aug 2005	19	4.4		2.2	5.2	Hill et al., 2007
Upland, CA		34.10	-117.65	Jan 1982	3	2.4		2.9	2.2	William Munger et al., 1983
Visalia, CA	43	36.33	-119.29	1983-1984	13	7		5.5	7.2	Jacob et al., 1986a
Western Washington		47.61	-122.33	Apr-May 1979	23	4.3		3.7	6.5	Hegg and Hobbs, 1981
Western Washington		47.61	-122.33	1979-1981	16	4.7		4.3	7	Hegg and Hobbs, 1982
Whiteface Mountain, NY	1500	44.48	-73.9	July-Aug 1976	50	3.6		3.4	4.2	Castillo et al., 1983
Whiteface Mountain, NY	1500	44.48	-73.9	Aug-Sept 1977		3.6		2.7	4.7	Falconer and Falconer, 1980
Whiteface Mountain, NY	1500	44.48	-73.9	Aug-Sept 1979		3.5		2.7	4.7	Falconer and Falconer, 1980
Whiteface Mountain, NY	1500	44.48	-73.9	Summer 1981	114	3.5				Castillo et al., 1985
Whiteface Mountain, NY	1500	44.48	-73.9	Summer 1982	167	3.7				Mohnen and Kadlecck, 1989
Whiteface Mountain, NY	1500	44.48	-73.9	Summer 1982	68	3.9				Castillo et al., 1985
Whiteface Mountain, NY	1500	44.48	-73.9	1983	400	3.6				Mohnen and Kadlecck, 1989
Whiteface Mountain, NY	1500	44.48	-73.9	June-Aug 1983	72	3.7		3.1	4.7	Van Valin et al., 1987
Whiteface Mountain, NY	1500	44.48	-73.9	1984	249	3.5				Mohnen and Kadlecck, 1989
Whiteface Mountain, NY	1500	44.48	-73.9	Winter 1985	38	3.5				Mohnen and Kadlecck, 1989
Whiteface Mountain, NY	1500	44.48	-73.9	1984-1985	10	3.9		3.2	4.6	Weathers et al., 1988
Whiteface Mountain, NY	1483	44.38	-73.08	1986-1988	634	4.1				Li and Aneja, 1992; Mohnen and Vong, 1993; Vong and Guttorp, 1991
Whiteface Mountain, NY	1438	44.38	-73.08	1986-1988	66	3.9				Li and Aneja, 1992; Mohnen and Vong, 1993; Vong and Guttorp, 1991
Whiteface Mountain, NY	1500	44.48	-73.9	1986	375	3.6				Mohnen and Kadlecck, 1989

Location	Altitude (m)	Latitude (°N)	Longitude (°E)	Period (mo/yr)	N	Mean (pH)	Median (pH)	Min (pH)	Max (pH)	Reference
Whiteface Mountain, NY	1500	44.48	-73.9	Summer 1987	91	3.8				Mohnen and Kadlec, 1989
Whiteface Mountain, NY	1512	44.39	-73.09	July 1987	16	3.5		3.1	4.4	Khwaja et al., 1995
Whiteface Mountain, NY	1512	44.39	-73.09	Aug 1987	6	3.4		3.6	4	Khwaja et al., 1995
Whiteface Mountain, NY	1512	44.39	-73.09	July-Aug 1988	10	3.3		3.1	3.7	Husain, 1989
Whiteface Mountain, NY	1512	44.39	-73.09	Aug 1989	2	4.4				Husain et al., 1991; Miller et al., 1993
Whiteface Mountain, NY		44.39	-73.09	July-Aug 1992	73	3.2		2.9	4.5	Dutkiewicz et al., 1996
Whiteface Mountain, NY	1512	44.39	-73.09	June 1993	23	3.6		3.0	4.7	Siefert et al., 1998
Whiteface Mountain, NY		44.39	-73.09	Sept-Oct 1993	73	3.6		3.3	4	Dutkiewicz et al., 1996
Whiteface Mountain, NY	1620	44.39	-73.09	Sept 1993	4	4.6		4.4	4.7	Rao and Collett, 1995
Whiteface Mountain, NY	1512	44.39	-73.09	June-July 1994	26	3.9		3.2	5.4	Siefert et al., 1998
Whiteface Mountain, NY	1483	44.37	-73.90	June-Oct 1994	223	3.6		2.5	5.8	Anderson et al., 1999; Baumgardner Jr et al., 2003
Whiteface Mountain, NY	1483	44.37	-73.90	June-Oct 1995	523	3.7		2.6	6.5	Anderson et al., 1999; Baumgardner Jr et al., 2003
Whiteface Mountain, NY	1483	44.37	-73.90	June 1995	367	3.4		2.9	4.2	Husain et al., 2000
Whiteface Mountain, NY	1483	44.37	-73.90	June-Oct 1996	569	3.8		2.8	5.9	Anderson et al., 1999; Baumgardner Jr et al., 2003
Whiteface Mountain, NY	1483	44.37	-73.90	June-Oct 1997	393	3.7		2.9	6.1	Anderson et al., 1999; Baumgardner Jr et al., 2003
Whiteface Mountain, NY	1483	44.37	-73.90	June-Oct 1998	387	3.8		2.8	5.9	Anderson et al., 1999; Baumgardner Jr et al., 2003
Whiteface Mountain, NY	1483	44.37	-73.90	June-Sept 1999	473	3.7		2.9	6.1	Anderson et al., 1999; Baumgardner Jr et al., 2003
Whiteface Mountain, NY	1484	44.4	-73.85	July 1998	49	3.1		2.9	3.8	Moore et al., 2004
Whiteface Mountain, NY	1484	44.4	-73.85	July-Oct 1994		3.7				Aleksic et al., 2009
Whiteface Mountain, NY	1484	44.4	-73.85	May-Oct 1995		3.7				Aleksic et al., 2009
Whiteface Mountain, NY	1484	44.4	-73.85	June-Oct 1996		4.0				Aleksic et al., 2009
Whiteface Mountain, NY	1484	44.4	-73.85	June-Oct 1997		3.8				Aleksic et al., 2009
Whiteface Mountain, NY	1484	44.4	-73.85	June-Oct 1998		3.7		2.8	4.7	Aleksic et al., 2009; Rattigan et al., 2001
Whiteface Mountain, NY	1484	44.4	-73.85	June-Oct 1999		3.8				Aleksic et al., 2009
Whiteface Mountain, NY	1484	44.4	-73.85	June-Sept 2000		3.9				Aleksic et al., 2009
Whiteface Mountain, NY	1484	44.4	-73.85	June-Oct 2001		4.0				Aleksic et al., 2009
Whiteface Mountain, NY	1484	44.4	-73.85	June-Sept 2002		3.9				Aleksic et al., 2009
Whiteface Mountain, NY	1484	44.4	-73.85	July-Sept 2003		3.9				Aleksic et al., 2009
Whiteface Mountain, NY	1484	44.4	-73.85	July-Aug 2004		4.0				Aleksic et al., 2009



Location	Altitude (m)	Latitude (°N)	Longitude (°E)	Period (mo/yr)	N	Mean (pH)	Median (pH)	Min (pH)	Max (pH)	Reference
Whiteface Mountain, NY	1484	44.4	-73.85	June-Sept 2005		3.7				Aleksic et al., 2009
Whiteface Mountain, NY	1484	44.4	-73.85	May-Sept 2006		3.9				Aleksic et al., 2009
Whiteface Mountain, NY	1484	44.4	-73.85	Aug-Sept 2014	8	4.8		4.1	5.3	Cook et al., 2017
Whitetop Mountain, VA	1689	36.64	-81.61	Summer 1986	32	3.5		2.8	4.3	Reisinger and Imhoff, 1989
Whitetop Mountain, VA	1689	36.64	-81.61	Summer 1987	18	3.2		2.9	3.7	Reisinger and Imhoff, 1989
Whitetop Mountain, VA	1689	36.64	-81.61	1986-1988	601	3.8				Li and Aneja, 1992; Mohnen and Vong, 1993
Whitetop Mountain, VA	1686	36.64	-81.61	June-Oct 1994	141	3.8		2.8	5.4	Anderson et al., 1999; Baumgardner Jr et al., 2003
Whitetop Mountain, VA	1686	36.64	-81.61	June-Oct 1995	550	3.8		2.6	5.2	Anderson et al., 1999; Baumgardner Jr et al., 2003
Whitetop Mountain, VA	1686	36.64	-81.61	June-Oct 1996	181	3.8		2.8	5.5	Anderson et al., 1999; Baumgardner Jr et al., 2003
Whitetop Mountain, VA	1686	36.64	-81.61	June-Oct 1997	501	3.9		2.8	6.1	Anderson et al., 1999; Baumgardner Jr et al., 2003
Whitetop Mountain, VA	1686	36.64	-81.61	June-Oct 1998	271	3.7		2.7	5.3	Anderson et al., 1999; Baumgardner Jr et al., 2003
Whitetop Mountain, VA	1686	36.64	-81.61	June-Sept 1999	143	4.2		2.7	5.4	Anderson et al., 1999; Baumgardner Jr et al., 2003
<b>Canada</b>										
Mt. Tremblant	860	46.21	-74.56	1985	11	4.7				Schemenauer et al., 1995
Mt. Tremblant	860	46.21	-74.56	1986	31	5.0				Schemenauer et al., 1995
Mt. Tremblant	860	46.21	-74.56	1987	79	5.0				Schemenauer et al., 1995
Mt. Tremblant	860	46.21	-74.56	1988	52	4.8				Schemenauer et al., 1995
Mt. Tremblant	860	46.21	-74.56	1989	48	4.7				Schemenauer et al., 1995
Mt. Tremblant	860	46.21	-74.56	1990	75	4.9				Schemenauer et al., 1995
Mt. Tremblant	860	46.21	-74.56	1991	66	5.0				Schemenauer et al., 1995
North Bay		46.33	-79.43	Jan-Feb 1984	148		3.6	2.3	5.5	Isaac and Daum, 1987
Nova Scotia	8	44.68	-63.42	Aug 1975	15	3.4		3	6.2	Bressan and Larson, 1979
Ontario		34.05	-117.6	1988	117	3.4				Liu et al., 1993
Roundtop Ridge	850	45.09	-72.56	July-Dec 1985	2	4.0				Schemenauer, 1986
Roundtop Ridge	845	45.09	-72.56	1986	22	4.6				Schemenauer et al., 1995
Roundtop Ridge	845	45.09	-72.56	1987	52	5.1				Schemenauer et al., 1995
Roundtop Ridge	845	45.09	-72.56	1988	63	5.2				Schemenauer et al., 1995
Roundtop Ridge	845	45.09	-72.56	1989	53	5				Schemenauer et al., 1995
Roundtop Ridge	845	45.09	-72.56	1990	54	5.1				Schemenauer et al., 1995
Roundtop Ridge	845	45.09	-72.56	1991	35	4.9				Schemenauer et al., 1995
Whistler Mountain		50.06	-122.96	2010		4.4				Ervens et al., 2013

348 <sup>1</sup>SNP: Sequoia National Park <sup>2</sup>YNP: Yosemite National Park

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**Table S9.** Select data from the complete datasets used for comparing MOSAIC against ISORROPIA-II. Concentrations are expressed in  $\mu\text{g m}^{-3}$ , temperature in K and relative humidity as percent (%).

ID	Na $\mu\text{g m}^{-3}$	SO <sub>4</sub> $\mu\text{g m}^{-3}$	TNH <sub>4</sub> $\mu\text{g m}^{-3}$	TNO <sub>3</sub> $\mu\text{g m}^{-3}$	Cl $\mu\text{g m}^{-3}$	Ca $\mu\text{g m}^{-3}$	K $\mu\text{g m}^{-3}$	Mg $\mu\text{g m}^{-3}$	RH %	T K	Campaign, Characteristics
1	0.000	1.490	12.851	4.729	0.071	0.000	0.000	0.039	61.5	271.093	Cabauw, High pH, High NO <sub>3</sub> , Low SO <sub>4</sub> , Low RH
2	0.000	8.479	19.306	15.144	0.278	0.000	0.037	0.022	92.9	271.079	Cabauw, High pH, High NO <sub>3</sub> , High SO <sub>4</sub> , High RH
3	0.000	7.964	32.317	16.207	0.074	0.000	0.000	0.000	86.6	271.936	Cabauw, High pH, High NO <sub>3</sub> , High SO <sub>4</sub> , High RH
4	0.000	1.088	49.943	3.851	0.381	0.058	0.000	0.045	71.4	296.264	Cabauw, High pH, High NO <sub>3</sub> , Low SO <sub>4</sub> , Sulfate poor, Intermediate RH
5	0.000	2.037	0.589	0.043	0.010	0.000	0.000	0.0000	88.5	297.3	SOAS, Low pH, High SO <sub>4</sub> , Low NO <sub>3</sub> , High RH
6	0.000	0.847	0.517	0.013	0.020	0.000	0.000	0.000	65.3	293.9	SOAS, Low pH, High SO <sub>4</sub> , Low NO <sub>3</sub> , Low RH
7	0.000	2.150	1.136	0.060	0.010	0.000	0.000	0.000	69.0	300.5	SOAS, Low pH, High SO <sub>4</sub> , Low NO <sub>3</sub> , Low RH
8	0.000	1.040	0.253	0.013	0.007	0.000	0.000	0.000	90.2	294	SOAS, Low pH, High SO <sub>4</sub> , Low NO <sub>3</sub> , High RH
9	0.000	0.776	1.965	1.095	0.603	0.000	0.000	0.000	60.8	288.15	CalNex, Intermediate pH, Comparable SO <sub>4</sub> , NO <sub>3</sub> , Low RH, Low PM
10	0.000	6.430	6.178	33.384	4.355	0.000	0.000	0.000	54.0	298.45	CalNex, Intermediate pH, Comparable SO <sub>4</sub> , NO <sub>3</sub> , Low RH, High PM
11	0.000	3.130	3.672	11.086	0.369	0.000	0.000	0.0000	90.2	286.883	CalNex, Intermediate pH, Comparable SO <sub>4</sub> , NO <sub>3</sub> , High RH, High PM
12	0.000	0.878	0.847	3.751	0.502	0.000	0.000	0.000	90.8	286.85	CalNex, Intermediate pH, Comparable SO <sub>4</sub> , NO <sub>3</sub> , High RH, Low PM
13	0.000	0.059	0.058	0.057	0.000	0.000	0.000	0.000	0.603	277.42	WINTER, Low RH, High SO <sub>4</sub> , low pH
14	0.000	0.001	0.001	0.002	0.000	0.000	0.000	0.000	0.646	264.91	WINTER, Low RH, Low SO <sub>4</sub> , low pH
15	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.868	256.02	WINTER, High RH, High SO <sub>4</sub> , low pH
16	0.000	0.041	0.005	0.096	0.000	0.000	0.000	0.000	0.801	275.51	WINTER, High RH, Low SO <sub>4</sub> , low pH
17	0.041	0.106	2.223	0.202	0.227	0.030	0.030	0.001	0.612	298.25	Tianjin, Low RH, High SO <sub>4</sub>
18	0.032	0.447	2.376	0.539	0.063	0.042	0.020	0.001	0.625	303.15	Tianjin, Low RH, Low SO <sub>4</sub>
19	0.031	0.209	2.035	0.182	0.140	0.034	0.017	0.001	0.595	302.05	Tianjin, High RH, High SO <sub>4</sub>
20	0.026	0.394	2.411	0.809	0.089	0.024	0.013	0.001	0.819	300.25	Tianjin, High RH, Low SO <sub>4</sub>

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355 **Table S10.** Locations used in the box model intercomparison and literature evaluating fine aerosol pH in CTMs.  
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Region/ Season	Specific Location(s)	Time	Observationally- constrained aerosol pH <sub>F</sub>	CMAQ fine aerosol pH <sub>F</sub>	GEOS- Chem fine aerosol pH <sub>F</sub>	WRF- Chem pH <sub>F</sub>
Southeast US Summer	Centreville, AL, USA; various	Summer 2013	0.9 ± 0.6 (a), 1.1 ± 0.4 (b)	1.6 ± 0.5 (i), 0.82 (j), 1.8 ± 1.0 (k), 0.9 ± 0.9 (l)	1.3 (o)	1.5 (r)
Eastern US Winter	Aircraft mission WINTER	February 2015	0.8 ± 1.0 (c)		1.3 (p)	
Los Angeles Basin Summer	Pasadena, CA, USA	May-June 2010	2.7 ± 0.3 (d)	3.0 ± 1.7 (i)		3.4 (r) 3.5 (s)
China Winter	Beijing, China	Winter	4.2 (e)	4.5 ± 0.8 (m)	4.3 ± 0.7 (q)	
China Summer	Tianjin, China	August 2015	3.4 ± 0.5 (f)	3.1 ± 1.5 (n)		
Netherlands	Cabauw	June, July, December 2012-2013	3.7 (g)	3.2 ± 0.7 (n)		
Mediterranean Summer/Fall	Finokalia, Crete	Aug- Nov 2012	1.25 ± 1.14 (h)	0.4 ± 1 (n)		

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 358 Data sources:  
 359 <sup>a</sup>Guo et al. (2015) SOAS Centreville, AL ground site ISORROPIA estimates  
 360 <sup>b</sup>Xu et al. (2016) SENEX aircraft summer 2013 ISORROPIA estimates  
 361 <sup>c</sup>Guo et al. (2016) WINTER 2015 aircraft campaign ISORROPIA estimates  
 362 <sup>d</sup>Guo et al. (2017a) PM<sub>2.5</sub> for the last week of CalNex  
 363 <sup>e</sup>Guo et al. (2017b) Beijing PM<sub>1</sub> ISORROPIA estimates  
 364 <sup>f</sup>Shi et al. (2019) ISORROPIA estimates  
 365 <sup>g</sup>Guo et al. (2018)  
 366 <sup>h</sup>Bougiatioti et al. (2016) excluding water associated with organics. pH=1.38 ± 1.1 including organic water.  
 367 <sup>i</sup>This work, CMAQv5.2 June 2016 predicted mean and standard deviation of hourly predictions (108×108km  
 368 horizontal resolution)  
 369 <sup>j</sup>Vasilakos et al. (2018) CMAQ prediction for summer 2011 Centreville, AL  
 370 <sup>k</sup>Calculated from CMAQ simulations of Pye et al. (2018) for CTR June 2013 including nonvolatile cations  
 371 <sup>l</sup>Calculated from CMAQ simulations of Pye et al. (2018) for CTR June 2013 excluding nonvolatile cations  
 372 <sup>m</sup>This work, CMAQv5.2 February 2016 predicted mean and standard deviation  
 373 <sup>n</sup>This work, CMAQv5.2 annual average 2016  
 374 <sup>o</sup>Marais et al. (2016) SEAC4RS aircraft campaign predicted by GEOS-Chem  
 375 <sup>p</sup>Shah et al., (2018) WINTER 2015 aircraft campaign predicted by GEOS-Chem  
 376 <sup>q</sup>Shao et al. (2018) Beijing Autumn/Winter, mean and range, predicted by GEOS-Chem  
 377 <sup>r</sup>This work, WRF-Chem with MOSAIC LWC-weighted PM<sub>2.5</sub>  
 378 <sup>s</sup>Knote et al. (2014) CALNex, CARES WRF-Chem simulation

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**Table S11.** Select cloud pH measurements (See Table S8) and spatially-corresponding (vertically averaged) cloud pH from CTMs

Location	Latitude (°)	Longitude (°)	Altitude (km)	Time Period (observed)	Cloud/fog pH <sub>F</sub> obs (mean)	Model <sup>1</sup>	Vertical average pH
Delta Barrage, Egypt	30.2	31.12		2015-2016	7.6	CAM-chem CMAQ	5.6 5.3
Mt. Boutmezguida, Morocco	29.2	-10.02	1.23	Nov 2013-Jun 2015	7.3	CAM-chem CMAQ	3.6 6.6
Mt. Tai, China	36.3	117.22	1.55	Jun-Aug 2015	4.9	CAM-chem CMAQ	4.4 4.6
Shanghai, China	31.3	121.48		2009-2010	6.0	CAM-chem CMAQ	4.2 4.3
Fresno, CA	36.83	-119.75		Jan 2010	6.7 (median)	CAM-chem CMAQ WRF-Chem	n/a 6.4 n/a
Whiteface Mtn, NY	44.4	-73.85	1.48	Aug-Sep 2014	4.8	CAM-chem CMAQ WRF-Chem	4.2 4.6 4.6
Po Valley, Italy	44.65	11.62		Nov 2013	4.6 (median)	CAM-chem CMAQ	n/a 5.3
Puy de Dome, France	48	2	1.46	2010-2013	5.6	CAM-chem CMAQ	4.1 5.2
Near Paris, France	48.7	2.2		2012-2013	5.2	CAM-chem CMAQ	4.1 4.6
Sundsbo, Norway	60.77	5.2		Fall 2011	5.0	CAM-chem CMAQ	4.8 4.6
California Marine Stratocumulus	36.98	-123.1	0.08 - 1.0	Summer aircraft flights between 2005 and 2018	4.4	CAM-chem CMAQ WRF-Chem	3.8 5.3 5.5

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**Notes:**

<sup>1</sup>CAM-chem simulation time period is June 2015; CMAQ simulation time period is Jan-Dec 2016; WRF-Chem simulation time period is June 1-15, 2013;

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