

Author responses in blue. Planned additions to the manuscript are underlined (or shown via screen shot).

Anonymous Referee #2

Pye et al. provides a detailed review concerning aerosol and cloud acidity. This is a much needed review, as the studies concerning aerosol acidity has increased. I applaud the large author list on this work, as it is very extensive and impressive. The review provides a much needed discussion concerning various aspects of cloud and aerosol pH, including definition, measurements and calculations, observations, and comparisons with models. I foresee this paper becoming an important source, both in the field and for introducing this subject in classes. This paper should be published upon consideration of the following comments:

We thank the reviewer for the supportive comments.

1.) Similar to Reviewer #1, occult is not a term that I am familiar with. If it could be defined, or a synonym could be used, that would be appreciated.

Now defined in response to reviewer #1

2.) I appreciate Table 1, as there are numerous abbreviations throughout the manuscript. However, there appears to be some abbreviations missing, such as NVCs. Please include all abbreviations in the manuscript into Table 1, as it can be hard to find them in this large manuscript.

Appendix A has been expanded to include additional abbreviations. A second appendix (B) was added listing the models, datasets, and other related information. Items in appendix B are often abbreviations (e.g. WRF-Chem), but knowing their definition is not critical to the message of the paper. In some cases (e.g. ISORROPIA, SPECIATE) the name in B is not an abbreviation.

Updated Appendix A and new Appendix B:

Appendix A: Nomenclature

Symbol	Description
a_{H^+}	The activity of hydrogen ions in aqueous solution on a molality basis
$a_{\text{H}^+}^{(c)}$	The activity of H^+ ions on a molarity (<u>concentration</u>) basis
$a_{\text{H}^+}^{(x)}$	The activity of H^+ ions on a mole fraction basis
α_{HSO_4}	Fraction of HSO_4^- dissociated into H^+ and SO_4^{2-}
a_i	The activity of species i (usually molality-based for ions in aqueous solutions)
ACSM	Aerosol chemical speciation monitor
adjGR	Adjusted gas ratio (see Table 2)
ALPHA	Adapted Low-cost Passive High-Absorption
ALWC	Aerosol liquid water content (mass per volume of air)
AMS	Aerosol mass spectrometer
c^\ominus	The standard state (unit) molarity
c_{H^+}	The molarity or “molar concentration” of hydrogen ions in an aqueous solution (also written using square brackets as $[\text{H}^+]$)
CCN	Cloud condensation nuclei
CTM	Chemical transport model
DELTA	Denuder for Long Term Atmospheric sampling
DON	Degree of neutralization (see Table 2)
DSN	Degree of sulfate neutralization (see Table 2)
$f_{\text{H}^+}^*$	The (rational) activity coefficient based on the mole fraction concentration scale
$F_{n,i}$	Fraction of species i in the particle vs particle + gas phase
FR	Flex Ratio, identifies the NH_3 emissions level at which the nitrate concentration switches from NH_3 -insensitive (or negative sensitivity) to positive NH_3 sensitivity
$f_{\text{H}^+}^*$	The (rational) activity coefficient based on the mole fraction concentration scale
GR	Gas ratio (see Table 2)
H_{air}^+	Concentration of aerosol H^+ per volume of air (e.g., moles per m^{-3} of air)
$\text{H}_{\text{air,cb}}^+$	H_{air}^+ determined from charge balance (see Table 2)

<u>HO_x</u>	<u>Hydrogen oxides (OH + HO₂)</u>
<u>K_a</u>	<u>Acid dissociation constant for $HX \rightleftharpoons H^+ + X^-$</u>
<u>K_a</u>	<u>Acid association constant for $H^+ + X^- \rightleftharpoons HX$ or $H^+ + X^- \rightleftharpoons XH^+$</u>
<u>K_H</u>	<u>Dimensionless Henry's law constant</u>
K _w	Activity-based equilibrium constant for the dissociation of water into H ⁺ and OH ⁻ (see Bandura and Lvov (2005) for tabulation of values)
LLPS	Liquid–liquid phase separation
KMT	Kinetic mass transfer
LLPS	Liquid–liquid phase separation
m [⊖]	The standard state (unit) molality
m _{H⁺}	Molality of H ⁺ (mol kg ⁻¹ solvent)
M _w	Molar mass of water: 0.018015 kg mol ⁻¹
n _i	Number (e.g., moles) of species <i>i</i>
NEI	National Emission Inventory (for the United States)
NO _x	Nitrogen oxides (NO + NO₂)
NVC	Non-volatile cations
PAHs	polycyclic aromatic hydrocarbons
PFASs	polyfluoroalkyl substances
PFSAs	Perfluoroalkyl sulfonic acids perfluoroalkane sulfonic acid
PFCAs	Perfluoroalkyl carboxylic acids
pH	Hydrogen ion potential with activity coefficient and concentration expressed on a molality concentration scale (see Table 1)
<u>pH_c</u>	pH on a concentration (molarity) basis
<u>pH_x</u>	pH on a mole fraction basis
<u>pH_t</u>	“Total” pH based on the molality of sulfate and bisulfate ions (see Table 1)
<u>pH_f</u>	“Free ion” approximation of pH obtained when the activity coefficient of H ⁺ is unity (see Table 1)
pH _± (H, X)	Approximation of pH using the mean molal ion activity coefficient of an H ⁺ and anion X pair (see Table 1)
pK _{wi}	-log ₁₀ of (K_wK_i) (see Bandura and Lvov (2005) for tabulation of values)

<u>PM</u>	<u>Particulate matter, synonymous with aerosol</u>
<u>PM₁</u>	<u>Particulate mass with an equivalent diameter below 1 μm</u>
<u>PM_{2.5}</u>	<u>Particulate mass with an aerodynamic equivalent diameter below 2.5 μm</u>
<u>PSC</u>	<u>Pitzer-Simonson-Clegg (model)</u>
<u>R</u>	<u>Universal gas constant</u>
<u>r²</u>	<u>Coefficient of determination</u>
<u>RH</u>	<u>Relative humidity</u>
<u>RRF</u>	<u>Relative response factor, relative change in concentration due to relative change in emission</u>
<u>SO_x</u>	<u>Sulfur oxides (usually SO₂ + TSO₄)</u>
<u>T</u>	<u>Temperature</u>
<u>TMI</u>	<u>Transition Metal Ions</u>
<u>TCl</u>	<u>Total chloride (sum of gas-phase hydrochloric acid and aerosol chloride)</u>
<u>TNO₃</u>	<u>Total nitrate (sum of gas-phase nitric acid and particulate nitrate)</u>
<u>TNH₄</u>	<u>Total ammonia (sum of gas-phase ammonia and particulate ammonium)</u>
<u>TSO₄</u>	<u>Total particulate sulfate (sum of sulfate and bisulfate)</u>
<u>VOCs</u>	<u>Volatile organic compounds</u>
<u>WSOC</u>	<u>Water-soluble organic compounds</u>
<u>x_{H⁺}</u>	<u>The mole fraction of H⁺ in the solution</u>
<u>X_T</u>	<u>Molal sulfate ratio indicating sulfate- rich vs poor domain (Eq. 9)</u>
<u>Z</u>	<u>Charge balance on total gas and particle phases used to estimate initial amount of H⁺ (Eq. 19)</u>
<u>ZSR</u>	<u>Zdanovskij-Stokes-Robinson (method for calculation of aerosol water)</u>
<u>γ_{H⁺}</u>	<u>The molal activity coefficient of species <i>i</i></u>
<u>γ_{H⁺}^(c)</u>	<u>The molarity-based activity coefficient of H⁺</u>
<u>γ_{±,HX}[□]</u>	<u>The meanSingle ion activity coefficient (for monovalent acid HX)</u>
<u>ρ₀</u>	<u>The density of the reference solvent (water)</u>

<u>Appendix B: Models, data, and related methods discussed in the text</u>	
<u>Type</u>	<u>Examples</u>
<u>Activity coefficient models</u>	<u>AIOMFAC, UNIFAC</u>
<u>Gas-particle thermodynamic models</u>	<u>AIOMFAC-GLE, ADDEM, E-AIM, EQSAM, EQUISOLV II, GFEMN, ISORROPIA II, MOSAIC, SCAPE, UHAERO</u>
<u>3-D models</u>	<u>CAM-Chem, CESM, CMAQ, GEOS-Chem, GISS, PM-CAMx, TM4-ECPL, WRF-Chem</u>
<u>Supporting algorithms/chemistry/databases</u>	<u>ASTEM, CAM6, HETV, MAM, MESA, MOZART, MTEM, SPECIATE</u>
<u>Observational data sets (networks, satellites, field campaigns)</u>	<u>ACTRIS, AIRS, AMoN, CalNex, CASTNET, CrIS, CSN, EMEP/EBAS, IASI, IMPROVE, MAN, NAMN, SEARCH, SEAC4RS, SOAS, WINTER</u>

3.) I think a Table that summarizes Section 2.6 would improve the quality of the paper. This will be one of the many reasons people will want to read and cite the paper, concerning a discussion and comparison of the thermodynamic models in order to calculate pH. Including a table that summarizes how each model calculates pH, the pros and cons of each model, any assumptions, and etc., would help the readers remember that discussion better.

We added a table summarizing the four thermodynamic models used most extensively in the paper. One sentence was added to the first paragraph of section 2.6:

[Advantages and disadvantages of four common thermodynamic models are summarized in Table 2.](#)

The following new table was added:

Table 2: Common box models used to calculate acidity.

Model	Input	Acidity Output	Advantages	Disadvantages
E-AIM	Gas + particle or equilibrium particle composition (H^+ , NH_4^+ , Na^+ , SO_4^{2-} , HSO_4^- , NO_3^- , Cl^- , Br^- , organic acids, amines) in moles in overall electroneutral conditions (see Eq. 19 for Z); RH, T.	pH at equilibrium	pH via recommended Eq. 1 Considered the most accurate inorganic thermodynamic model Some ionizing organic species (e.g. organic acids, amines) considered	Computationally intensive T and RH restricted for some compositions to preserve accuracy
AIOMFAC-GLE	Gas + particle or equilibrium particle composition (H^+ , Li^+ , Na^+ , K^+ , NH_4^+ , Mg^{2+} , Ca^{2+} , Cl^- , Br^- , NO_3^- , HSO_4^- , SO_4^{2-} , organic species and/or organic functional groups) in $mol\ m^{-3}$ air for electroneutral conditions; RH, T	pH at equilibrium	pH via recommended Eq. 1 Accounts for organic--inorganic interactions and liquid-liquid equilibrium in consistent framework Code publicly distributed through repository	Limited support for solid-liquid equilibria of diverse inorganic salts (presently) Optimized for temperatures near 298 K, with limited accuracy for much colder atmospheric temperatures
MOSAIC	Distinct gas and particle composition (H^+ , NH_4^+ , Na^+ , Ca^{2+} , SO_4^{2-} , HSO_4^- , $CH_3SO_3^-$, NO_3^- , Cl^- , and CO_3^{2-}) in $mol\ m^{-3}$ air; RH, and T. Automatic adjustments applied to non-electroneutral input particle-phase composition.	pH_F by default (pH_{\pm} with modification) for each particle size bin (or mode) at each time step while dynamically solving gas-particle mass transfer	Provides size-resolved pH_F and pH_{\pm} to account for compositional heterogeneity across particles of different sizes and origins Does not require equilibrium assumption	Gas-particle and solid-liquid equilibrium constants depend on temperature, but activity coefficients are limited to 298.15 K.
ISORROPIA II	Gas + particle or particle composition (TSO_4 , TCl , TNO_3 , TNH_4 , Na , K , Ca , Mg) in $mol\ m^{-3}$ or $\mu g\ m^{-3}$ air; RH, T. Automatic adjustments applied to non-electroneutral input particle-phase composition.	pH_F by default (pH_{\pm} with modification) at equilibrium	Computationally efficient Code has widespread public distribution and incorporation in CTMs	Approximations employed (e.g. some activity coefficients treated as 1, minor species do not perturb equilibrium) Segmented solution approach leads to discontinuous solution surface

4.) Throughout the sections, it is apparent that different people wrote them with different styles coming through in each section. For example, some sections briefly state future research while other sections devote a subsection about future research (and some sections do not have any discussion about questions/future work). Starting with at least Section 3, if not some aspects of Section 2 (i.e., Measuring pH), it would be beneficial for the authors to include a description of what they consider future questions/work to be.

In some cases, concepts span multiple sections. For example, proxies were introduced in section 4, but the box model intercomparison in section 6 had summarizing messages. We have reorganized the paper to improve the flow/connection throughout. The new order will be:

- i. Introduction
- ii. Definition
- iii. Proxies (former 4)
- iv. Box model comparison & w/proxies (former 6)
- v. Role of processes: Aqueous Chemistry (former 3)
- vi. Role of size, composition, mass transfer (former 7)
- vii. Observations (former 5)
- viii. CTM results
- ix. Conclusions

This moves the box model intercomparison (*iv*) immediately after definitions of pH and proxies. The box model intercomparison contains a recap of concepts introduced in the preceding two sections (in Section 4.3: Recommendations on the calculation of pH by approximation and proxy) and thus serves as a synthesis of sections *ii-iv*.

New section *v* (old section 3) covers the role of aqueous chemistry. Since the section is already short, summary in nature, and the subject of a companion paper, an additional summary paragraph was not added.

For new Section *vi* (old 7) on size, composition, mass transfer, the last two subsections end with specific conclusions regarding the roles of size, composition, mass transfer, and organics/LLPS. No further conclusions/future directions were added.

Section *vii* (old 5/Observations) is divided into section 7.1 (aerosols) and 7.2 (clouds). Section 7.2 already had a section on the need for future monitoring of cloud pH. Section 7.2.4 was promoted up to 7.3 and expanded to include aerosol pH:

7.3 Need for future monitoring of cloud and aerosol pH

Although cloud and fog sampling is generally more challenging than aerosol collection, pH measurement of the collected cloud/fog water is simpler due to its much larger volume and much lower ionic strength. As a result, Over the past several decades, fogs and clouds have been sampled and their pH determined in areas around the globe with more temporal and spatial coverage than for aerosol pH. Depending on inputs of key acids and bases, cloud/fog pH has been observed to range from below 2 to greater than 7-, slightly higher, but similar to fine aerosol pH that ranges from below 0 to near 7. Programs designed to target reductions in acid rain have had

direct impacts on cloud and fog pH-, but aerosol pH has been much more constant than cloud pH in the southeastern US and southeastern Canada over time. Analysis of cloud pH observations over the past 25 – 30 years reveals that cloud/fog acidity in many regions has decreased as anthropogenic emissions of the important acid precursors, SO₂ and NO_x, have decreased. A continued rise in cloud/fog pH is likely in many regions with planned, future decreases in NO_x and SO₂ emissions and stable or increasing NH₃ emissions. Future changes in emissions could eventually be significant enough to lead to fine aerosol pH changes as well. Increases in cloud pH are expected to enhance the solubility of gas phase organic acids, potentially shortening their atmospheric lifetimes- while increases in aerosol pH could lead to more nitrate aerosol formation and allow previously unfavorable kinetic reactions to occur.

As emissions evolve with time, continued characterization of cloud and particle pH is needed to understand how anthropogenic activities affect condensed-phase acidity and downstream endpoints in the earth system. Much remains to be learned about factors controlling cloud/fog pH in the atmosphere and the influence of this acidity on aqueous phase chemistry, including the aqueous phase uptake and oxidation of soluble gases to form secondary inorganic or organic aerosol. More detailed measurements of organic acids and bases, and their influence on cloud pH, will be increasingly important as sulfate and nitrate concentrations decline. Likewise, there is a need for more systematic monitoring of cloud and fog composition in key environments, as opposed to the more ad hoc past sampling approaches driven primarily by the objectives of process-based research. Because fogs and clouds are good integrators of atmospheric acids and bases in both the gas and particle phases, they may offer a convenient and practical basis for ongoing monitoring of atmospheric acidity. Future monitoring strategies should consider long-term monitoring at surface sites as well as periodic measurements of cloud, particle, and gas-phase composition from aircraft in order to enhance our understanding of acidity at higher elevations in the troposphere. Future measurements should also better document heterogeneity of acidity across individual drops within a cloud or fog or aerosol population, for example ~~looking at~~ determining the size-dependence of ~~drop~~ pH. Aerosol pH estimates will likely continue to be primarily based on thermodynamic models in the near future and thus require simultaneous particle- and gas-phase measurements (specifically of ammonia) to improve the spatial and temporal scales over which fine particle pH is currently characterized.

Section *viii* (CTM predictions) already contains a summary/future directions section.

Multiple minor changes were made throughout the manuscript for the new section order (will be provided in the future tracked changes document).

5.) For Section 3, it would be good to include a couple of items in discussion, including: (a) How many of the reactions have been conducted for dilute, laboratory conditions, therefore, for aerosol, where the ion activity is higher and water is lower, there is large uncertainty in how the reactions may occur. (b) How there is debate occurring the field about various reactions (e.g., production of sulfate in aerosol in China) and the questions/future studies needed to move this questions forward. (c) N₂O₅ chemistry appears to be missing in your discussion throughout, including in Section 5.

The current paper is already quite long (~140 pages). The topic of kinetic drivers of pH and how pH affects kinetics is a large topic and warrants a separate companion paper. Former section 3 (now 5) was meant to highlight some examples of the kinetics-pH interplay, but not intended to be comprehensive. We will defer the bulk of these reviewer suggestions to the companion paper. However, we added mention of N_2O_5 in the introduction along with two references:

The concentration of fine particulate matter ($PM_{2.5}$) is directly modulated by pH through its effects on gas-particle partitioning, and pH-dependent condensed-phase reactions, and other particle processes influenced by pH. For example, N_2O_5 heterogeneous hydrolysis significantly affects tropospheric chemistry (Dentener and Crutzen, 1993) and depends strongly on particle composition (Chang et al., 2011), including formation of organic coatings due to liquid-liquid phase separation influenced by acidity (see Section 6.3 for a discussion of phase separation in the context of acidity).

Chang, W. L., Bhave, P.V., Brown, S.S., Riemer, N., Stutz, J., and Dabdub, D.: Heterogeneous atmospheric chemistry, ambient measurements, and model calculations of N_2O_5 : A review, *Aerosol Sci. Technol.*, 45, 6665-6695, <https://doi.org/10.1080/02786826.2010.551672>, 2011.

Dentener, F. J., and Crutzen, P. J.: Reaction of N_2O_5 on tropospheric aerosols: Impact on the global distributions of NO_x , O_3 , and OH, *J. Geophys. Res.*, 98(D4), 7149–7163, <https://doi.org/10.1029/92JD02979>, 1993.

6.) For Section 8, a table that summarizes the CTMs with the thermodynamic models they use and the species they use to calculate pH would help with the discussion.

We added a new table, Table 7, that summarizes the CTM calculations of pH. We included models used in this work only (rather than characterizing all CTMs in literature). A sentence was added to the first paragraph of Section 8:

Table 7 summarizes the species considered in the calculation of pH for each model displayed in this work.

The following new table was added:

Table 7: Species and methods used to calculate acidity in CTMs. Bulk cloudwater pH is calculated assuming electroneutrality, generally using model-specific algorithms. Dissolved gases in cloudwater are determined using Henry's law coefficients. Configurations are specific to this work.

Model	Aerosol size information	Species/sources considered in aerosol pH calculation	Fine aerosol pH calculation method	Species/sources considered in cloud pH calculation
CMAQ v5.3	Fine aerosol: explicit Aitken and accumulation modes. Coarse mode acidity not explicitly calculated but included in determination of dynamic mass transfer and composition.	TSO ₄ , TCl, TNO ₃ , TNH ₄ , Na, K, Ca, Mg from sea salt, dust, wildland fires, and anthropogenic activities.	ISORROPIA II pH _F for inorganic-only composition of combined fine modes. Condensed water associated with organic species is also predicted (not considered in fine aerosol pH _F in this work).	Aqueous species: H ⁺ , OH ⁻ , HSO ₃ ⁻ , SO ₃ ²⁻ , HSO ₄ ⁻ , SO ₄ ²⁻ , HCO ₃ ⁻ , CO ₃ ²⁻ , HCO ₂ ⁻ , NH ₄ ⁺ , NO ₃ ⁻ , Cl ⁻ , Ca ²⁺ , Na ⁺ , K ⁺ , Mg ²⁺ Dissolved gases: SO ₂ , CO ₂ , NH ₃ , HCl, HNO ₃ , HCOOH, H ₂ SO ₄ (as sulfate), N ₂ O ₅ (as 2×HNO ₃)
GEOS-Chem v12.0.0	Bulk fine aerosol. Coarse mode acidity not explicitly calculated but included in determination of dynamic mass transfer and composition.	TSO ₄ , HCl, TNO ₃ , TNH ₄ , and fine mode Ca, Mg, Na, Cl from anthropogenic, sea salt, and dust sources (dust contributions not considered in default GEOS-Chem predictions but Ca and Mg from dust considered in this work).	ISORROPIA II pH _F .	Aqueous species: SO ₄ ²⁻ , NO ₃ ⁻ , NH ₄ ⁺ Dissolved gases: CO ₂ , SO ₂ , NH ₃ , HNO ₃
TM4-ECPL	Fine (externally mixed dust) and coarse (internally mixed dust) aerosol.	SO ₄ ²⁻ , NH ₃ , NH ₄ ⁺ , HNO ₃ and NO ₃ ⁻ ; sea salt and dust assumed to be externally mixed with fine mode sulfate and not considered in the fine acidity calculation.	ISORROPIA II pH _F for inorganic-only composition of fine and coarse modes (each in equilibrium with gas). Condensed water associated with organic species is also predicted (not considered in fine aerosol pH _F in this work).	Aqueous species: SO ₄ ²⁻ , CH ₃ O ₃ S ⁻ , NO ₃ ⁻ , NH ₄ ⁺ , Na ⁺ , Ca ²⁺ , K ⁺ , Cl ⁻ , Mg ²⁺ Dissolved gases: SO ₂ , CO ₂ , HNO ₃ , NH ₃ , oxalic acid
WRF-Chem	Four aerosol size bins (0.039–0.156, 0.156–0.625, 0.625–2.5, 2.5–10 μm in diameter) treated dynamically.	sulfate, HNO ₃ /NO ₃ ⁻ , NH ₃ /NH ₄ ⁺ , CH ₃ O ₃ S ⁻ , Cl ⁻ , CO ₃ ²⁻ , Na, Ca; HCl not considered with MOZART chemistry (no displacement of Cl ⁻ from sea salt aerosols allowed).	MOSAIC size-resolved pH _F .	Aqueous species: OH ⁻ , HCO ₃ ⁻ , CO ₃ ²⁻ , CO ₃ ⁻ , HSO ₃ ⁻ , SO ₃ ²⁻ , HSO ₄ ⁻ , SO ₄ ²⁻ , SO ₄ ⁻ , SO ₅ ⁻ , HSO ₅ ⁻ , HOCH ₂ SO ₃ ⁻ , ⁻ OCH ₂ SO ₃ ⁻ , NO ₂ ⁻ , NO ₃ ⁻ , HO ₂ ⁻ , O ₂ ⁻ , HCOO ⁻ , Cl ⁻ , Cl ₂ ⁻ , ClOH ⁻ , NH ₄ ⁺ , Fe ³⁺ , Mn ²⁺ Dissolved gases: SO ₂ , CO ₂ , HNO ₃ , NH ₃ , HO ₂ , HCOOH, H ₂ O ₂
CAM-Chem	Four log-normal modes.	Inorganic aerosol composition considered: SO ₄ ²⁻ , NH ₄ ⁺ , soil dust, sea salt.	Not considered in this work.	Aqueous species: OH ⁻ , HCO ₃ ⁻ , NO ₃ ⁻ , HSO ₃ ⁻ , SO ₃ ²⁻ , SO ₄ ²⁻ , NH ₄ ⁺ Dissolved gases: SO ₂ , H ₂ SO ₄ , HNO ₃ , CO ₂ , NH ₃

7.) For Section 9, I appreciate that it summarizes the very large review. However, at this point, I really think a description of remaining questions, studies, observations and future outlook is necessary so that we, as a community, know what should be done to move forward.

The synthesized messages in section 9 were developed at a workshop involving the coauthors of this study. Coauthors submitted their thoughts on major messages ahead of discussion then messages were discussed, refined, and agreed upon by the group. As a result, we think the most important major messages (which include both summary information and future directions) have been captured. The individual sections contain additional information. Some future directions (e.g. understanding/improving bisulfate dissociation predictions to improve consistency among box models) were raised and discussed by the group but considered too detailed for the main messages.

We think that section 9 already provides guidance to the community. The following guidance is contained in section 9:

- We recommend researchers use specific nomenclature to document and communicate what metric of acidity they report
- pH is the ideal indicator of acidity and researchers should aim to report that value or an approximation (pH_{\pm} is best approximation)
- The role of kinetic-pH interactions is likely underappreciated and should be further examined to understand where H^+ is chemically generated (this is discussed in the companion paper)
- Experimental determination of aerosol pH is a current knowledge gap
- Heterogeneity of pH across the aerosol/cloud droplet population and within a given particle needs investigation (most methods/models use bulk techniques)
- Ammonia measurements are needed to facilitate determination of aerosol pH
- Ambient characterization of pH is spatially and temporally incomplete (more observations or observationally constrained estimates are needed-Section 7 highlights current areas without measurements and where trends are available).
- pH should be considered in the context of CTM evaluation and endpoints of interest.
- Considerable model diversity in predicted pH exists and Section 8 points to mixing state assumptions and composition as reasons and locations where model diversity is high and could be measured