Supplemental information for

Roles of pH, ionic strength and sulfate in the aqueous nitrate-mediated photooxidation of green leaf volatiles

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S1. Measurement of p-hydroxybenzoic acid

p-hydroxybenzoic acid, which is formed from the reaction of ·OH with BA (k_{BA+OH} = 5.9×10^9 M^{-1} s^{-1} (Herrmann et al., 2010)) at a yield of 0.17 (Anastasio and Mcgregor, 2001), was measured in separate experiments using an ultra-high performance liquid chromatography system (1290 system, Agilent) coupled to a high-resolution quadrupole-time-of-flight mass spectrometer (X500R QTOF MS/MS, Sciex) (UPLC-MS) equipped with an electrospray ionization (ESI) source that was operated in negative mode. A reverse phase Kinetex (Phenomenex) Polar C18 column (2.6 μm, 150 × 2.1 mm) equipped with a Polar C18 guard column was used for UPLC-MS analysis. The temperatures for the column oven and the UPLC autosampler were set to 25 ℃. A gradient elution program was used. For the mobile phase, eluent A was 10 mM ammonia acetate in Milli-Q water buffered with 0.03% acetic acid, and eluent B was pure methanol. A gradient elution program was used, and it was delivered at a flow rate of 0.3 mL min^{-1}. The following mobile phase gradient was used for the detection of BA and its product PHBA: 0 to 3 min 1% B, 3 to 5 min linear rise to 80% B and hold to 6 min, 6 to 6.5 min linear drop to 1% B and then hold to 10 min for equilibrium. The sample injection volume was set to 10 μL. The following tandem MS conditions were used: -4500 V ESI ion spray voltage, 80 V declustering potential, -20 V collision energy, 50 psi ion source gas, 25 psi curtain gas, and 450 ℃ source temperature. Solid phase extraction (SPE) was performed to desalt the samples using SPE cartridges (Oasis MAX, 60 mg, 3 cc, 60 μm, Waters). First, the sorbent was conditioned and equilibrated using 3 mL of methanol (LC-MS grade) followed by 3 mL of Milli-Q water. Next, the cartridge was loaded with 3 mL of 1× diluted sample solution and then purged with 6 mL of Milli-Q water. A vacuum pump was used to dry out the sorbent before elution using 3 mL of 2% formic acid in methanol. All the desalted samples were filtered using 0.2 μm nylon syringe filters to remove any particulates prior to UPLC-MS analysis.

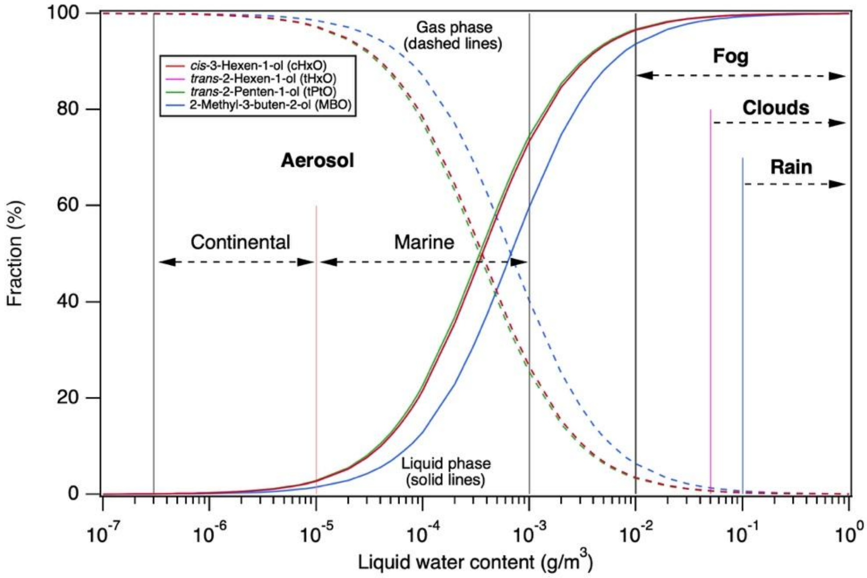

Figure S1. Calculated partitioning of the GLVs between the gas and aqueous phases as a function of liquid water content. The Henry's law solubility coefficients used for the calculation of cHxO, tHxO, tPtO, and MBO were 113 M atm^{-1}, 94 M atm^{-1},120 M atm^{-1}, and 61 M atm^{-1}, respectively (Sarang et al., 2021; Sander, 2023).

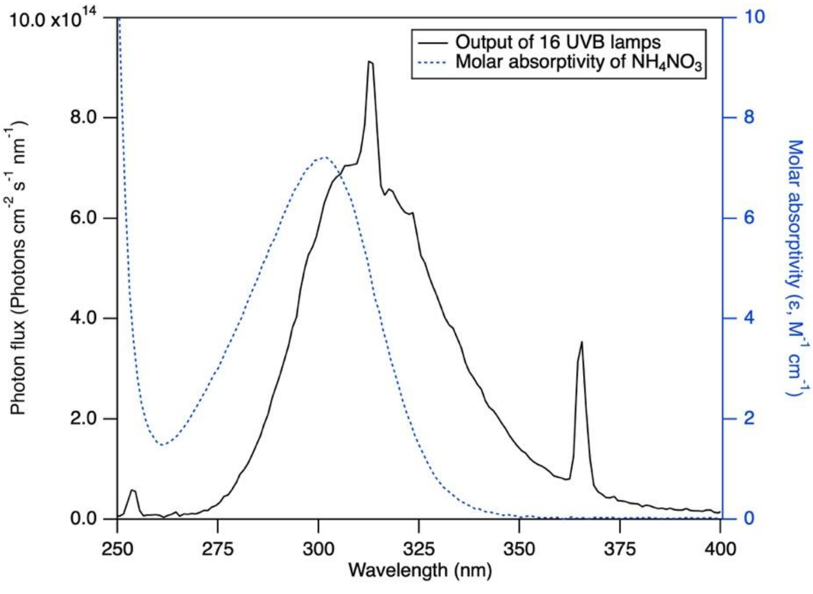

Figure S2. The photon flux of inside the photoreactor and the molar absorptivity (ε) of NH_4NO_3.

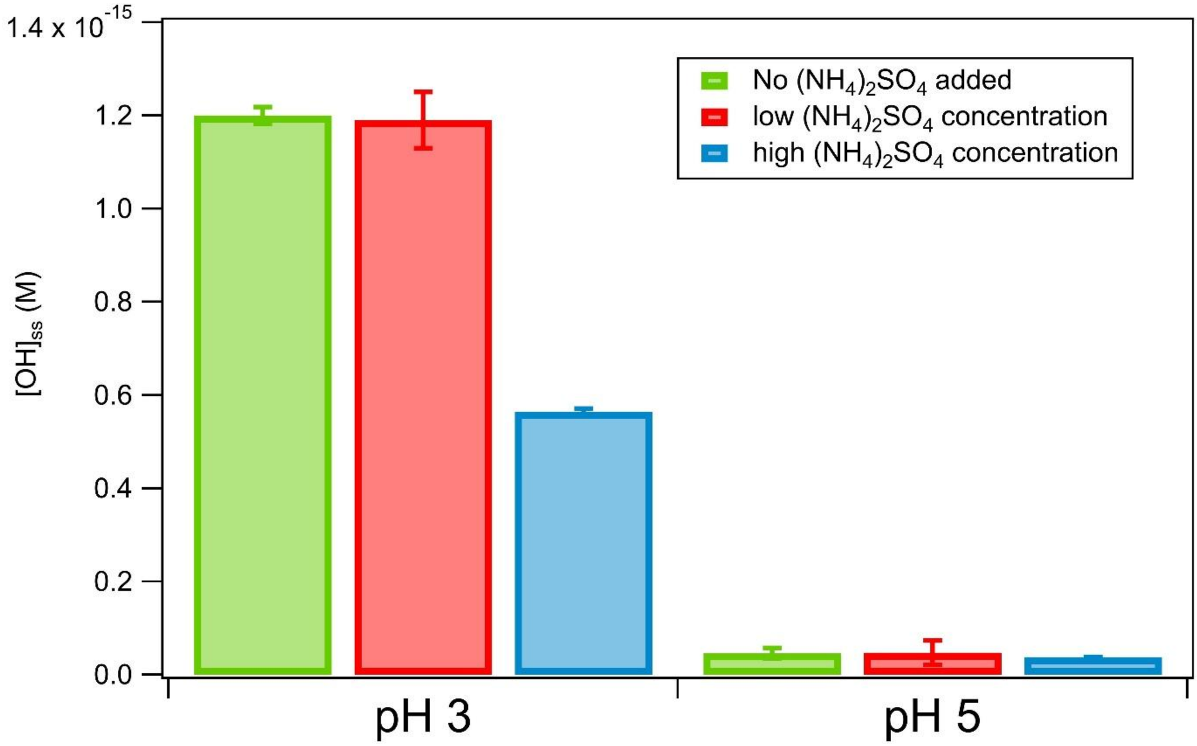

Figure S3. Estimated $[\cdot OH]_{ss}$ in nitrate-mediated photooxidation experiments under dilute cloud/fog-like conditions. These values were obtained from a separate set of experiments (i.e., GLVs were not present in the solutions) using benzoic acid (10 µM) as the ·OH probe compound and measuring the formation of p-hydroxybenzoic acid from the reaction of ·OH with BA (Lyu et al., 2023; Yang et al., 2021; Yang et al., 2023). The error bars represent one standard deviation originating from multiple experiments and measurements. For the low $(NH_4)_2SO_4$ concentration conditions (red bars), 0.135 mM and 0.583 mM of $(NH_4)_2SO_4$ was added into the solutions for pH 3 and 5, respectively (Table 1). For the high $(NH_4)_2SO_4$ concentration conditions (blue bars), 6.135 mM and 6.580 mM of $(NH_4)_2SO_4$ was added into the solutions for pH 3 and 5, respectively (Table 1).

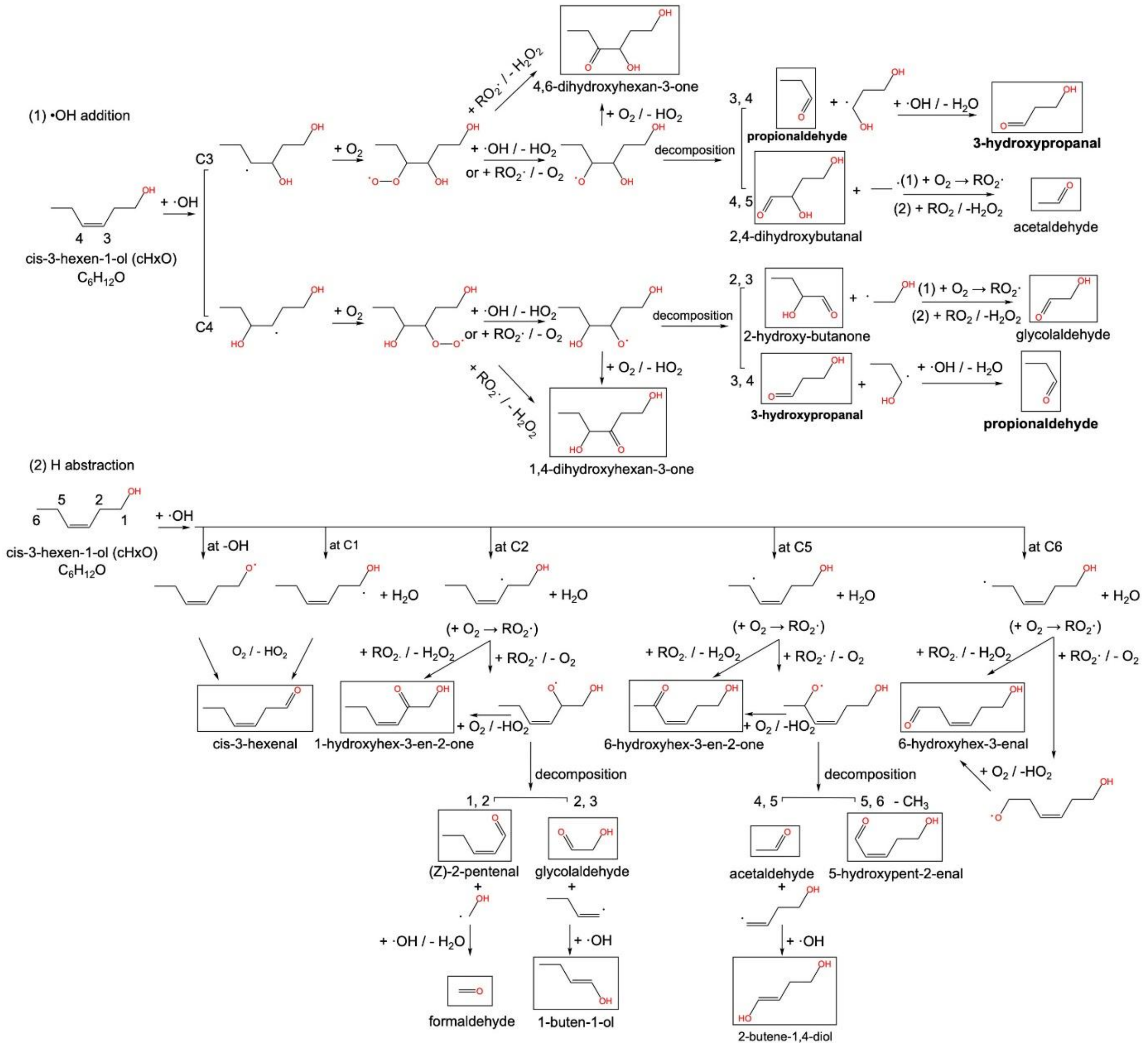

Figure S4. The proposed reaction mechanism and first-generation products via (1) ·OH addition, and (2) H abstraction for the oxidation cHxO by ·OH in the aqueous phase based on the existing literature (Reisen et al., 2003; Sarang et al., 2023). Similar reaction mechanisms are expected for the ·OH oxidation of tHxO and tPtO given their similar molecular structures. The expected products are shown in boxes, while expected key products are highlighted in boldface. Note that ·OH can also react with organic compounds through electron transfer pathways, which are not included here due to their expected minor roles in oxidation with GLVs. Bimolecular combination reaction pathways involving $RO_2\cdot$ and RO· (e.g., $RO_2\cdot$ + $RO_2\cdot$) that lead to oligomer formation are also not known here.

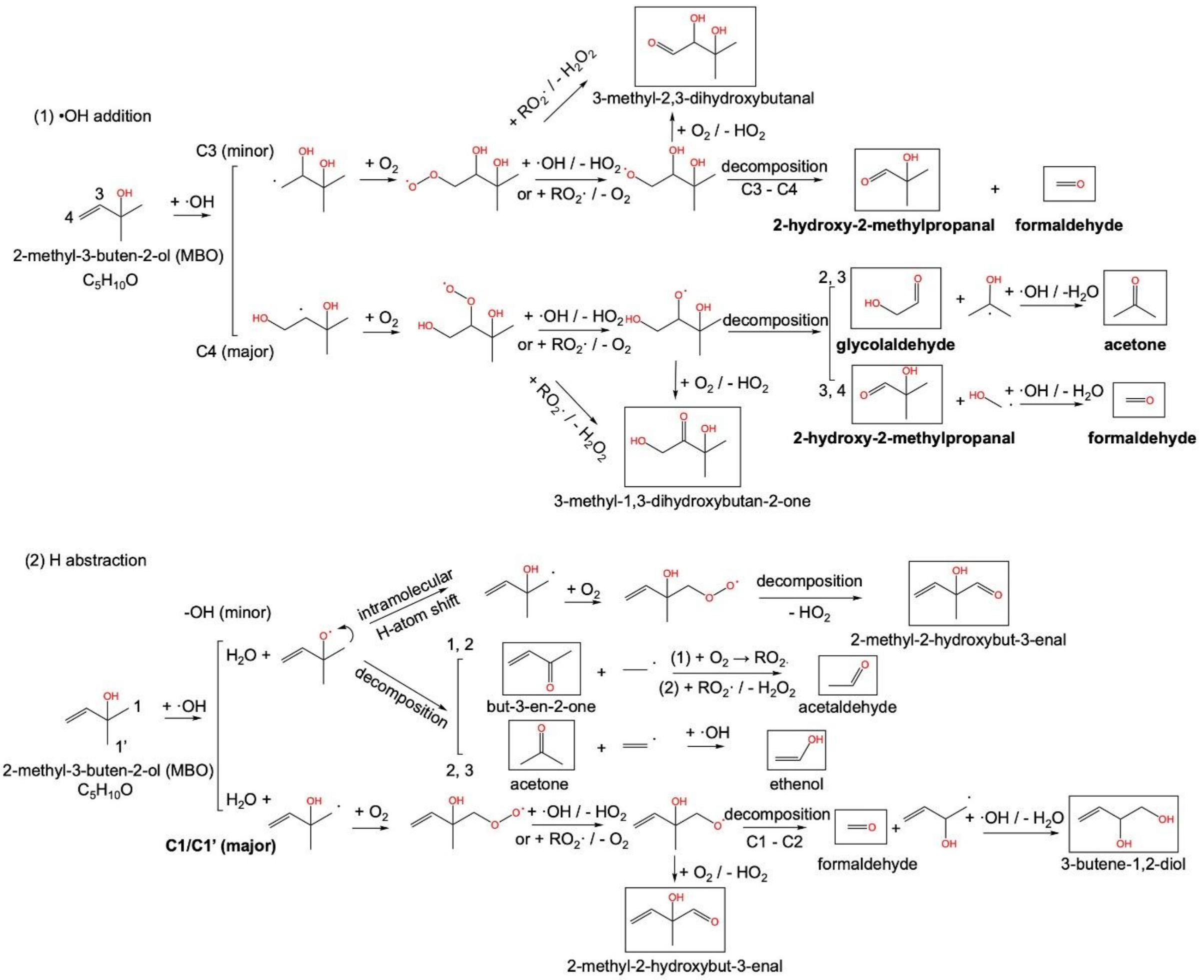

Figure S5. The proposed reaction mechanism and first generation products via (1) ·OH addition and (2) H abstraction for the oxidation MBO by ·OH in the aqueous phase based on the existing literature (Atkinson and Arey, 2003; Carrasco et al., 2007; Chan et al., 2009; Reisen et al., 2003; Sarang et al., 2023). The expected products are shown in boxes, while expected key products are highlighted in boldface. Note that ·OH can also react with MBO through electron transfer pathways, which are not included here due to their expected minor roles in oxidation with GLVs. Bimolecular combination reaction pathways involving RO_2· and RO· (e.g., RO_2· + RO_2·) that lead to oligomer formation are also not known here.

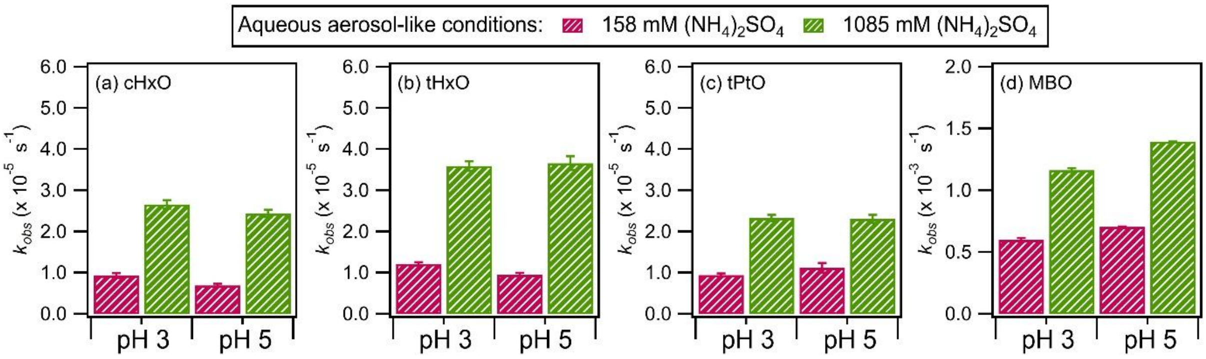

Figure S6. The k_{obs} values for the decays of the four GLVs upon irradiation when only sulfate (in the form of $(NH_4)_2SO_4$ and (for pH 3) H_2SO_4) was present in the solutions. The error bars represent one standard deviation originating from triplicate experiments and measurements.

Table S1. List of reactions pathways initiated by the aqueous photolysis of nitrate compiled from the literature (Gligorovski et al., 2015; Herrmann, 2007; Mack and Bolton, 1999; Marussi and Vione, 2021; Scharko et al., 2014).

No.	Reactions	Quantum yield (Φ)/ Acid dissociation constant (pK_a)
1	$NO_3^- + h\nu \rightarrow [NO_2\bullet + O\bullet^-]_{cage}$	$\Phi = 0.01$
2	$[NO_2\bullet + O\bullet^-]_{cage} \rightarrow NO_2\bullet + O\bullet^-$	—
3	$O\bullet^- + H_2O \rightleftharpoons \bullet OH + OH^-$	$pK_a(\bullet OH) = 11.9$
4	$[NO_2\bullet + O\bullet^-]_{cage} \rightarrow OONO^-$	—
5	$OONO^- + H^+ \rightleftharpoons HOONO$	$pK_a = 7$
6	$HOONO \rightarrow \bullet OH + NO_2\bullet$	—
7	$2NO_2\bullet \rightleftharpoons N_2O_4$	—
8	$N_2O_4 + H_2O \rightarrow HNO_2 + NO_3^- + H^+$	—
9	$HNO_2 \rightleftharpoons H^+ + NO_2^-$	$pK_a = \sim 3.3$
10	$NO_2^- + h\nu \rightarrow NO\bullet + O\bullet^-$	$\Phi = 0.025–0.065$
11	$NO_2^- + h\nu \rightarrow NO_2\bullet + e^-$	$\Phi = \sim 0.001$
12	$NO_2^- + \bullet OH \rightarrow NO_2\bullet + OH^-$	—
13	$NO\bullet + NO_2\bullet \rightleftharpoons N_2O_3$	—
14	$N_2O_3 + H_2O \rightarrow 2\ NO_2^- + 2\ H^+$	—
15	$HNO_2 + h\nu \rightarrow NO\bullet + \bullet OH$	$\Phi = 0.35$
16	$HNO_2 + \bullet OH \rightarrow NO_2\bullet + H_2O$	—
17	$2\ HNO_2 \rightarrow NO\bullet + NO_2\bullet + H_2O$	—

Table S2. Statistical analyses (student's *t* test) for the differences in k_{obs} at different pH and ionic strengths under cloud/fog-like conditions for cHxO (Figure 2 in the main text).

cHxO	pH 3 I_{total} = 0.002 M	pH 3 I_{total} = 0.02 M	pH 5 I_{total} = 0.002 M	pH 5 I_{total} = 0.02 M
pH 3 I_{total} = 0.002 M	/	N.S.S.	$p < 0.05$	N.S.S.
pH 3 I_{total} = 0.02 M	N.S.S.	/	N.S.S.	N.S.S.
pH 5 I_{total} = 0.002 M	$p < 0.05$	N.S.S.	/	N.S.S.
pH 5 I_{total} = 0.02 M	N.S.S.	N.S.S.	N.S.S.	/

Note: If the *p* value is smaller than 0.05, this indicates that the difference between the two variables in the student's *t* test is statistically significant. Conversely, if the *p* value is larger than 0.05, this indicates that the difference is not statically significant (N.S.S.).

Table S3. Statistical analyses (student's t test) for the differences in k_{obs} at different pH and ionic strengths under cloud/fog-like conditions for tHxO (Figure 2 in the main text).

tHxO	pH 3 I_{total} = 0.002 M	pH 3 I_{total} = 0.02 M	pH 5 I_{total} = 0.002 M	pH 5 I_{total} = 0.02 M
pH 3 I_{total} = 0.002 M	/	N.S.S.	$p < 0.05$	$p < 0.05$
pH 3 I_{total} = 0.02 M	N.S.S.	/	$p < 0.05$	$p < 0.05$
pH 5 I_{total} = 0.002 M	$p < 0.05$	$p < 0.05$	/	N.S.S.
pH 5 I_{total} = 0.02 M	$p < 0.05$	$p < 0.05$	N.S.S.	/

Note: If the p value is smaller than 0.05, this indicates that the difference between the two variables in the student's t test is statistically significant. Conversely, if the p value is larger than 0.05, this indicates that the difference is not statically significant (N.S.S.).

Table S4. Statistical analyses (student's t test) for the differences in k_{obs} at different pH and ionic strengths under cloud/fog-like conditions for tPtO (Figure 2 in the main text).

tPtO	pH 3 I_{total} = 0.002 M	pH 3 I_{total} = 0.02 M	pH 5 I_{total} = 0.002 M	pH 5 I_{total} = 0.02 M
pH 3 I_{total} = 0.002 M	/	N.S.S.	$p < 0.05$	$p < 0.05$
pH 3 I_{total} = 0.02 M	N.S.S.	/	$p < 0.05$	$p < 0.05$
pH 5 I_{total} = 0.002 M	$p < 0.05$	$p < 0.05$	/	N.S.S.
pH 5 I_{total} = 0.02 M	$p < 0.05$	$p < 0.05$	N.S.S.	/

Note: If the p value is smaller than 0.05, this indicates that the difference between the two variables in the student's t test is statistically significant. Conversely, if the p value is larger than 0.05, this indicates that the difference is not statically significant (N.S.S.).

Table S5. Statistical analyses (student's *t* test) for the differences in k_{obs} at different pH and ionic strengths under cloud/fog-like conditions for MBO (Figure 2 in the main text).

MBO	pH 3 I_{total} = 0.002 M	pH 3 I_{total} = 0.02 M	pH 5 I_{total} = 0.002 M	pH 5 I_{total} = 0.02 M
pH 3 I_{total} = 0.002 M	/	N.S.S.	$p < 0.05$	$p < 0.05$
pH 3 I_{total} = 0.02 M	N.S.S.	/	$p < 0.05$	$p < 0.05$
pH 5 I_{total} = 0.002 M	$p < 0.05$	$p < 0.05$	/	N.S.S.
pH 5 I_{total} = 0.02 M	$p < 0.05$	$p < 0.05$	N.S.S.	/

Note: If the *p* value is smaller than 0.05, this indicates that the difference between the two variables in the student's *t* test is statistically significant. Conversely, if the *p* value is larger than 0.05, this indicates that the difference is not statically significant (N.S.S.).

Table S6. Statistical analyses (student's *t* test) for the differences in Y_{SOA} between pH and ionic strength under cloud/fog-like conditions for cHxO (Figure 3 in the main text).

cHxO	pH 3 I_{total} = 0.002 M	pH 3 I_{total} = 0.02 M	pH 5 I_{total} = 0.002 M	pH 5 I_{total} = 0.02 M
pH 3 I_{total} = 0.002 M	/	N.S.S.	$p < 0.05$	$p < 0.05$
pH 3 I_{total} = 0.02 M	N.S.S.	/	$p < 0.05$	$p < 0.05$
pH 5 I_{total} = 0.002 M	$p < 0.05$	$p < 0.05$	/	$p < 0.05$
pH 5 I_{total} = 0.02 M	$p < 0.05$	$p < 0.05$	$p < 0.05$	/

Note: If the *p* value is smaller than 0.05, this indicates that the difference between the two variables in the student's *t* test is statistically significant. Conversely, if the *p* value is larger than 0.05, this indicates that the difference is not statically significant (N.S.S.).

Table S7. Statistical analyses (student's *t* test) for the differences in Y_{SOA} between pH and ionic strength under cloud/fog-like conditions for tHxO (Figure 3 in the main text).

tHxO	pH 3 I_{total} = 0.002 M	pH 3 I_{total} = 0.02 M	pH 5 I_{total} = 0.002 M	pH 5 I_{total} = 0.02 M
pH 3 I_{total} = 0.002 M	/	$p < 0.05$	N.S.S.	$p < 0.05$
pH 3 I_{total} = 0.02 M	$p < 0.05$	/	N.S.S.	N.S.S.
pH 5 I_{total} = 0.002 M	N.S.S.	N.S.S.	/	N.S.S.
pH 5 I_{total} = 0.02 M	$p < 0.05$	N.S.S.	N.S.S.	/

Note: If the *p* value is smaller than 0.05, this indicates that the difference between the two variables in the student's *t* test is statistically significant. Conversely, if the *p* value is larger than 0.05, this indicates that the difference is not statically significant (N.S.S.).

Table S8. Statistical analyses (student's *t* test) for the differences in Y_{SOA} between pH and ionic strength under cloud/fog-like conditions for tPtO (Figure 3 in the main text).

tPtO	pH 3 I_{total} = 0.002 M	pH 3 I_{total} = 0.02 M	pH 5 I_{total} = 0.002 M	pH 5 I_{total} = 0.02 M
pH 3 I_{total} = 0.002 M	/	N.S.S.	N.S.S.	$p < 0.05$
pH 3 I_{total} = 0.02 M	N.S.S.	/	N.S.S.	$p < 0.05$
pH 5 I_{total} = 0.002 M	N.S.S.	N.S.S.	/	$p < 0.05$
pH 5 I_{total} = 0.02 M	$p < 0.05$	$p < 0.05$	$p < 0.05$	/

Note: If the *p* value is smaller than 0.05, this indicates that the difference between the two variables in the student's *t* test is statistically significant. Conversely, if the *p* value is larger than 0.05, this indicates that the difference is not statically significant (N.S.S.).

Table S9. Statistical analyses (student's *t* test) for the differences in Y_{SOA} between pH and ionic strength under cloud/fog-like conditions for MBO (Figure 3 in the main text).

MBO	pH 3 I_{total} = 0.002 M	pH 3 I_{total} = 0.02 M	pH 5 I_{total} = 0.002 M	pH 5 I_{total} = 0.02 M
pH 3 I_{total} = 0.002 M	/	N.S.S.	N.S.S.	N.S.S.
pH 3 I_{total} = 0.02 M	N.S.S.	/	N.S.S.	N.S.S.
pH 5 I_{total} = 0.002 M	N.S.S.	N.S.S.	/	N.S.S.
pH 5 I_{total} = 0.02 M	N.S.S.	N.S.S.	N.S.S.	/

Note: If the *p* value is smaller than 0.05, this indicates that the difference between the two variables in the student's *t* test is statistically significant. Conversely, if the *p* value is larger than 0.05, this indicates that the difference is not statically significant (N.S.S.).

Table S10. Statistical analyses (student's *t* test) for the differences in k_{obs} at different pH and ionic strengths under aqueous aerosol-like conditions for cHxO (Figure 4 in the main text).

cHxO	pH 3 I_{total} = 0.5 M	pH 3 I_{total} = 3.3 M	pH 5 I_{total} = 0.5 M	pH 5 I_{total} = 3.3 M
pH 3 I_{total} = 0.5 M	/	$p < 0.05$	N.S.S.	$p < 0.05$
pH 3 I_{total} = 3.3 M	$p < 0.05$	/	$p < 0.05$	N.S.S.
pH 5 I_{total} = 0.5 M	N.S.S.	$P< 0.05$	/	$p < 0.05$
pH 5 I_{total} = 3.3 M	$p < 0.05$	N.S.S.	$p < 0.05$	/

Note: If the *p* value is smaller than 0.05, this indicates that the difference between the two variables in the student's *t* test is statistically significant. Conversely, if the *p* value is larger than 0.05, this indicates that the difference is not statically significant (N.S.S.).

Table S11. Statistical analyses (student's t test) for the differences in k_{obs} at different pH and ionic strengths under aqueous aerosol-like conditions for tHxO (Figure 4 in the main text).

tHxO	pH 3 I_{total} = 0.5 M	pH 3 I_{total} = 3.3 M	pH 5 I_{total} = 0.5 M	pH 5 I_{total} = 3.3 M
pH 3 I_{total} = 0.5 M	/	$p < 0.05$	N.S.S.	$p < 0.05$
pH 3 I_{total} = 3.3 M	$p < 0.05$	/	$p < 0.05$	$p < 0.05$
pH 5 I_{total} = 0.5 M	N.S.S.	$p < 0.05$	/	$p < 0.05$
pH 5 I_{total} = 3.3 M	$p < 0.05$	$p < 0.05$	$p < 0.05$	/

Note: If the p value is smaller than 0.05, this indicates that the difference between the two variables in the student's t test is statistically significant. Conversely, if the p value is larger than 0.05, this indicates that the difference is not statically significant (N.S.S.).

Table S12. Statistical analyses (student's t test) for the differences in k_{obs} at different pH and ionic strengths under aqueous aerosol-like conditions for tPtO (Figure 4 in the main text).

tPtO	pH 3 I_{total} = 0.5 M	pH 3 I_{total} = 3.3 M	pH 5 I_{total} = 0.5 M	pH 5 I_{total} = 3.3 M
pH 3 I_{total} = 0.5 M	/	$p < 0.05$	$p < 0.05$	$p < 0.05$
pH 3 I_{total} = 3.3 M	$p < 0.05$	/	$p < 0.05$	$p < 0.05$
pH 5 I_{total} = 0.5 M	$p < 0.05$	$p < 0.05$	/	$p < 0.05$
pH 5 I_{total} = 3.3 M	$p < 0.05$	$p < 0.05$	$p < 0.05$	/

Note: If the p value is smaller than 0.05, this indicates that the difference between the two variables in the student's t test is statistically significant. Conversely, if the p value is larger than 0.05, this indicates that the difference is not statically significant (N.S.S.).

Table S13. Statistical analyses (student's t test) for the differences in k_{obs} at different pH and ionic strengths under aqueous aerosol-like conditions for MBO (Figure 4 in the main text).

MBO	pH 3 I_{total} = 0.5 M	pH 3 I_{total} = 3.3 M	pH 5 I_{total} = 0.5 M	pH 5 I_{total} = 3.3 M
pH 3 I_{total} = 0.5 M	/	$p < 0.05$	$p < 0.05$	$p < 0.05$
pH 3 I_{total} = 3.3 M	$p < 0.05$	/	$p < 0.05$	$p < 0.05$
pH 5 I_{total} = 0.5 M	$p < 0.05$	$p < 0.05$	/	$p < 0.05$
pH 5 I_{total} = 3.3 M	$p < 0.05$	$p < 0.05$	$p < 0.05$	/

Note: If the p value is smaller than 0.05, this indicates that the difference between the two variables in the student's t test is statistically significant. Conversely, if the p value is larger than 0.05, this indicates that the difference is not statically significant (N.S.S.).

Table S14. Statistical analyses (student's *t* test) for the differences in Y_{SOA} between pH and ionic strength under aqueous aerosol-like conditions for cHxO (Figure 5 in the main text).

cHxO	pH 3 I_{total} = 0.5 M	pH 3 I_{total} = 3.3 M	pH 5 I_{total} = 0.5 M	pH 5 I_{total} = 3.3 M
pH 3 I_{total} = 0.5 M	/	$p < 0.05$	N.S.S.	$p < 0.05$
pH 3 I_{total} = 3.3 M	$p < 0.05$	/	$p < 0.05$	$p < 0.05$
pH 5 I_{total} = 0.5 M	N.S.S.	$p < 0.05$	/	$p < 0.05$
pH 5 I_{total} = 3.3 M	$p < 0.05$	$p < 0.05$	$p < 0.05$	/

Note: If the *p* value is smaller than 0.05, this indicates that the difference between the two variables in the student's *t* test is statistically significant. Conversely, if the *p* value is larger than 0.05, this indicates that the difference is not statically significant (N.S.S.).

Table S15. Statistical analyses (student's *t* test) for the differences in Y_{SOA} between pH and ionic strength under Aqueous aerosol-like conditions for tHxO (Figure 5 in the main text).

tHxO	pH 3 I_{total} = 0.5 M	pH 3 I_{total} = 3.3 M	pH 5 I_{total} = 0.5 M	pH 5 I_{total} = 3.3 M
pH 3 I_{total} = 0.5 M	/	$p < 0.05$	$p < 0.05$	$p < 0.05$
pH 3 I_{total} = 3.3 M	$p < 0.05$	/	$p < 0.05$	N.S.S.
pH 5 I_{total} = 0.5 M	$p < 0.05$	$P < 0.05$	/	$p < 0.05$
pH 5 I_{total} = 3.3 M	$p < 0.05$	N.S.S.	$p < 0.05$	/

Note: If the *p* value is smaller than 0.05, this indicates that the difference between the two variables in the student's *t* test is statistically significant. Conversely, if the *p* value is larger than 0.05, this indicates that the difference is not statically significant (N.S.S.).

Table S16. Statistical analyses (student's t test) for the differences in Y_{SOA} between pH and ionic strength under Aqueous aerosol-like conditions for tPtO (Figure 5 in the main text).

tPtO	pH 3 I_{total} = 0.5 M	pH 3 I_{total} = 3.3 M	pH 5 I_{total} = 0.5 M	pH 5 I_{total} = 3.3 M
pH 3 I_{total} = 0.5 M	/	$p < 0.05$	$p < 0.05$	$p < 0.05$
pH 3 I_{total} = 3.3 M	$p < 0.05$	/	$p < 0.05$	$p < 0.05$
pH 5 I_{total} = 0.5 M	$p < 0.05$	$p < 0.05$	/	$p < 0.05$
pH 5 I_{total} = 3.3 M	$p < 0.05$	$p < 0.05$	$p < 0.05$	/

Note: If the p value is smaller than 0.05, this indicates that the difference between the two variables in the student's t test is statistically significant. Conversely, if the p value is larger than 0.05, this indicates that the difference is not statically significant (N.S.S.).

Table S17. Statistical analyses (student's t test) for the differences in Y_{SOA} between pH and ionic strength under Aqueous aerosol-like conditions for MBO (Figure 5 in the main text).

MBO	pH 3 I_{total} = 0.5 M	pH 3 I_{total} = 3.3 M	pH 5 I_{total} = 0.5 M	pH 5 I_{total} = 3.3 M
pH 3 I_{total} = 0.5 M	/	$p < 0.05$	N.S.S.	$p < 0.05$
pH 3 I_{total} = 3.3 M	$p < 0.05$	/	$p < 0.05$	N.S.S.
pH 5 I_{total} = 0.5 M	N.S.S.	$p < 0.05$	/	$p < 0.05$
pH 5 I_{total} = 3.3 M	$p < 0.05$	N.S.S.	$p < 0.05$	/

Note: If the p value is smaller than 0.05, this indicates that the difference between the two variables in the student's t test is statistically significant. Conversely, if the p value is larger than 0.05, this indicates that the difference is not statically significant (N.S.S.).

References

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