Supplemental Information

Table S1. pH-indicator papers used in this study.

Type Nr. (classified in this work)	pH Range	Note
Type I	0.0 – 2.5	
Type II	2.5 – 4.5	
Type III	4.0 – 7.0	
Type IV	0.5 – 5.5	Model and producer for the Type V pH paper: Hydrion® Brilliant pH dip stiks, Lot Nr. 3110, Sigma-Aldrich
Type V	0.0 – 6.0	
Type VI	0.0 – 5.0	
Type VII	1.0 – 11.0	
Type VIII	1.0 – 14.0	
Type IX	0.0 – 14.0	

Table S2. Detailed information about the buffer solutions used in this study.

Self-prepared Buffer Solutions		Purchased Buffer Solutions			
Measured pH value[a]	SD[b]	Specified pH value[c]	Measured pH value[a]	SD[b]	Manufacturer
0.03	0.06	1.00	1.00	0.042	Carl Roth GmbH + Co. KG
0.61	0.043	1.68	1.66	0.036	VWR CHEMICALS
2.48	0.025	2.00	1.99	0.045	Fisher Scientific U.K. Limited
2.83	0.025	3.00	2.99	0.045	neoFroxx GmbH
3.58	0.026	4.00	4.00	0.042	neoFroxx GmbH
4.38	0.029	5.00	5.01	0.045	Fisher Scientific U.K. Limited
5.50	0.029	6.00	6.01	0.042	Fisher Scientific U.K. Limited
6.46	0.025	7.00	7.00	0.042	Fisher Scientific U.K. Limited

a: averaged pH values measured by the pH meter; b: standard deviations of three replicate pH measurements by the pH meter; c: pH values specified on the purchased buffer solution bottles.

Table S3. Composition of the salt systems used for interference check for different types of pH indicator papers.

Inorganic Acid Systems	Organic Acid Systems	Inorganic + Organic Acids Systems
$MgSO_4$-H_2SO_4	$MgSO_4$-$C_2H_2O_4$	$MgSO_4$-H_2SO_4-$C_2H_2O_4$
Na_2CO_3-HCl	Na_2CO_3-$C_2H_2O_4$	Na_2CO_3-HCl-$C_2H_2O_4$
$NaNO_3$-HNO_3	$NaNO_3$-$C_2H_2O_4$	$NaNO_3$-HNO_3-$C_2H_2O_4$
Na_2SO_4-H_2SO_4	Na_2SO_4-$C_2H_2O_4$	Na_2SO_4-H_2SO_4-$C_2H_2O_4$
NH_4NO_3-HNO_3	NH_4NO_3-$C_2H_2O_4$	NH_4NO_3-HNO_3-$C_2H_2O_4$
$(NH_4)_2SO_4$-H_2SO_4	$(NH_4)_2SO_4$-$C_2H_2O_4$	$(NH_4)_2SO_4$-H_2SO_4-$C_2H_2O_4$
Na_2HPO_4-H_2SO_4	Na_2HPO_4- $C_6H_8O_7$	NH_4NO_3-HNO_3-$C_3H_4O_4$
		$(NH_4)_2SO_4$-H_2SO_4-$C_3H_4O_4$

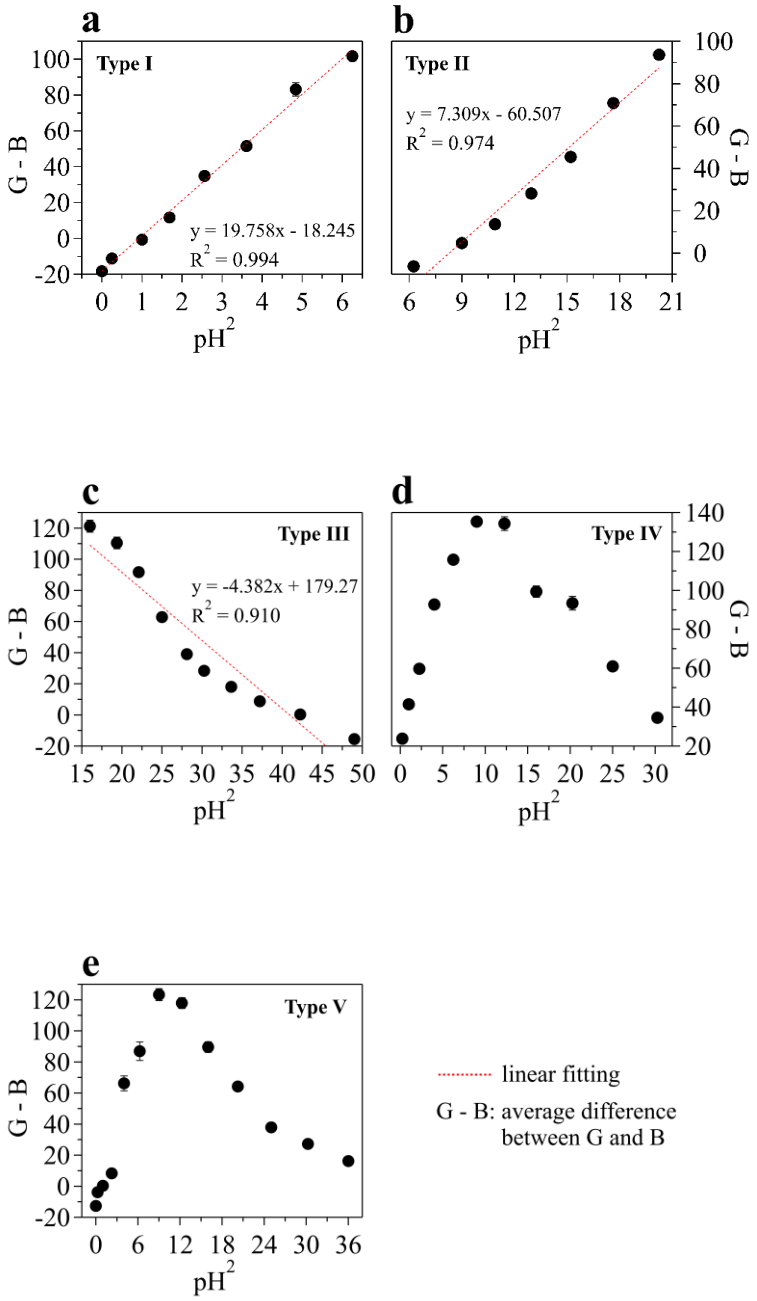


Figure S1. Relationship between the average difference of G and B (G – B) and pH^2, derived from the color charts of five different pH papers: Type I: 0 – 2.5, Type II: 2.5 – 4.5, Type III: 4.0 – 7.0, Type IV: 0.5 – 5.5 and Type V: 0 – 6.0 (summarized in Table S1).

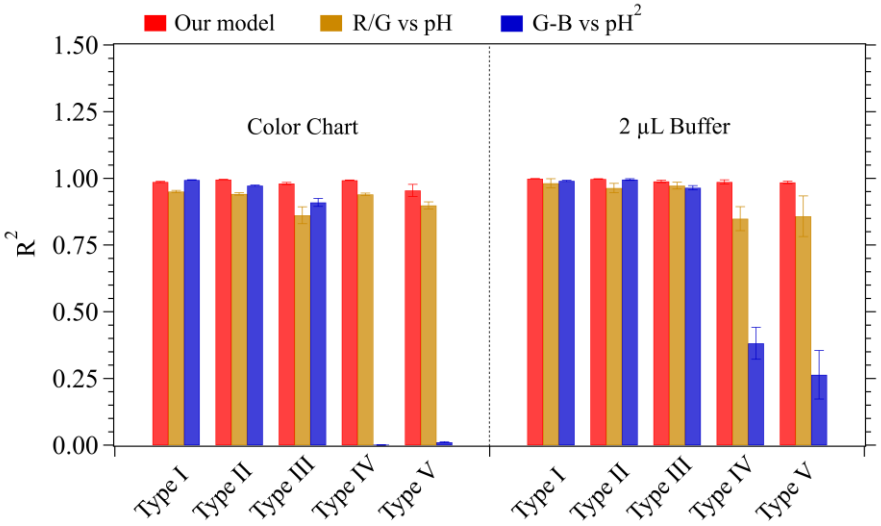


Figure S2. Comparison of the established linear correlation (R^2) using different RGB models for the first five types of pH papers adopted in this study. For each type, the comparison is made for its color chart and samples of 2 µL buffers, respectively. The model R/G vs pH was used by Selva Kumar et al. (2018) and G-B vs pH^2 by Craig et al. (2018). The error bars represent the standard deviation of five to six replicate experiments.

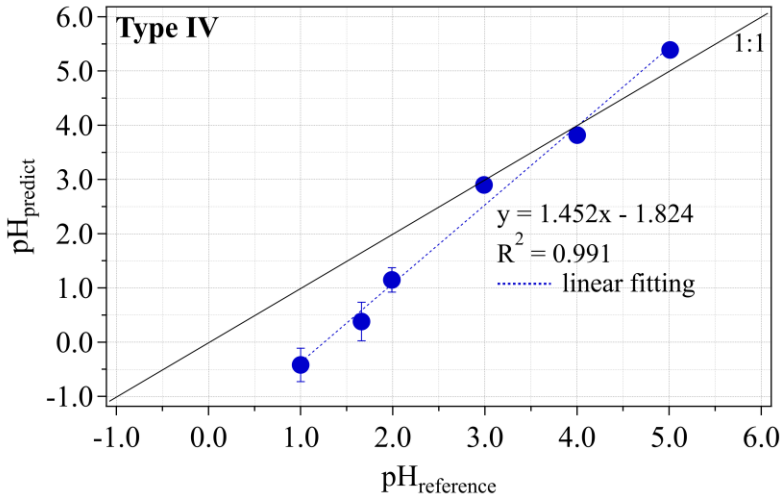


Figure S3. $pH_{predict}$ versus $pH_{reference}$ for 2 μL buffer samples on the type IV pH paper. The $pH_{predict}$ are calculated using the coefficient vector [*a*, *b*, *c*] derived from regression analysis on the color chart. The error bars represent the standard deviation of five replicate experiments.

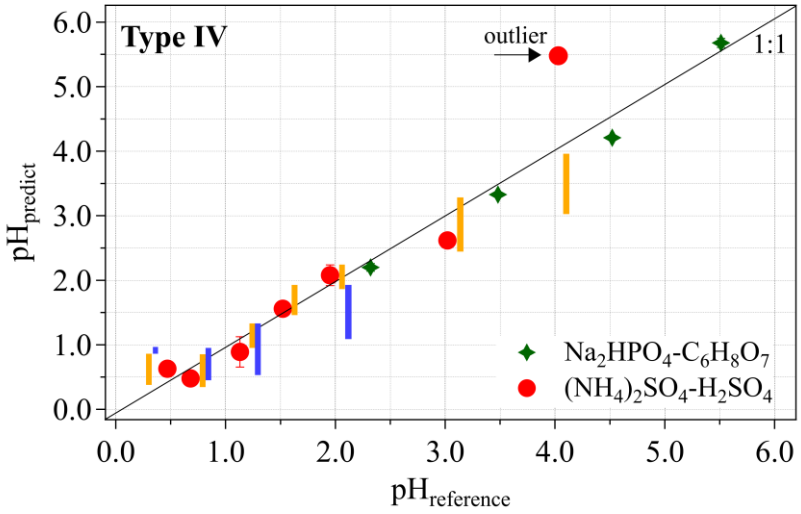


Figure S4. Estimation of samples pH using the type IV pH paper. The adopted samples include a series of 2 µL lab-prepared aerosol surrogates ($(NH_4)_2SO_4$-H_2SO_4, red dot) and self-prepared buffers (Na_2HPO_4-$C_6H_8O_7$, green star). $pH_{predict}$ are calculated with the averaged coefficient vector [*a*, *b*, *c*] derived from the standard buffers from three to six replicate experiments under constant photographing conditions. The error bars represent the standard deviation of three to six replicate experiments. The heights of the orange and blue bars indicate the reported pH ranges measured with pH papers and Raman spectroscopy respectively, for $(NH_4)_2SO_4$ - H_2SO_4 aerosols with particle sizes larger than 2.5 µm in Craig et al. (2018). Note that, each orange or blue bar has the same $pH_{reference}$ as of the red symbol close to it.

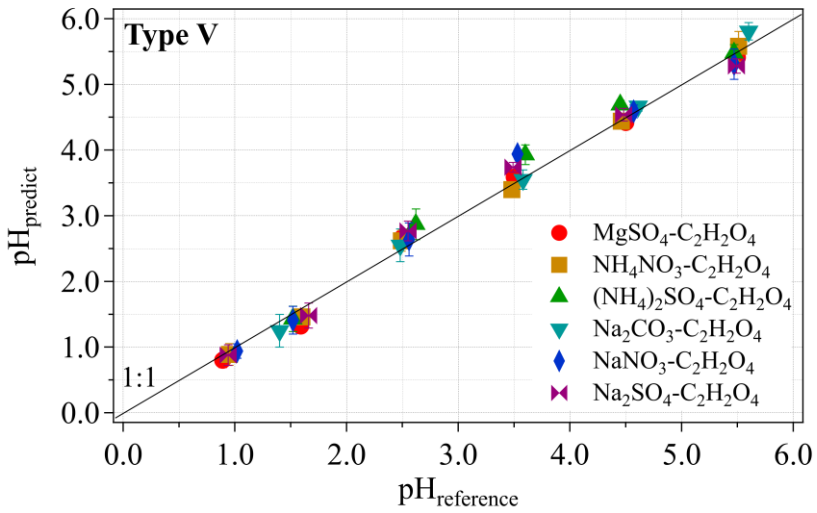


Figure S5. pH estimation using the type V pH paper for salt systems with oxalic acid. $pH_{predict}$ are calculated with the averaged coefficient vector [*a*, *b*, *c*] derived from three replicate calibration experiments with 2 µL standard buffers and under constant photographing conditions. The error bars represent the standard deviation of three to four replicate experiments.

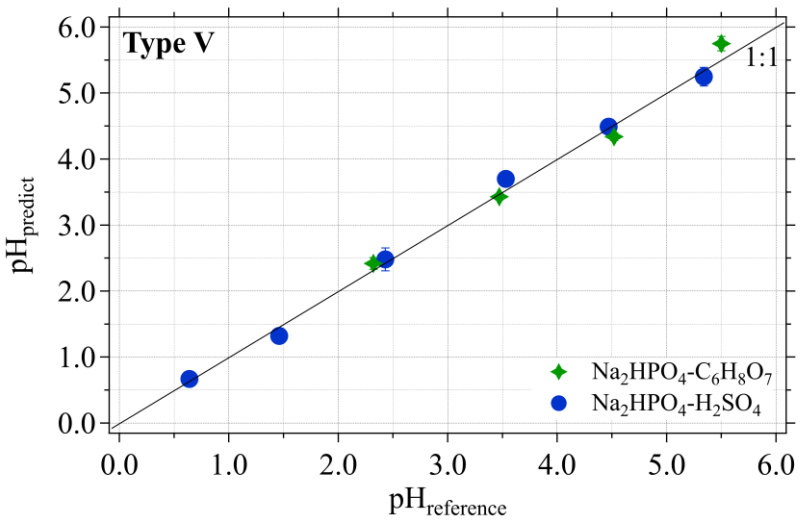
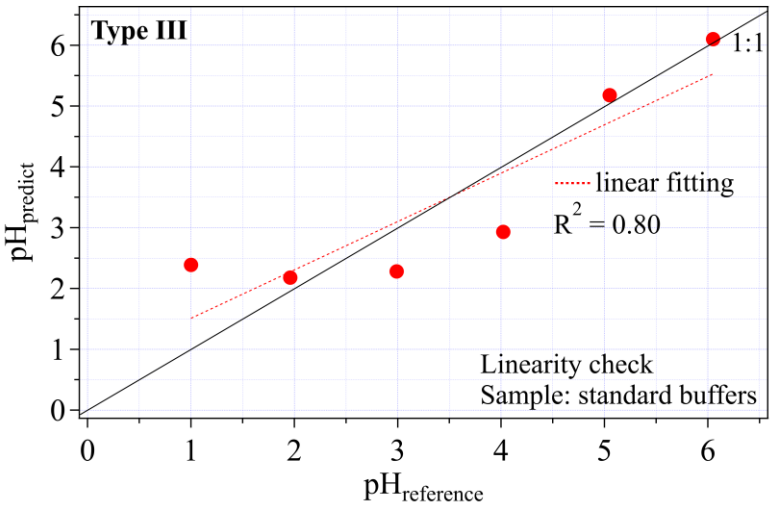
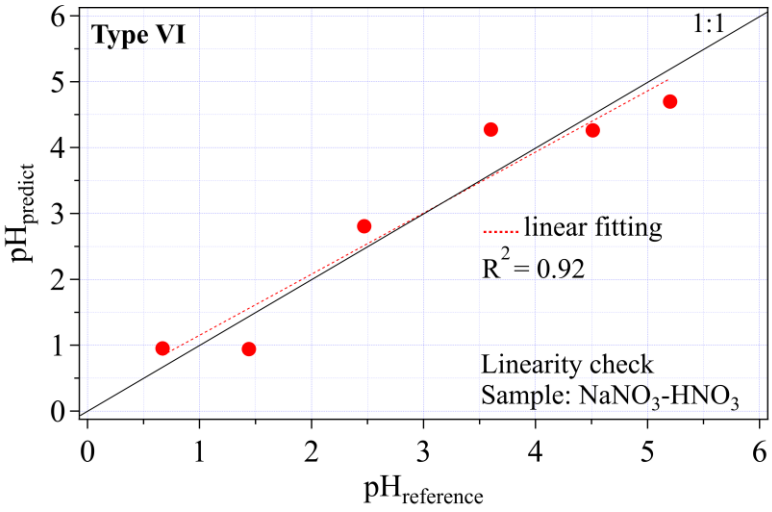


Figure S6. pH estimation using the type V pH paper for phosphate systems. $pH_{predict}$ are calculated with the averaged coefficient vector [*a*, *b*, *c*] derived from three replicate calibration experiments with 2 µL standard buffers and under constant photographing conditions. The error bars represent the standard deviation of three replicate experiments.

Interference test for different types of pH indicator papers (Type III and VI-IX)

The test includes two steps: Step1 is to check the established linearity between $pH_{predict}$ and $pH_{reference}$ directly with Eqn (5); Step2 is to predict sample pH (with Eqn (4)) using the coefficient vector [*a*, *b*, *c*] derived from linear regression analyses on standard buffers (i.e. the standard-buffer-calibration method) and is only conducted when the linearity check (in Step1) provides a good linearity with $R^2 \geq 0.95$. Note that if the linearity check in Step1 gives a poor linearity ($R^2 < 0.95$), this result is enough (the obtained R^2 in Step1 is always larger than the R^2 in Step2) to demonstrate the tested pH paper is not suitable for pH measurements of atmospheric aerosols due to either its limited ability to achieve accurate pH or potential interferences from various chemical compositions common in aerosols. As shown in Fig. S7, for the Step1 test, poor linearity is found for type III, VI, VII and IX pH papers against different test samples whose pH were measured by a pH bench meter beforehand. Good linearity in Step1 ($R^2 \geq 0.95$), however, cannot guarantee the good applicability of a tested pH paper for aerosol pH estimation and therefore needs to be checked through Step2 to further inspect the effectiveness of the adopted colorimetric method on this specific pH paper type. In Fig. S7, the type VIII pH paper still predicts largely deviated $pH_{predict}$ when $pH_{reference}$ in the range of 4 – 6, even though it presents a good linearity in the Step1 linearity check. All the results demonstrate that these tested pH paper types are not suitable for pH measurement of ambient aerosols.





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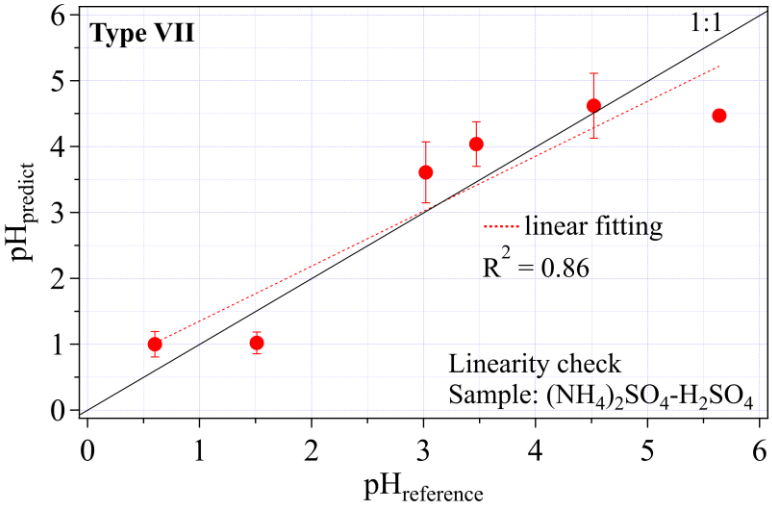

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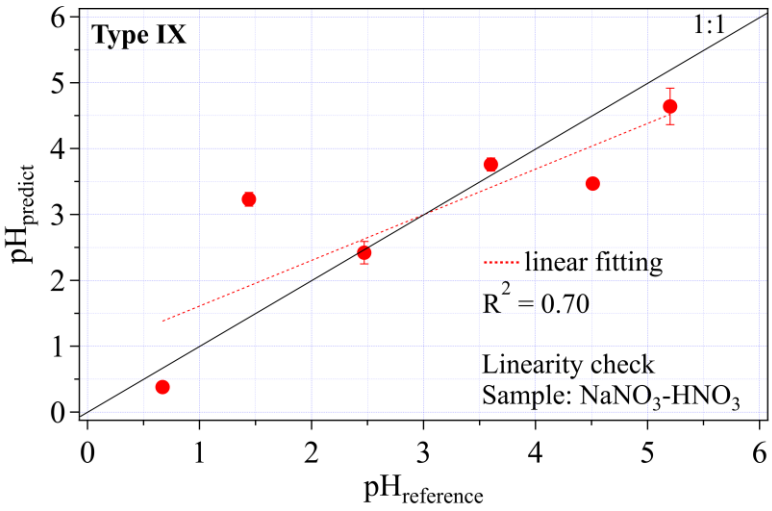

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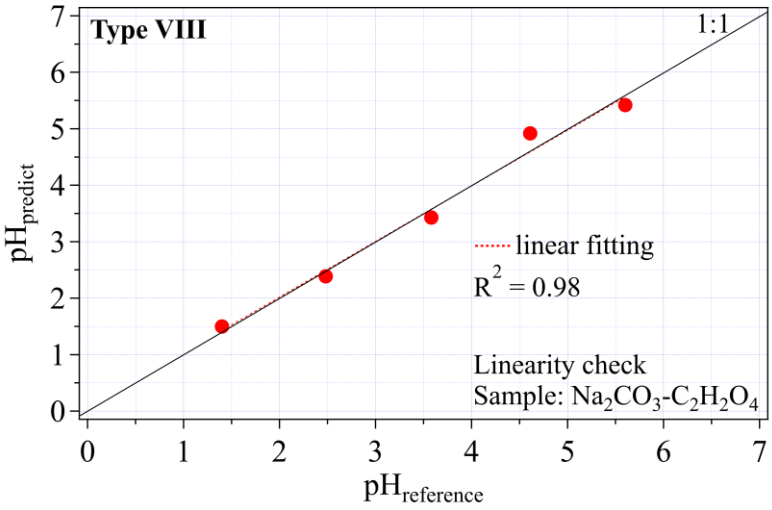

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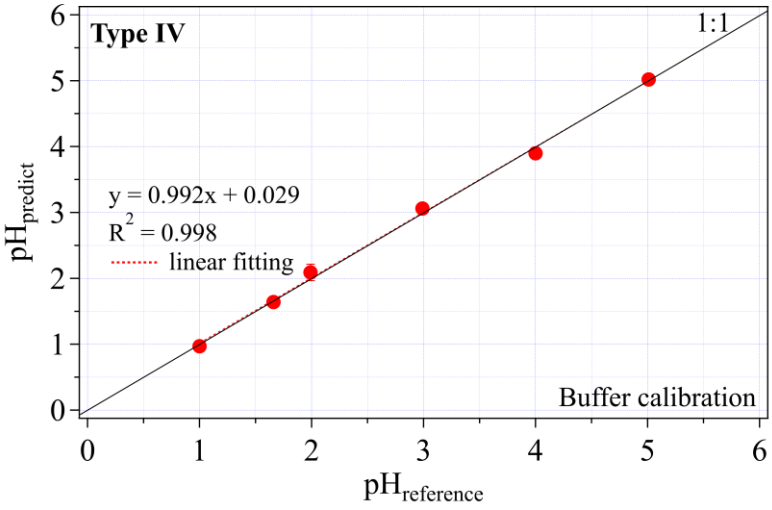

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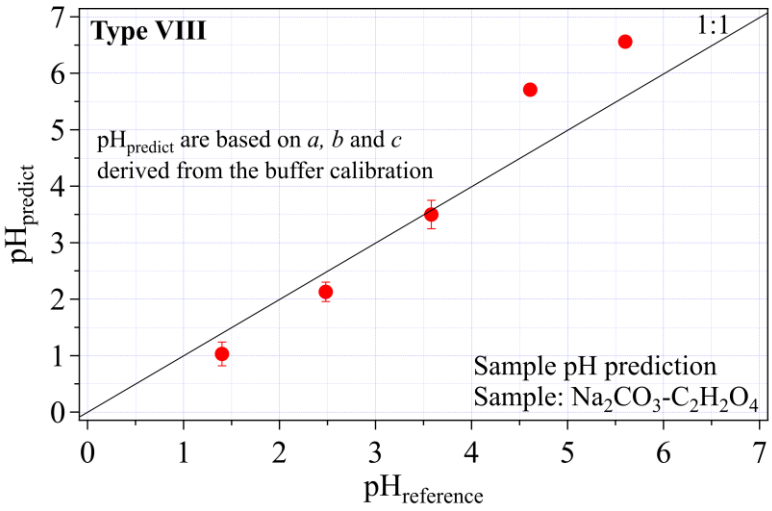

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260 **Figure S7.** Interference check for different types of pH indicator papers through lab-prepared aerosol surrogates
261 (2 µL). The error bars represent the standard deviation of three to four replicate experiments.

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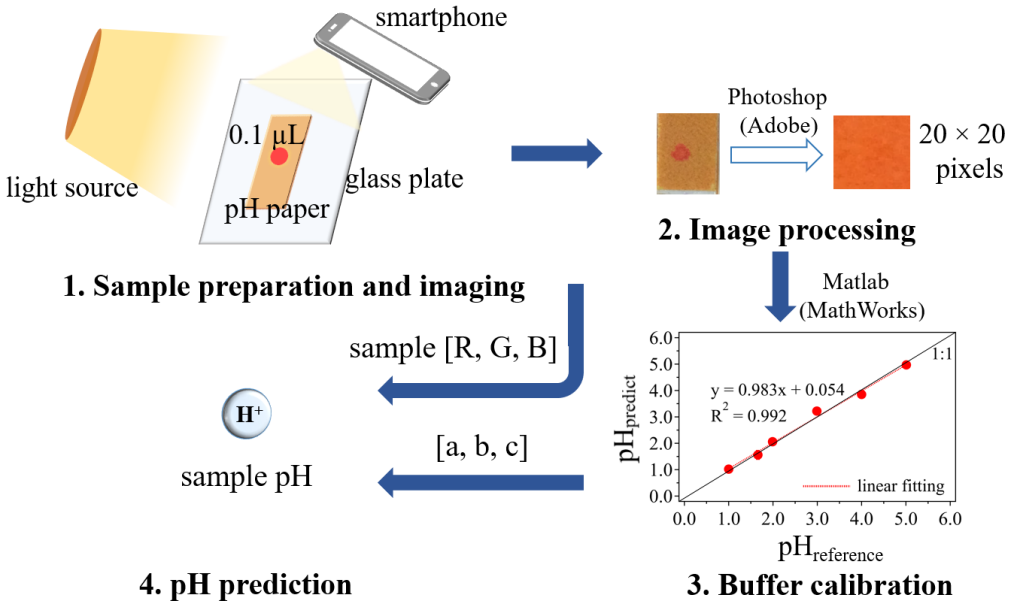

Figure S8. Schematic of using the RGB-based colorimetric method for pH estimation of 0.1 μL aerosol samples. Note that when using the buffer calibration results to predict the pH of aerosol samples, the photographing conditions for the samples are the same as those of the buffer calibration.

References

Craig, R. L., Peterson, P. K., Nandy, L., Lei, Z., Hossain, M. A., Camarena, S., Dodson, R. A., Cook, R. D., Dutcher, C. S., and Ault, A. P.: Direct Determination of Aerosol pH: Size-Resolved Measurements of Submicrometer and Supermicrometer Aqueous Particles, Analytical Chemistry, 90, 11232-11239, 10.1021/acs.analchem.8b00586, 2018.

Selva Kumar, R., Kumar, S. K. A., Vijayakrishna, K., Sivaramakrishna, A., Brahmmananda Rao, C. V. S., Sivaraman, N., and Sahoo, S. K.: Development of the Smartphone-Assisted Colorimetric Detection of Thorium by Using New Schiff's Base and Its Applications to Real Time Samples, Inorganic Chemistry, 57, 15270-15279, 10.1021/acs.inorgchem.8b02564, 2018.