

# SUPPLEMENTARY MATERIAL

## **Fine particle pH and its impact on PM<sub>2.5</sub> control in a megacity of Central China**

Mingming Zheng <sup>1</sup>, Ke Xu <sup>2</sup>, Lianxin Yuan <sup>2</sup>, Nan Chen <sup>2\*</sup>, Menghua Cao <sup>4\*</sup>

<sup>1</sup>*School of Chemical and Environmental Engineering, Wuhan Polytechnic University,*

*Wuhan, 430023, China*

<sup>2</sup>*Hubei Environmental Monitoring Center, Wuhan, 430072, China*

<sup>3</sup>*College of Resources and Environment, Huazhong Agricultural University, Wuhan*

*430070, China*

*Corresponding author: Nan Chen (nanchen2020@126.com), Menghua Cao  
(caomenghua@mail.hzau.edu.cn)*

**The SM contains 1 text, 5 figures and 1 table.**

### **Text 1 Instrument descriptions**

Water-soluble ions including  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{K}^+$ ,  $\text{Na}^+$ ,  $\text{NH}_4^+$ ,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ , and  $\text{Cl}^-$  in  $\text{PM}_{2.5}$  and gases including  $\text{HCl}$ ,  $\text{HNO}_3$ , and  $\text{NH}_3$  were synchronously observed using an online ion chromatograph (Model ADI 2080, Metrohm Applikon B.V., Schiedam, Netherlands). A wet rotating denuder was used to concentrate the gases. At a rate of  $16.7 \text{ L min}^{-1}$ , The aerosol was collected with a steam jet aerosol collector. The extracts of particles and gases were monitored by ion chromatography. There was a regular calibration and routine maintenance of the instrument. The sampling flow was calibrated within 5%. Internal calibration by using LiBr was conducted hourly in each analysis of the sample. The RSD (relative standard deviation) was lower than 10%. Monthly, the standard mixed cationic and anionic solution ( $\text{NH}_4^+$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{K}^+$ ,  $\text{Na}^+$ ,  $\text{Br}^-$ ,  $\text{Cl}^-$ ,  $\text{Li}^+$ ,  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$ ) was conducted to establish the standard calibration curve, with recovery rates higher than 95%. hydrogen peroxide or peracetic acid were used for routine liquid flow path cleaning to disinfect bacterial.

The in-situ ambient temperature (Temp) and relative humidity (RH) were monitored hourly using an automatic weather station (LUFFT WS600).

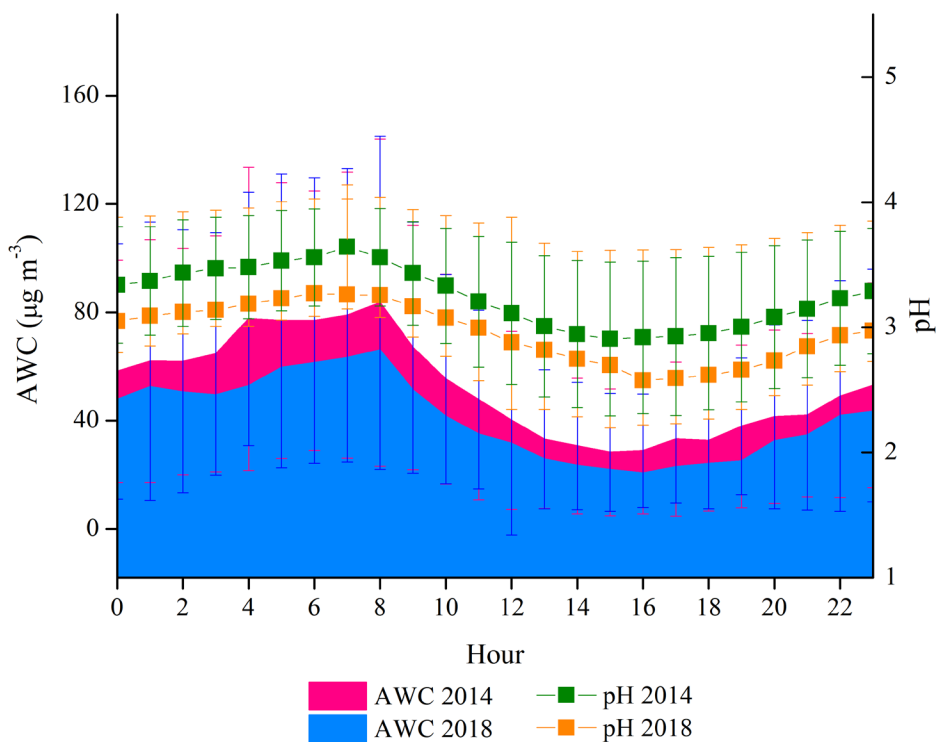


Figure S1 Diurnal variations of fine particle pH and aerosol water content (AWC) in 2014 and 2018.

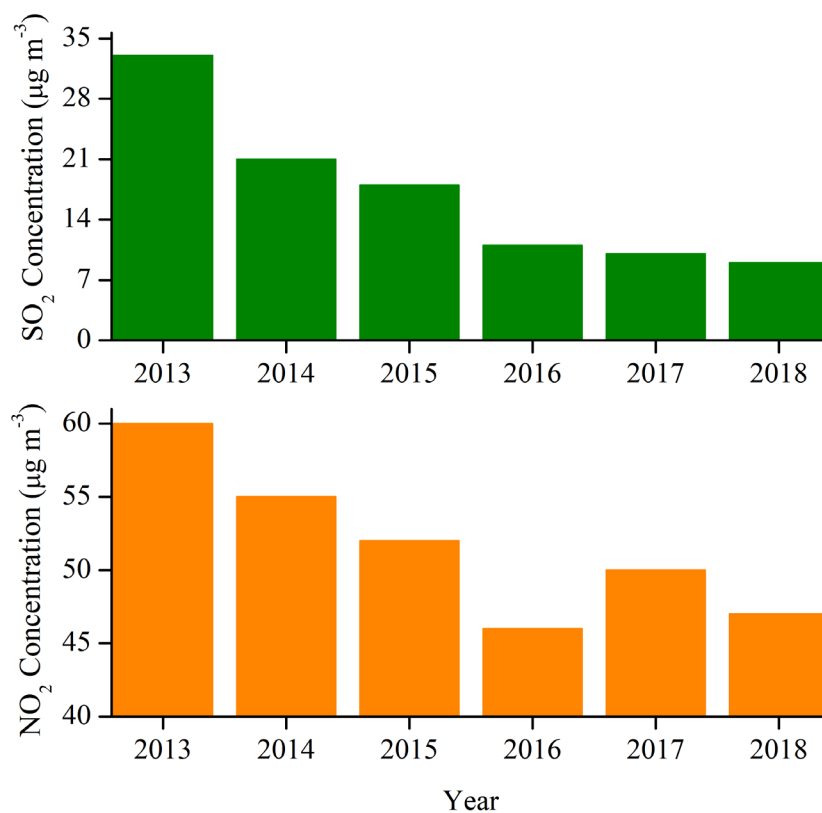


Figure S2 Atmospheric  $\text{SO}_2$  and  $\text{NO}_x$  concentrations from 2013 to 2018 (consecutive observations at the same site as water-soluble ions).

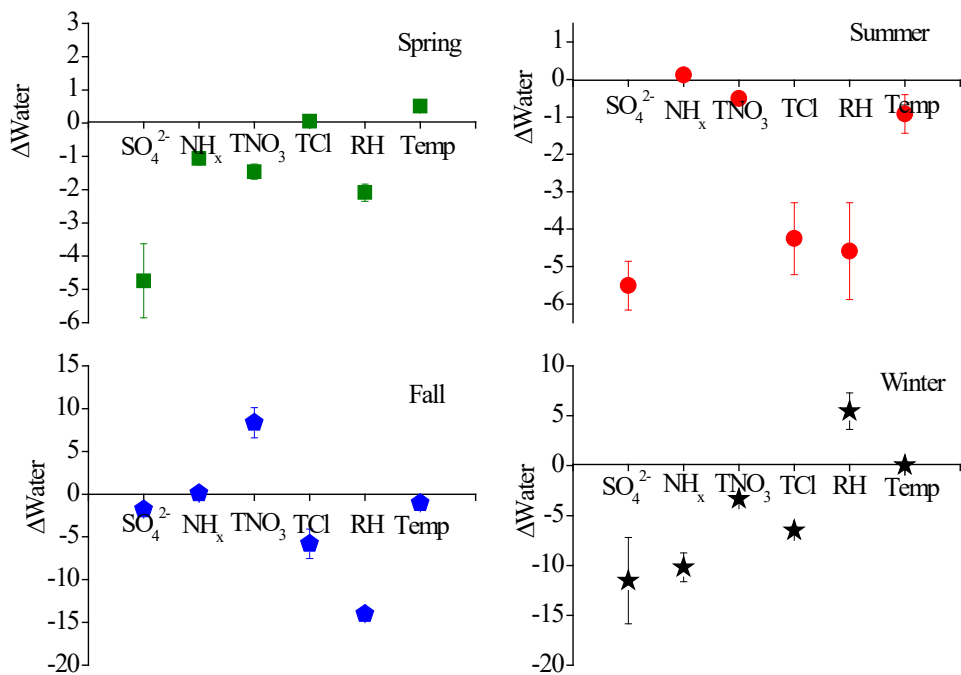


Figure S3 Factors contributing to AWC changes from 2014 to 2018.

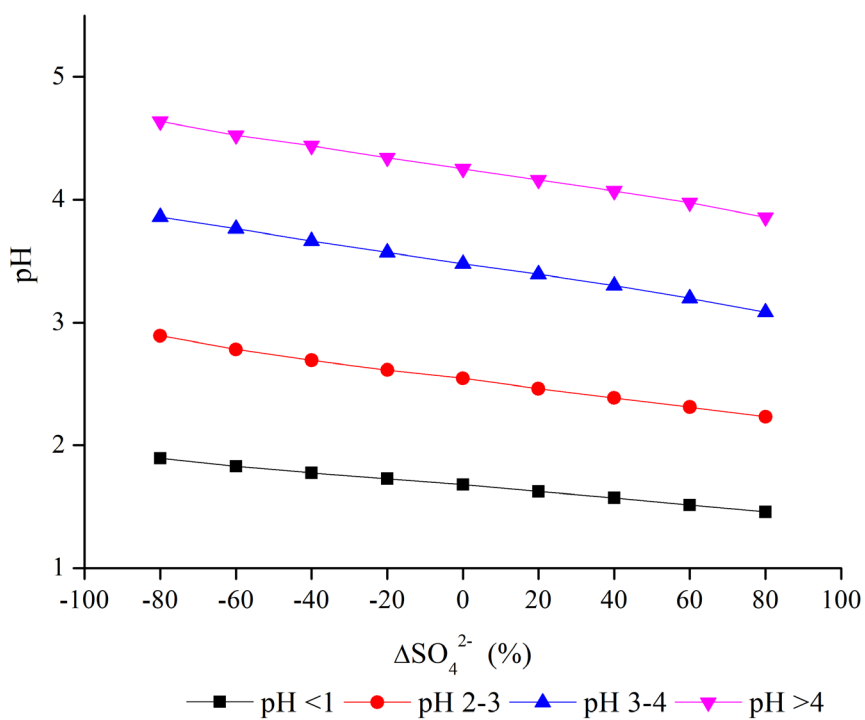


Figure S4 Fine particle pH as functions of sulfate changes at different pH ranges.

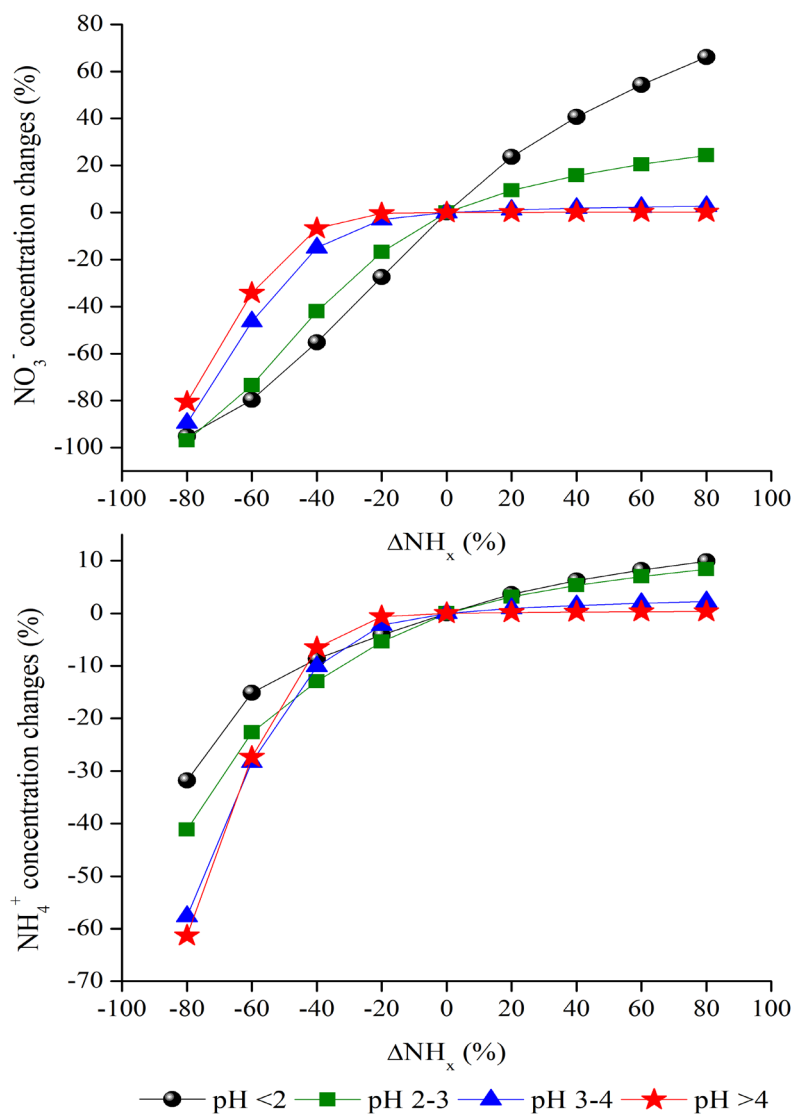


Figure S5 Changes of  $\text{NO}_3^-$  and  $\text{NH}_4^+$  concentrations as functions of  $\Delta\text{NH}_x$  in different pH ranges.

Table S1 Correlation coefficients of particulate inorganic ions in 2014 and 2018.

	Cl <sup>-</sup>	NO <sub>3</sub> <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	NH <sub>4</sub> <sup>+</sup>	Na <sup>+</sup>	K <sup>+</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>
Cl <sup>-</sup>	1	0.62	0.15	0.52	0.32	0.54	0.29	0.23
NO <sub>3</sub> <sup>-</sup>	0.28	1	0.40	0.87	-0.03	0.55	0.30	0.18
SO <sub>4</sub> <sup>2-</sup>	0.21	0.73	1	0.59	-0.32	0.31	0.14	0.08
NH <sub>4</sub> <sup>+</sup>	0.42	0.91	0.86	1	-0.31	0.55	0.26	0.14
Na <sup>+</sup>	0.28	0.51	0.34	0.46	1	0.24	-0.10	-0.06
K <sup>+</sup>	0.34	0.38	0.34	0.36	0.33	1	0.23	0.21
Mg <sup>2+</sup>	0.30	0.28	0.20	0.26	0.33	0.76	1	0.53
Ca <sup>2+</sup>	0.30	0.30	0.12	0.27	0.52	0.30	0.65	1

The correlation coefficients were based on hourly-observed data.

All correlation is significant at 0.01 level except one cell at 0.05 level.

Orange: 2014.

Blue: 2018.