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Supplement of

Long-term trends and drivers of aerosol pH in eastern China

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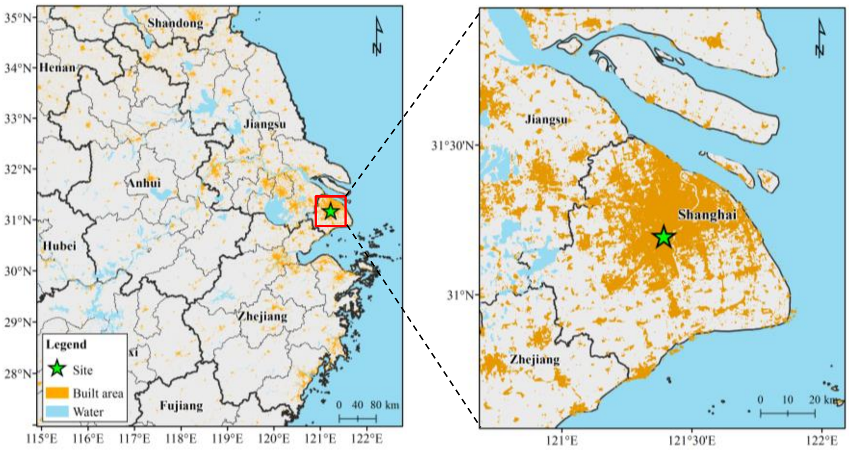

Figure S1 Location of the sampling site

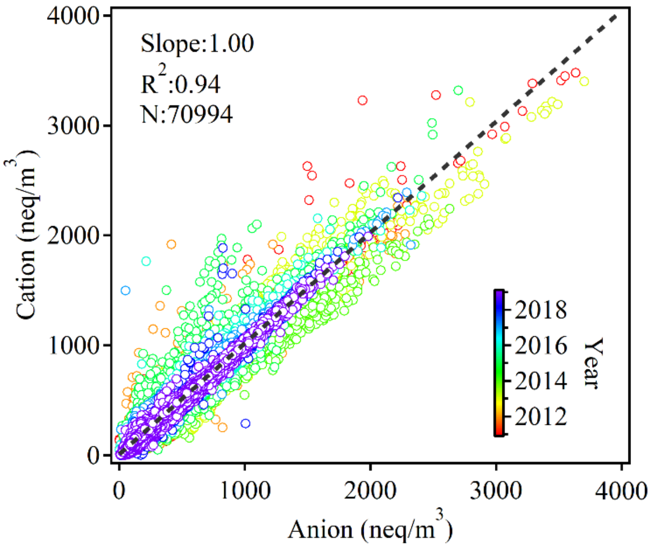

Figure S2 Comparisons of anions and cations in Shanghai during 2011-2019

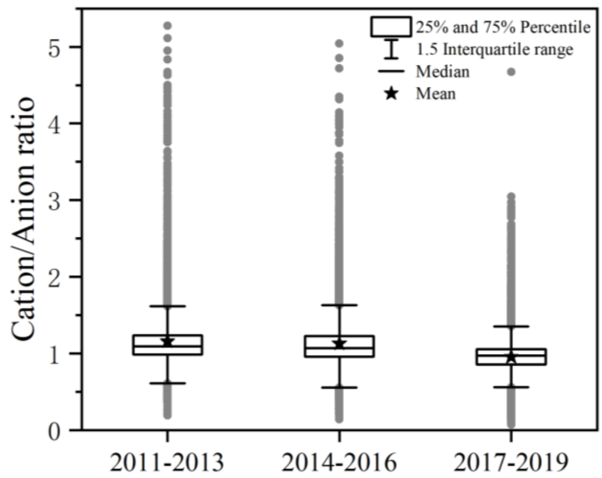

Figure S3 The box plots of Cation/Anion ratios during 2011-2013, 2014-2016, and 2017-2019

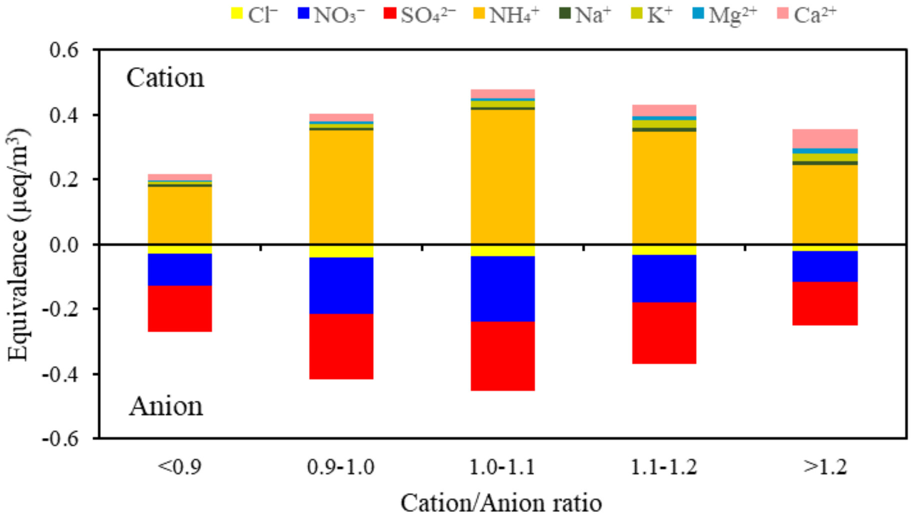

Figure S4 Average equivalence concentrations of cation and anion at different level of Cation/Anion ratio

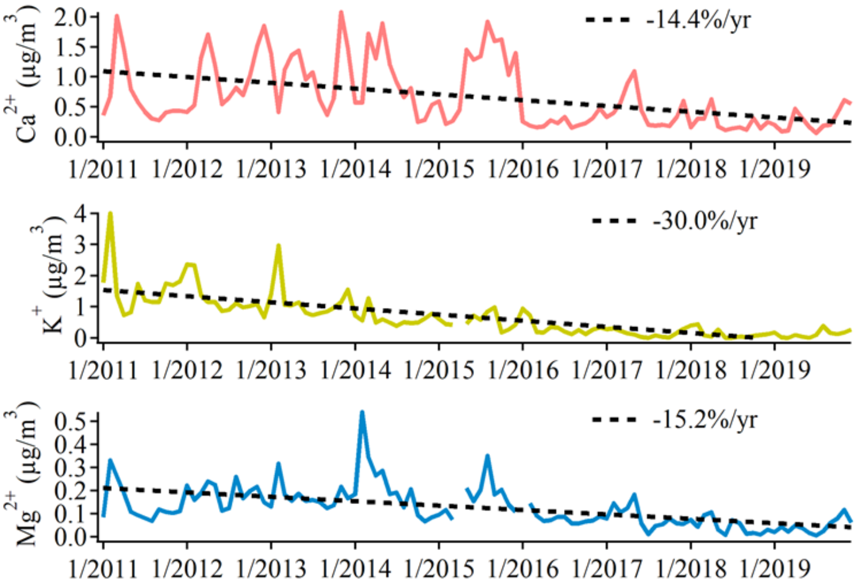

Figure S5 Monthly mean concentrations of Ca^{2+}, K^{+} and Mg^{2+} from 2011 to 2019

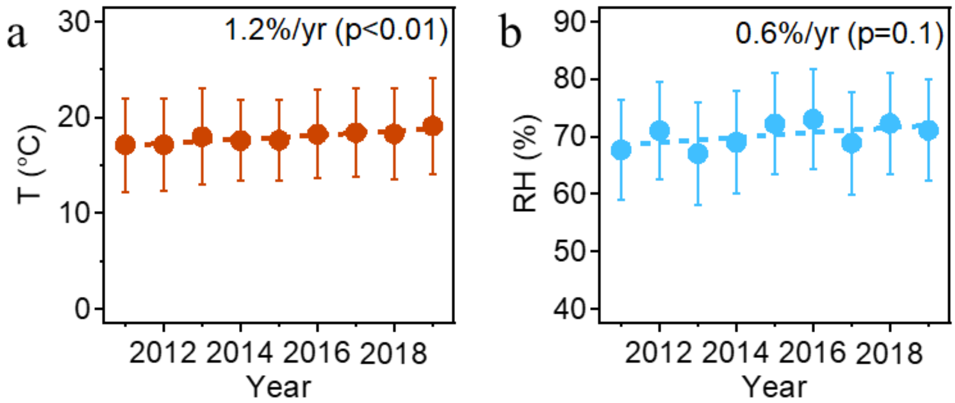

Figure S6 Annual values of temperature (T) and relative humidity (RH) from 2011 to 2019

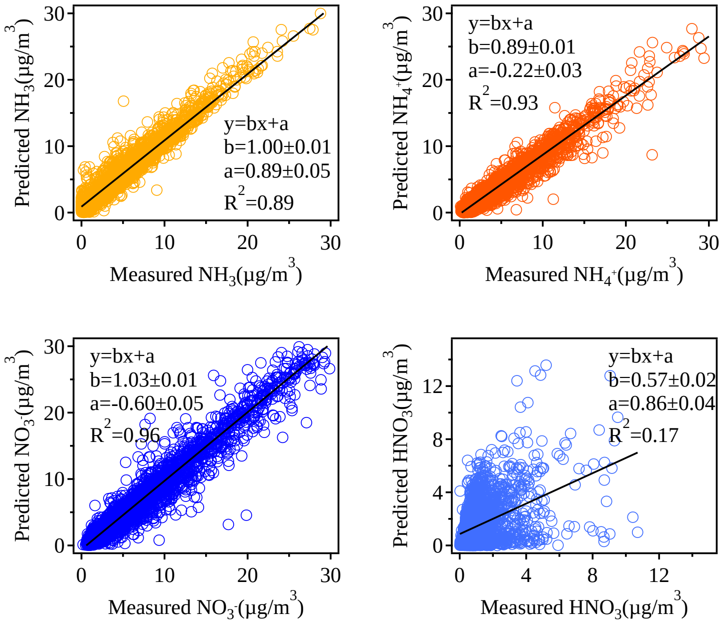

Figure S7 Comparisons of predicted and measured NH_3, NH_4^+, NO_3^- and HNO_3 in Shanghai during 2011-2019

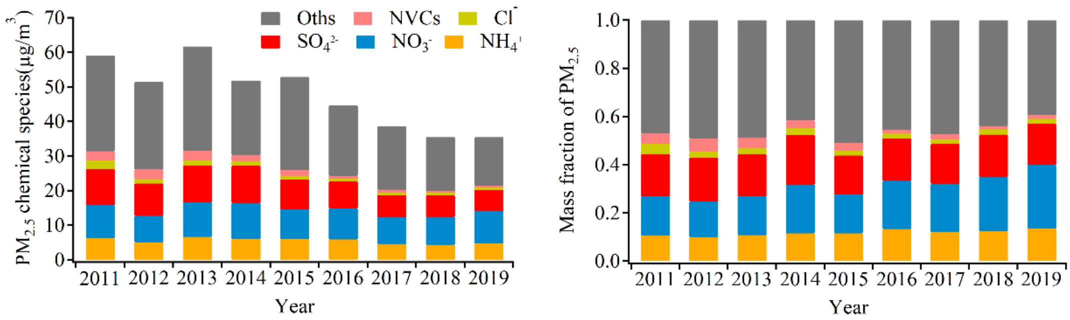

Figure S8 Annual variations of different chemical species in $PM_{2.5}$ from 2011 to 2019

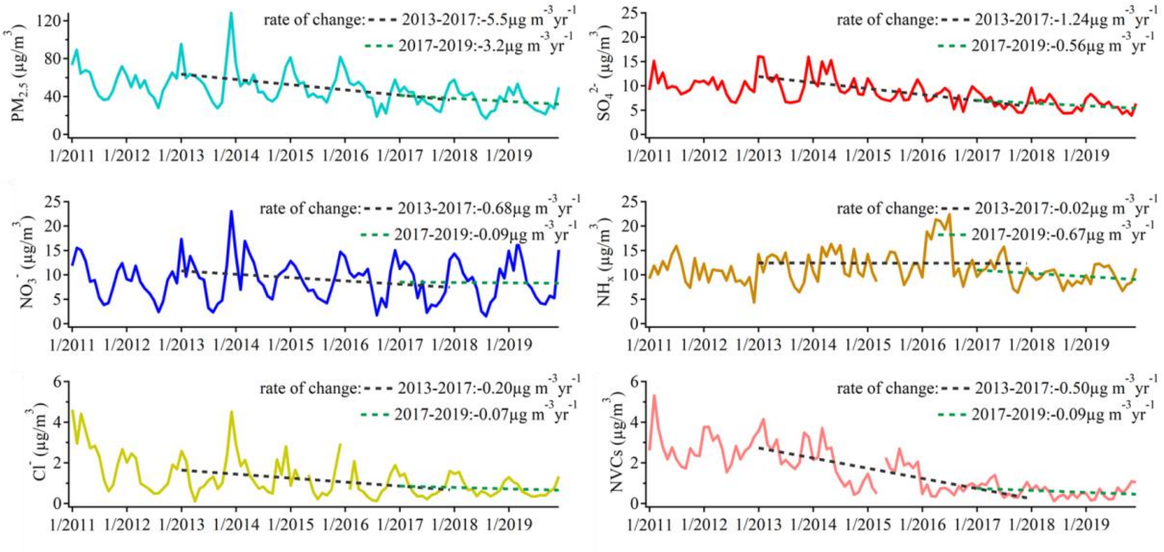

Figure S9 Monthly mean of $PM_{2.5}$, SO_4^{2-}, NO_3^-, NH_x, Cl^- and NVCs from 2011 to 2019. The years of 2011-2013, 2013-2017 and 2017-2019 represent the Pre-Action Plan, Action Plan and Post-Action Plan period, respectively. Here we focused on the changes in trends between the Action Plan (2013-2017; black dashed lines) and Post-Action Plan (2017-2019; green dashed lines) periods.

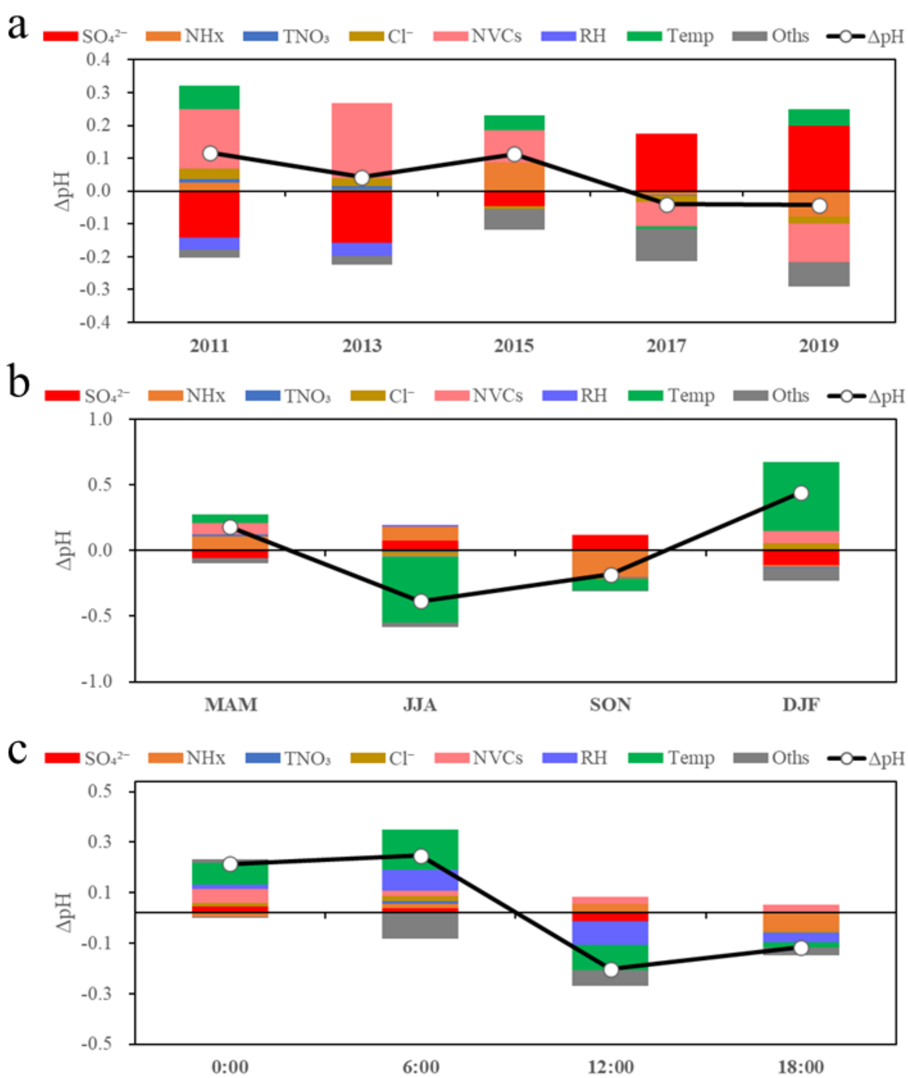

Figure S10. Fractional contribution of individual drivers to the variations in aerosol pH from average conditions (i.e., averages of all observational data) during 2011–2019. (a) Annual variation; (b) Seasonal variation, and (c) diurnal variation. The meanings of the abbreviations: RH, relative humidity; Temp, temperature; NVCs, non-volatile cations; NH_x, total ammonia; TNO_3, total nitrate.

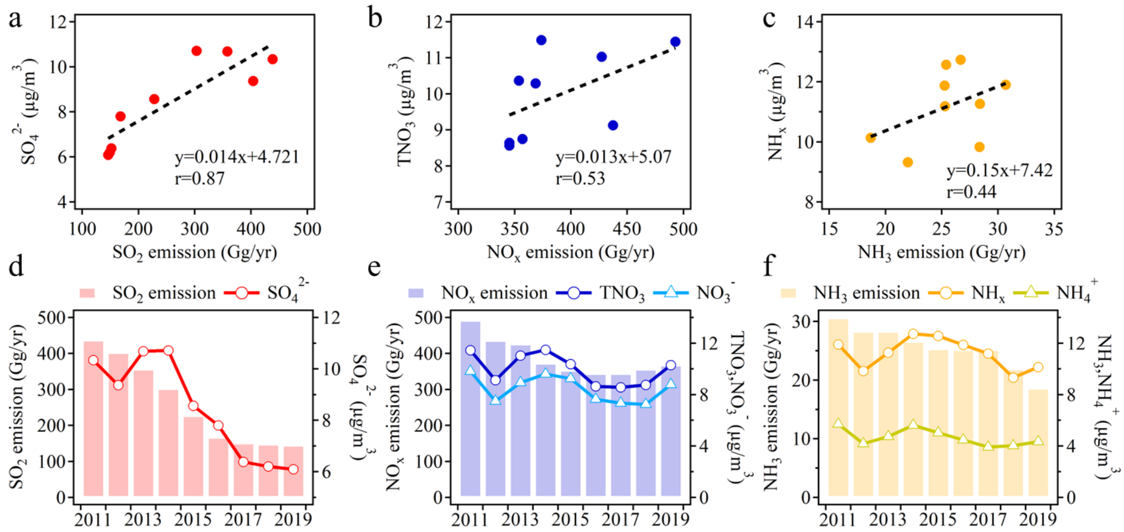

Figure S11 (a)-(c) Correlations of aerosol abundances to precursor emissions from 2011 to 2019, including (a)SO_4^{2-} vs. SO_2, (b) TNO_3 vs. NO_x and (c)NHx vs. NH_3. (d)-(f) Annual values of aerosol abundances and precursor emissions from 2011 to 2019, including (d)SO_4^{2-} and SO_2 emission, (e)TNO_3, NO_3^-, and NO_x emission, and (f) NHx, NH_4^+, and NH_3 emission.

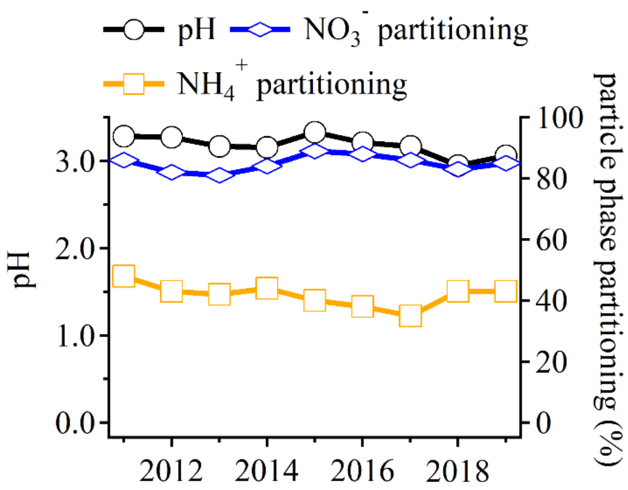

Figure S12 Annual values of pH, NO_3^- partitioning (NO_3^- / (NO_3^- + HNO_3)), and NH_4^+ partitioning (NH_4^+ / (NH_4^+ + NH_3)) from 2011 to 2019.

Table S1 Comparison of the $PM_{2.5}$ pH values in this study with other sites

Summer Site	Aerosol size	Measurement periods	pH	Reference	Winter Site	Aerosol size	Measurement periods	pH	Reference
Guangzhou	$PM_{2.5}$	2013	2.8	(Jia et al., 2018)	Shanghai	$PM_{2.5}$	2011-2019	3.52	This study
Shanghai	$PM_{2.5}$	2011-2019	2.93	This study	Anyang	$PM_{2.5}$	2018	4.8	(Wang et al., 2020)
Tianjin	$PM_{2.5}$	2015	3.4	(Shi et al., 2019)	Zhengzhou	$PM_{2.5}$	2018	4.5	(Wang et al., 2020)
Beijing	$PM_{2.5}$	2014	1.82	(Tan et al., 2018)	Beijing	$PM_{2.5}$	2014	4.11	(Tan et al., 2018)
Beijing	$PM_{2.5}$	2017	3.8	(Ding et al., 2019)	Beijing	$PM_{2.5}$	2017	4.5	(Ding et al., 2019)
Inner Mongolia	$PM_{2.5}$	2014	5	(Wang et al., 2019)	Inner Mongolia	$PM_{2.5}$	2015	5.7	(Wang et al., 2019)
Cabauw	$PM_{2.5}$	2013	3.3	(Guo et al., 2018)	Cabauw	$PM_{2.5}$	2012	3.9	(Guo et al., 2018)
Po Valley	$PM_{2.5}$	2012	2.3	(Masiol et al., 2020)	Po Valley	$PM_{2.5}$	2012/2013	3.9	(Masiol et al., 2020)
JST	$PM_{1.0}$	2011	0.55	(Guo et al., 2015)	JST	PM_1	2012	2.2	(Guo et al., 2015)
Yorkville	$PM_{1.0}$	2012	1.1	(Guo et al., 2015)	Yorkville	PM_1	2012	1.8	(Guo et al., 2015)
Atlanta	$PM_{1.0}$	2012	1.1	(Guo et al., 2015)	Atlanta	PM_1	2012	1.3	(Guo et al., 2015)
Centreville	$PM_{2.5}$	2013	1.1	(Pye et al., 2018)	Yorkville	PM_1	2016	2.2	(Nah et al., 2018)

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