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How alkaline compounds control atmospheric aerosol particle acidity

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Supplementary Materials

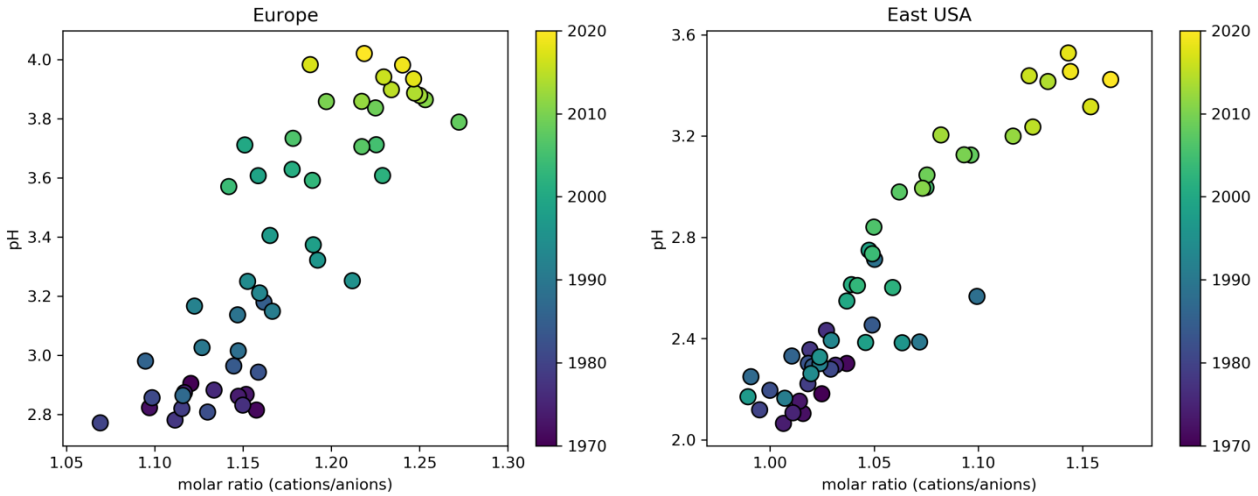

Figure S1: Time evolution of annual average pH as a function of cation/anion molar ratio over Europe (left) and the Eastern USA (right) during the period 1970-2020. The calculated ratio includes all ions from both solid salts and the liquid phase.

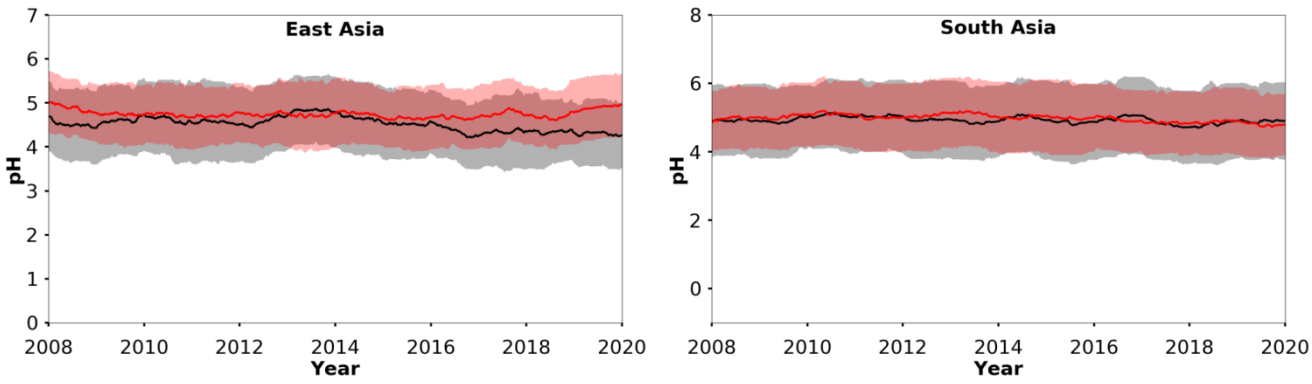

Figure S2: Temporal pH evolution in East and South Asia during the period 2008-2020. Black lines represent the reference simulation. Red lines show the sensitivity simulation in which SO_2 emissions are reduced by 75% in East Asia and increased by 50% in South Asia. Ranges represent the 1σ standard deviation.

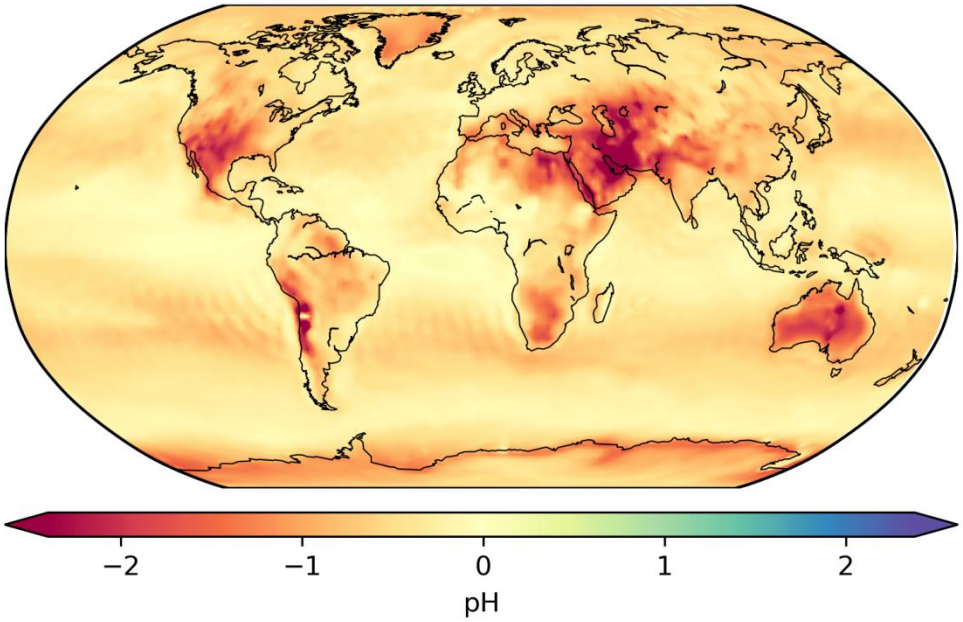

Figure S3: Absolute change in the calculated mean near-surface fine aerosol particle pH during the period 2010-2015 (cf. central panel in Fig. 1) by assuming that aerosols are always aqueous solution droplets (metastable state). A negative change corresponds to more acidic particles compared to the stable state assumption.

Table S1: Fractional emission factors of aerosol components for biofuel combustion, and savannah and tropical forest biomass burning (Akagi et al., 2011), and for sea salt (Seinfeld and Pandis, 2006).

Source	SO_4^{2-}	NO_3^-	Cl^-	Na^+	K^+	Mg^{2+}	Ca^{2+}	NH_4^+
Biofuel combustion	-	0.014	-	-	0.093	0.022	0.073	-
Grassfire burning	0.05	0.04	0.62	0.01	0.62	0.04	0.06	0.01
Forest fire burning	0.25	0.21	0.29	0.01	0.56	0.08	0.16	0.01
Sea salt	0.077	-	0.55	0.306	0.011	0.037	0.012	-

Table S2: Fractional chemical composition of mineral dust emissions (Karydis et al., 2016).

Desert	Na^+	K^+	Ca^{2+}	Mg^{2+}	Other
Great Basin	0.064	0.023	0.053	0.018	0.842
Mojave	0.015	0.027	0.059	0.019	0.880
Sonoran	0.025	0.012	0.037	0.006	0.920
Patagonia	0.012	0.015	0.021	0.013	0.939
Monte	0.023	0.018	0.025	0.009	0.925
Atacama	0.069	0.007	0.018	0.005	0.901
Kalahari/ Namibia	0.030	0.050	0.120	0.090	0.710
Sahara	0.011	0.035	0.075	0.030	0.849
Saudi Arabia	0.010	0.004	0.034	0.006	0.946
Thar/Lut	0.022	0.033	0.082	0.022	0.841
Taklimakan	0.012	0.030	0.120	0.028	0.810
Gobi	0.012	0.021	0.077	0.017	0.873
Great Sandy/ Simpson	0.028	0.001	0.005	0.003	0.963
Other	0.012	0.015	0.024	0.009	0.940

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