



1 **Dust-driven changes in particle pH differentially regulate secondary inorganic**
2 **aerosol formation in central China**

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16



17 **Abstract**

18 Dust episodes facilitate the long-range transport of crustal materials (CM), thereby increasing their
19 contribution to particulate matter and influencing secondary inorganic aerosol (SNA) formation
20 through changes in particle acidity. However, variations in precursor emissions, meteorological
21 conditions, and particle acidity along dust transport pathways may cause the effects of dust input on
22 SNA formation to differ among cities. Such intercity variability during the same regional dust episode
23 remains insufficiently investigated. To address this gap, synchronous observations were conducted in
24 seven cities in central China during a regional dust episode in April 2023. The results showed that,
25 across the seven cities, the mean contribution of CM to PM_{2.5} increased from 22.6% on clean days to
26 53.2% on dust days. Correspondingly, particle pH generally increased to 4.24–4.94 during the dust
27 episode, mainly driven by non-volatile cations such as Ca²⁺ and Mg²⁺. Calculations of aqueous-phase
28 sulfate formation rates indicated that transition metal ion (TMI) catalyzed oxidation consistently
29 dominated sulfate formation. However, this pathway was generally suppressed during the dust episode
30 as particle pH increased. Although the O₃ and NO₂ oxidation pathways were enhanced, their increases
31 could not offset the decrease in the TMI-catalyzed pathway. In contrast, elevated particle pH promoted
32 nitrate partitioning. In cities with relatively low clean-day pH values within the rapid-response range
33 of $\epsilon(\text{NO}_3^-)$, even a non-maximum dust-induced pH increase could result in a pronounced increase in
34 particulate nitrate. Therefore, future nitrate pollution control should consider the coordinated
35 management of NO_x emissions and fugitive dust sources.

36 **Keywords:** PM_{2.5}; Crustal materials; Secondary inorganic aerosol; Particle acidity

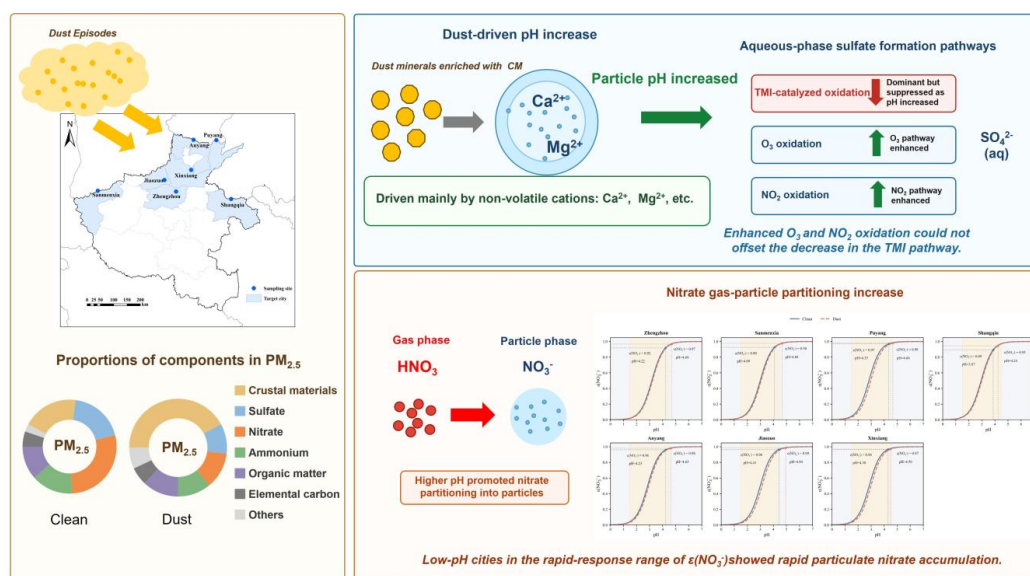


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38 **Synopsis:** Dust-derived crustal materials elevated particle pH, suppressing sulfate formation but

39 promoting particulate nitrate partitioning during a regional dust episode.

40 **Graphical abstract:**



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43 **Highlights:**

- 44 • Dust input made crustal materials the dominant PM_{2.5} component;
- 45 • Non-volatile cations substantially elevated particle pH during the dust episode;
- 46 • Elevated particle pH suppressed TMI-catalyzed sulfate formation but promoted nitrate partitioning
- 47 into the particle phase.

48



49 **1 Introduction**

50 Crustal materials (CM), mainly composed of elements or ions such as Si, Fe, Al, Ca, Mg, and Na,
51 are important components of atmospheric particulate matter (Després et al., 2012). CM are widely
52 present in the atmosphere, with particle sizes ranging from less than 0.1 μm to more than 100 μm ,
53 spanning over three orders of magnitude (Mahowald et al., 2014; Ryder et al., 2019). Globally, the
54 annual emission of CM is second only to sea salt, at approximately 1840 Tg yr⁻¹. The atmospheric
55 burden of CM is about 19.2 Tg, accounting for more than two-thirds of the global aerosol mass (Textor
56 et al., 2006).

57 CM originate from both anthropogenic and natural sources. Anthropogenic CM are mainly derived
58 from fugitive dust generated by human activities, such as road dust and construction dust (Wu et al.,
59 2020; Chen et al., 2023). Natural CM are mainly associated with dust transport and wind-driven
60 resuspension of exposed surfaces or soils (McKendry et al., 2007; Chen et al., 2018). As an important
61 source of CM, dust episodes can transport large amounts of crustal materials and rapidly increase
62 particulate matter concentrations in downwind cities (Chen et al., 2024). The North China Plain is
63 frequently affected by dust episodes under the influence of synoptic systems such as Mongolian
64 cyclones and cold high-pressure systems, making dust transport an important input of crustal materials
65 into atmospheric particulate matter (Chen et al., 2023; Huo et al., 2025). During dust episodes, coarse
66 particle concentrations can increase to 1.39–19.7 times the pre-dust levels, with maximum
67 concentrations reaching 3076 $\mu\text{g}/\text{m}^3$. Meanwhile, CM concentrations in PM_{2.5} may increase to 54.0
68 $\mu\text{g}/\text{m}^3$, contributing up to 71.8% of PM_{2.5} mass (Dong et al., 2020; Wei et al., 2022; Zhang et al., 2025).



69 Secondary inorganic aerosols (SNA), consisting of SO_4^{2-} , NO_3^- , and NH_4^+ , are major components of
70 $\text{PM}_{2.5}$. In eastern China, their contribution can exceed 50% (Wei et al., 2023) and can further increase
71 to 67% during pollution episodes (Wang et al., 2023). SO_4^{2-} is mainly formed through SO_2 oxidation
72 (Tao et al., 2020). In the gas phase, SO_2 can be oxidized by $\bullet\text{OH}$ to form H_2SO_4 , which subsequently
73 partitions into the particle phase (Song et al., 2021; Wang et al., 2022). In the aqueous phase, SO_2
74 dissolves into aerosol water to form S(IV) , which is further oxidized to SO_4^{2-} by H_2O_2 , O_3 , NO_2 , and
75 transition metals such as Fe and Mn (Tao et al., 2020; Wang et al., 2022). During the daytime, nitrate
76 is mainly formed through the reaction of NO_2 with $\bullet\text{OH}$ to produce HNO_3 , followed by partitioning
77 into the particle phase (Liu et al., 2025; Nenes et al., 2020). At night, NO_2 is first oxidized by O_3 to
78 form $\text{NO}_3^\bullet$, which further reacts with NO_2 to produce N_2O_5 (Yan et al., 2019; Ma et al., 2023).
79 Subsequent heterogeneous hydrolysis of N_2O_5 leads to nitrate formation (Nenes et al., 2020; Zhao et
80 al., 2023). Aerosol pH is a key factor regulating SNA formation. For sulfate, pH can alter the aqueous-
81 phase speciation of S(IV) , thereby regulating the relative contributions of different sulfate formation
82 pathways (Pye et al., 2020; Tilgner et al., 2021). For nitrate, its strong semi-volatility makes gas-
83 particle partitioning highly sensitive to aerosol pH (Zhou et al., 2022). Dust-derived CM, including
84 non-volatile cations such as Ca^{2+} , Mg^{2+} , and Na^+ , as well as elements such as Ca, Al, and Fe, can
85 neutralize particle acidity, increase aerosol pH, and further affect aerosol liquid water content, thereby
86 influencing SNA formation (Karydis et al., 2021; Itahashi et al., 2022; Lang et al., 2025). In addition,
87 the large specific surface area of mineral particles provides heterogeneous reaction interfaces for
88 precursors such as SO_2 and NO_2 (Jia et al., 2021; Liu et al., 2022; Li et al., 2025). Transition metals
89 such as Fe and Mn can participate in oxidation reactions and promote sulfate formation (Wang et al.,



2021; Zhang et al., 2024). CM can heterogeneously react with NO_2 and take up gaseous HNO_3 , leading to particulate nitrate formation through surface reactions and acid–base neutralization (Jia et al., 2021; Hrdina et al., 2021; Milousis et al., 2025). Lang et al. (2025) found that the input of abundant non-volatile cations during dust episodes enhanced the buffering capacity of particle acidity and reduced the dependence of nitrate formation on NH_3 , thereby weakening the effect of NH_3 emission reduction on nitrate formation to some extent. These findings suggest that SNA formation mechanisms may change substantially during dust episodes. However, as dust plumes pass through different cities, SO_2 , NO_x , and NH_3 emissions, meteorological conditions, and particle acidity can vary considerably. The effects of dust input on SNA formation may therefore differ among cities, but this issue remains insufficiently investigated.

Central China is frequently affected by dust transported from northern source regions toward the North China Plain and central-eastern China (Tang et al., 2020; Zhang et al., 2024). In spring, enhanced Mongolian cyclones, cold high-pressure systems, and cold frontal activities can transport dust southeastward from the Mongolian Plateau and affect the region (Chen et al., 2023; Borjigin et al., 2024; Huo et al., 2025). Multiple dust transport events have also been observed in this region in spring, autumn, and winter (Dong et al., 2020; Tang et al., 2020; Huo et al., 2025). Meanwhile, SNA are the dominant components of $\text{PM}_{2.5}$ in this region, contributing 53%–63% of $\text{PM}_{2.5}$ mass and increasing to 67% during heavy pollution episodes (Wang et al., 2022, 2023). Previous studies further showed that particle pH in this region is jointly affected by TNH_x and non-volatile cations, indicating that local SNA formation and gas-particle partitioning are sensitive to the aerosol acid-base environment (Wang et al., 2020).



111 In this study, observational data collected synchronously across seven cities in an urban
112 agglomeration in central China agglomeration during April 2023 were analyzed. We compared PM_{2.5}
113 chemical composition between clean and dust days, explored the causes of intercity differences in
114 particle pH under these two conditions, and analyzed the effects of dust episodes on sulfate formation
115 rates and nitrate gas-particle partitioning. This study aims to clarify the impacts of dust input on particle
116 acidity and SNA formation in central China, and to provide a scientific basis for the coordinated control
117 of particulate nitrate and crustal materials.

118 **2 Experiment and methods**

119 **2.1 Observation Sites**

120 Online measurements of particulate matter and its chemical components were conducted in seven
121 cities in Henan Province, China, from 7 to 17 April 2023. The observation sites included Zhengzhou
122 (34.78° N, 113.81° E), Sanmenxia (34.80° N, 111.17° E), Puyang (36.20° N, 115.17° E), Shangqiu
123 (34.57° N, 115.67° E), Anyang (36.21° N, 114.40° E), Jiaozuo (35.10° N, 113.42° E), and Xinxiang
124 (35.38° N, 114.32° E). Descriptions and the spatial distribution of the seven observation sites in Henan
125 Province are shown in Fig. S1 and Table S4.

126 **2.2 Measurements**

127 A Monitor for AeRosols and Gases in ambient Air (MARGA, Metrohm, Switzerland) was used to
128 measure water-soluble inorganic ions (Na⁺, NH₄⁺, K⁺, Mg²⁺, Ca²⁺, Cl⁻, NO₃⁻, and SO₄²⁻) and gaseous
129 species (NH₃, HCl, and HONO) at a 1 h time resolution. The MARGA system was also configured to



130 measure gaseous HNO_3 at all seven sites. However, HNO_3 concentrations were generally low during
131 this campaign, resulting in small chromatographic peaks and relatively large uncertainties in peak
132 integration. Previous MARGA evaluations have shown that low HNO_3 concentrations can increase
133 relative variability and bias, while adsorption onto inlets and sampling tubing can further impair
134 measurement accuracy (Rumsey et al., 2014; Rumsey and Walker, 2016; Chen et al., 2017).
135 Consequently, gaseous HNO_3 measurements from all seven sites could not be quality assured and were
136 excluded from the final dataset and subsequent quantitative analyses. Ambient air was introduced
137 through a $\text{PM}_{2.5}$ inlet, after which particles and gases entered a wet rotating denuder for separation and
138 dissolution. Particles were collected after hygroscopic growth and condensation in a steam-jet aerosol
139 collector and were then analyzed by ion chromatography. Gaseous species were oxidized by H_2O_2 in
140 the denuder, absorbed into the liquid solution, and subsequently quantified by ion chromatography
141 (Rumsey et al., 2014; Trebs et al., 2004). Inorganic elements in $\text{PM}_{2.5}$ were measured using an energy-
142 dispersive X-ray fluorescence analyzer (Xact 625, Cooper Environmental Services, USA) with a time
143 resolution of 1 h. The measured elements included Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe,
144 Co, Ni, Cu, Zn, Ga, As, Se, Sr, Sn, Sb, Ba, and Pb. Ambient particles passed through a $\text{PM}_{2.5}$ inlet and
145 were deposited on a Teflon filter tape. After approximately 1 h of sampling, the filter spot was moved
146 to the analysis position, where elemental concentrations were determined by X-ray fluorescence and
147 converted to mass concentrations using the instrument software. Organic carbon (OC) and elemental
148 carbon (EC) were measured using a semi-continuous carbon analyzer (Model-4, Sunset Laboratory,
149 USA). The measurement was based on the thermal-optical transmittance method following the
150 NIOSH-5040 protocol. Carbon detected under a pure He atmosphere was defined as OC, while carbon



151 oxidized under a mixed atmosphere of 90% He and 10% O₂ was defined as EC.

152 PM_{2.5}, PM₁₀, and gaseous pollutants, including O₃, SO₂, and NO₂, were obtained from online
153 analyzers (Thermo Electron, USA). Meteorological parameters, including wind speed (WS),
154 temperature (T), relative humidity (RH), and pressure (P), were recorded simultaneously using an
155 automatic weather station (LUFFT-WS500, Sutron Corporation, Germany). The accuracies were 0.1
156 m/s for WS, ±0.2 °C for T, ±2% for RH, and ±0.5 hPa for P. Detailed operating principles and quality
157 assurance procedures are provided in Supplementary Text S1.

158 **2.3 Data Analysis**

159 **2.3.1 Mass reconstruction**

160 The calculation method for CM is as follows (Tian et al., 2016):

$$161 \quad [CM] = 1.89 \times [Al] + 2.14 \times [Si] + 1.4 \times [Ca] + 1.43 \times [Fe] + 1.94 [Ti] \quad (1)$$

162 where [Al], [Si], [Ca], [Fe], and [Ti] represent the concentrations of the respective elements (μg/m³).

163 As Ti was not available in the present dataset, the CM concentration was calculated using the measured
164 concentrations of Al, Si, Ca, and Fe only.

165 **2.3.2 Thermodynamic model**

166 The particle pH was calculated using the ISORROPIA-II model (version 2.1,
167 <http://isorropia.eas.gatech.edu>). The input data (excluding RH ≤ 30%), including SO₄²⁻, TNO₃, TNH_x
168 (NH₃+ NH₄⁺), Ca²⁺, K⁺, Na⁺, Mg²⁺, Cl⁻, RH and T. For the TNO₃ input, the measured particulate NO₃
169 concentration was used directly at all seven sites, and no gaseous HNO₃ concentration was included.



As described in Sect. 2.2, the MARGA system was configured to measure gaseous HNO_3 , but quality-assured HNO_3 data were unavailable because the ambient concentrations were generally low and the corresponding chromatographic peaks were subject to relatively large integration uncertainty. The concentrations of hydrogen ions in air (H_{air}^+) and ALWC were derived from the Na^+ - K^+ - Ca^{2+} - Mg^{2+} - NH_4^+ - SO_4^{2-} - NO_3^- - Cl^- - H_2O equilibrium composition system. Activity coefficients for H^+ and OH^- were fixed at unity, while other ion pairs (e.g., H^+ - Cl^-) employed the Kusik-Meissner parameterization for ionic activity calculations (Fountoukis and Nenes, 2007). pH values were calculated using the following formula:

$$\text{pH} = -\log_{10} \text{H}_{\text{aq}}^+ \cong -\log_{10} \frac{1000 \text{H}_{\text{air}}^+}{\text{ALWC}_i + \text{ALWC}_o} \cong -\log_{10} \frac{1000 \text{H}_{\text{air}}^+}{\text{ALWC}_i} \quad (2)$$

$$\text{ALWC}_o = \frac{m_{\text{org}} \rho_w}{\rho_w} \left(\frac{\kappa_{\text{org}}}{\left(\frac{1}{\text{RH}} - 1 \right)} \right) \quad (3)$$

where ALWC_i and ALWC_o refer to the ALWC for inorganic and organic components, respectively. m_{org} denotes the mass of organic aerosol, ρ_w is the density of water (1.0 g/cm^3), ρ_{org} is the density of organic material (1.4 g/cm^3) (Guo et al., 2015); κ_{org} is the hygroscopicity parameter for organic aerosol (0.087) (Chang et al., 2010; Li et al., 2016). The ISORROPIA-II model operated under metastable conditions in the forward mode.

2.3.3 HYSPLIT analysis

Backward trajectories were calculated using the mixed-particle Lagrangian integrated trajectory method (HYSPLIT, https://www.ready.noaa.gov/HYSPLIT_traj.php). Meteorological input data were from the Global Data Assimilation System (GDAS) with 3D wind vectors, temperature, relative



189 humidity, geopotential height, surface pressure, and boundary layer diagnostics. 24-hour backward
190 trajectories were simulated for air masses arriving at 500 m above ground level in each city, and
191 trajectories from 11 to 13 April 2023 were further clustered to characterize the air-mass transport
192 pathways during the dust pollution episode.

193 **2.3.4 Calculation of aqueous-phase sulfate formation rates**

194 Aqueous-phase SO₂ oxidation includes several pathways, such as oxidation by H₂O₂, O₃, and
195 NO₂, transition metal ion (TMI)-catalyzed oxidation, methyl hydroperoxide oxidation, and peracetic
196 acid oxidation (Zheng et al., 2015; Wang et al., 2021). Among TMIs, Ti(III), V(III), Cr(III), Co(II),
197 Ni(II), Cu(II), and Zn(II) generally show relatively low catalytic activity. In addition, atmospheric
198 concentrations of methyl hydroperoxide and peracetic acid are usually low. Therefore, only oxidation
199 catalyzed by Fe(III) and Mn(II) was considered in this study. Because H₂O₂ measurements were not
200 available, the H₂O₂ oxidation pathway was not calculated. The calculation methods for different
201 oxidation pathways are summarized in Table S1, and the detailed equations and procedures are
202 provided in Supplementary Text S2.

203 **2.3.5 Calculation of nitrate gas-particle partitioning**

204 Under conditions favorable for particulate nitrate formation, ambient HNO₃ concentrations are
205 typically substantially lower than particulate NO₃⁻ concentrations (Ding et al., 2019). Previous
206 thermodynamic studies have shown that when aerosol pH exceeds approximately 2.5–3.0, most
207 inorganic nitrate resides in the particle phase, and $\varepsilon(\text{NO}_3^-)$ is comparatively insensitive to moderate pH
208 variations (Vasilakos et al., 2018; Guo et al., 2018). The modeled particle pH values in this study were



generally above this threshold. Therefore, gaseous HNO_3 was expected to account for only a minor fraction of TNO_3 , and its omission from the TNO_3 input was expected to have a limited influence on the estimated $\varepsilon(\text{NO}_3^-)$. According to the method proposed by Guo et al. (2016), the estimated nitrate partitioning ratio was calculated as follows:

$$\varepsilon(\text{NO}_3^-) = \frac{H_{\text{HNO}_3} \times K_{\text{n1}} \times \text{ALWC} \times RT(0.987 \times 10^{-14})}{\gamma_{\text{NO}_3^-} \cdot \gamma_{\text{H}^+} \times 10^{-\text{pH}} + H_{\text{HNO}_3} \times K_{\text{n1}} \times \text{ALWC} \times RT(0.987 \times 10^{-14})} \quad (4)$$

where H_{HNO_3} is the Henry's law constant of HNO_3 , with units of atm^{-1} ; K_{n1} is the dissociation constant of HNO_3 , which depends on temperature, with units of $\text{mol}^2 \cdot \text{kg}^{-2}$; ALWC is the mean aerosol liquid water content derived from the ISORROPIA-II thermodynamic model using the measured water-soluble inorganic ions as input, with units of $\mu\text{g}/\text{m}^3$; R is the gas constant, with units of $\text{J} \cdot \text{mol}^{-1} \cdot \text{K}^{-1}$; T is the mean temperature during the observation period, with units of K; 0.987×10^{-14} is the unit conversion factor; and $\gamma_{\text{NO}_3^-} \cdot \gamma_{\text{H}^+}$ represents the activity coefficient product of NO_3^- and H^+ , which is assumed to be 1.0 under ideal conditions. The product $H_{\text{HNO}_3} \times K_{\text{n1}}$ is the overall equilibrium constant based on the molar concentration of HNO_3 . It was calculated using Eq. (40) reported by Clegg et al. (1998), with the unit conversion from atm^{-1} to $\text{mol}^2 \text{kg}^{-2} \text{atm}^{-1}$ performed according to their Eq. (5).

3 Results and discussion

3.1 Identification and evolution of the regional dust episode

A relatively severe dust episode occurred across central China in April 2023. As shown in Fig. 1, the concentrations of PM_{10} , CM, and Ca^{2+} increased synchronously in Zhengzhou (ZZ), Sanmenxia (SMX), Puyang (PY), Shangqiu (SQ), Anyang (AY), Jiaozuo (JZ), and Xinxiang (XX) during the



228 observation period, with two peaks observed from 11 to 13 April and from 14 to 16 April. The
229 backward trajectories on 13 April (Fig. S2) showed that all seven cities were influenced by air masses
230 transported from the northwest. These air masses originated from Mongolia, Inner Mongolia, and
231 northwestern dust source regions, and were transported to Henan through the North China region,
232 indicating that this episode was closely related to the long-range transport of dust-laden air masses
233 from the northwest. During this period, PM₁₀ concentrations in all seven cities increased to more than
234 twice their pre-dust levels. AY and PY were the most strongly affected by dust transport, with PM₁₀
235 concentrations increasing by factors of 4.7 and 4.1, respectively. The mean CM concentrations on dust
236 days also varied substantially among cities. During the dust episode, ZZ exhibited the highest
237 concentration of CM in PM_{2.5} ($65.8 \pm 34.3 \mu\text{g}/\text{m}^3$), followed by AY ($48.7 \pm 27.3 \mu\text{g}/\text{m}^3$) and JZ (44.8
238 $\pm 26.3 \mu\text{g}/\text{m}^3$), whereas XX had the lowest concentration ($18.3 \pm 10.6 \mu\text{g}/\text{m}^3$). Although ZZ was located
239 downstream of the dust transport pathway, it exhibited the highest CM concentration and a relatively
240 long pollution duration. This may be attributed to the dominance of moderate and low wind speeds
241 during the dust episode, which was unfavorable for dust dispersion and may have also facilitated the
242 resuspension of local fugitive dust. Therefore, although this dust episode showed a clear regional
243 transport signal, the increases in PM₁₀, CM, and Ca²⁺ concentrations, as well as the duration of their
244 peaks, differed among cities. This suggests that atmospheric particles and their chemical components
245 were still affected by local emissions during dust transport. During the second peak, wind speeds were
246 generally lower than those during the main dust period, accompanied by lower relative humidity and
247 higher temperature (Figs. S3 and S4). This peak was likely associated with the combined effects of
248 regional dust retention and local fugitive dust resuspension.



249 **3.2 Changes in PM_{2.5} chemical composition during the dust episode**

250 Figure 2 shows the mass concentrations and fractional contributions of reconstructed PM_{2.5}
251 components in ZZ, SMX, PY, SQ, AY, JZ, and XX. On clean days, PM_{2.5} composition in these cities
252 was strongly influenced by secondary formation, with secondary inorganic aerosols (SNA, including
253 SO₄²⁻, NO₃, and NH₄⁺) representing the major component in most cities. Except for ZZ (39.1%) and
254 SMX (36.4%), the SNA contribution exceeded 40% in the other five cities, and reached 57.4%, 50.5%,
255 and 53.9% in XX, AY, and PY, respectively. This pattern is generally consistent with previous studies
256 (Dong et al., 2020; Wang et al., 2022). Notably, CM already accounted for a considerable fraction of
257 PM_{2.5} on clean days. Its contribution exceeded 20% in all cities and was higher than 30% in ZZ and
258 SMX, indicating that crustal materials remained an important component of regional PM_{2.5} even
259 outside dust periods.

260 During the dust episode, both the concentration and fractional contribution of CM increased
261 rapidly, making CM the dominant component of PM_{2.5} in all cities. The largest increases in CM
262 concentration were observed in PY and AY, where the mean concentrations increased from 3.9 ± 1.6
263 and 8.8 ± 3.2 µg/m³ on clean days to 25.8 ± 15.3 and 48.7 ± 27.3 µg/m³ on dust days, respectively,
264 corresponding to increases by factors of 5.6 and 4.5. The average CM contribution across the seven
265 cities increased from 22.6% on clean days to 53.2% on dust days, corresponding to a 2.4-fold increase.
266 ZZ showed the highest CM contribution on dust days, reaching 68.1%, about twice that on clean days.
267 The CM contributions in AY and JZ increased to 62.2% and 58.2%, respectively, both corresponding
268 to approximately 2.7-fold increases. Although the CM contributions in PY and XX were lower than
269 those in ZZ, AY, and JZ on dust days, their increases were more pronounced because of their relatively



low clean-day levels, reaching 3.8 and 3.2 times the clean-day values, respectively. Ca^{2+} also showed a clear response to dust input. The most pronounced increases in Ca^{2+} concentration occurred in PY, SQ, and AY, where concentrations increased from 0.7 ± 0.2 , 0.5 ± 0.2 , and $0.8 \pm 0.2 \mu\text{g}/\text{m}^3$ on clean days to 3.3 ± 2.3 , 2.3 ± 2.1 , and $3.5 \pm 1.8 \mu\text{g}/\text{m}^3$ on dust days, respectively, increasing by factors of 3.8, 3.6, and 3.5. The fractional contribution of Ca^{2+} to $\text{PM}_{2.5}$ also increased markedly during the dust episode. The increases were more evident in SQ and PY, where the Ca^{2+} contribution increased from 1.3% and 1.9% to 3.8% and 5.4%, respectively, representing increases of approximately 1.9 and 1.8 times the clean-day levels. The Ca^{2+} contributions in XX and AY increased moderately by factors of approximately 1.5 and 1.3, respectively. In contrast, SMX showed the weakest increase, with only an approximately 0.5-fold increase in Ca^{2+} contribution. These results suggest that the response of the Ca^{2+} contribution to dust input differed among cities.

In addition to CM and Ca^{2+} , the mean concentrations of OM, Na^+ , and Mg^{2+} also increased on dust days. However, their fractional contributions to $\text{PM}_{2.5}$ did not increase accordingly because of the rapid increase in CM. For example, OM concentrations in PY and AY increased from 8.9 and $6.3 \mu\text{g}/\text{m}^3$ on clean days to 11.3 and $8.3 \mu\text{g}/\text{m}^3$ on dust days, whereas their contributions decreased from 25.2% and 16.4% to 18.6% and 10.7%, respectively. EC concentrations changed only slightly, and their contributions decreased in all cities. The largest decreases occurred in AY and SQ, where EC contributions declined from 3.4% and 2.9% on clean days to 1.5% and 1.4% on dust days, respectively. Although the rapid increase in crustal materials reduced the fractional contribution of SNA on dust days, SNA concentrations remained relatively high. For example, SNA concentrations reached $14.2 \mu\text{g}/\text{m}^3$ in PY and $14.1 \mu\text{g}/\text{m}^3$ in SQ. Among the three SNA components, NO_3^- remained the dominant



species on dust days. NO_3^- concentrations reached $6.4 \mu\text{g}/\text{m}^3$ in PY and $6.3 \mu\text{g}/\text{m}^3$ in SQ, both higher than the corresponding SO_4^{2-} and NH_4^+ concentrations. SO_4^{2-} also remained at relatively high levels during the dust episode, especially in SMX, SQ, and JZ, with concentrations of 4.6 , 4.5 , and $4.4 \mu\text{g}/\text{m}^3$, respectively. The variations in SOR and NOR further support this interpretation (Figs. 3 and 4). On dust days, SOR values in SMX, PY, and ZZ were 0.36 , 0.32 , and 0.30 , respectively, while NOR values in AY, ZZ, and XX were 0.56 , 0.50 , and 0.47 , respectively. Although sulfur and nitrogen oxidation ratios decreased compared with clean days, they remained at relatively high levels, indicating that substantial nitrate and sulfate formation still occurred in central China during the dust episode.

3.3 Drivers of particle pH during the dust episode

Figure 5 shows the particle pH values calculated using the ISORROPIA-II model for seven cities in central China, under clean and dust conditions. During the dust episode, particle pH increased markedly in all cities under the influence of long-range dust transport. Except for XX, particle pH increased by more than 0.6 units in each city. JZ showed the largest increase, with particle pH rising from 4.19 to 4.94 , corresponding to an increase of approximately 0.75 units. In contrast, XX was least affected by dust input, with particle pH increasing only from 4.05 to 4.50 , by 0.45 units. This pattern is generally consistent with previous studies (Wang et al., 2020; Zhang et al., 2024). The increase in particle pH can be mainly attributed to the input of alkaline crustal materials during dust transport, which buffered acidic species in particles, thereby reducing particle acidity and increasing particle pH (Ding et al., 2019; Karydis et al., 2021). Notably, similar to clean days, substantial intercity differences in particle pH were still observed during the dust episode. The mean particle pH values on dust days



311 followed the order JZ (4.94) > ZZ (4.68) > PY (4.67) > SMX (4.66) > AY (4.65) > XX (4.50) > SQ
312 (4.24), indicating that particle acidity during the dust episode was still strongly influenced by local
313 emission characteristics.

314 To identify the main factors responsible for intercity differences in particle pH on clean days,
315 sensitivity analyses were performed for key influencing parameters. For each sensitivity test, the range
316 of a given parameter was input into the ISORROPIA-II model, while the other parameters were fixed
317 at their mean values. The corresponding particle pH was then calculated under different values of the
318 selected parameter. The sensitivity results showed that particle pH in all cities increased with
319 increasing concentrations of TNH_x , Ca^{2+} , Mg^{2+} , and other cations (Figs. S5, S6, and S8), but decreased
320 with increasing SO_4^{2-} concentration (Fig. S7). Notably, particle pH also showed an increasing trend
321 with increasing TNO_3 concentration in all cities (Fig. S9). This may be related to the overall low
322 concentrations of chemical components and relatively high levels of crustal cations on clean days.
323 Under such conditions, increased TNO_3 was more likely to combine with alkaline cations to form
324 nitrate salts, so that the H^+ concentration did not increase accordingly (Fig. S10), whereas aerosol
325 liquid water content (ALWC) increased (Fig. S11), leading to an increase in particle pH.

326 On clean days, when particles were mainly influenced by local conditions, JZ showed the highest
327 particle pH, with a value of 4.19. This was likely associated with its relatively high TNH_x concentration,
328 which averaged $32.7 \mu\text{g}/\text{m}^3$ and was much higher than those in the other cities. In addition, Ca^{2+} and
329 Mg^{2+} concentrations in JZ were also relatively high, reaching 1.0 and $0.2 \mu\text{g}/\text{m}^3$, respectively. Although
330 JZ had the highest SO_4^{2-} concentration among the seven cities, at $5.8 \mu\text{g}/\text{m}^3$, its high TNH_x and Ca^{2+}
331 concentrations substantially neutralized acidic species, resulting in the lowest particle acidity among



the cities. In contrast, SQ had the lowest particle pH, with a value of only 3.58. This was mainly due to its relatively low TNH_x and Ca^{2+} concentrations, which were 21.1 and 0.5 $\mu\text{g}/\text{m}^3$, respectively, together with a high SO_4^{2-} concentration of 5.8 $\mu\text{g}/\text{m}^3$. These conditions led to insufficient buffering of particle acidity and made SQ the most acidic city among the seven sites. It is worth noting that SMX had the lowest TNH_x concentration among the seven cities, at only 17.5 $\mu\text{g}/\text{m}^3$, but its particle pH was 4.03, much higher than that in SQ. This was likely because SMX had the highest levels of crustal components among the seven cities, with Ca^{2+} and Mg^{2+} concentrations of 1.6 and 0.3 $\mu\text{g}/\text{m}^3$, respectively, which greatly enhanced the buffering capacity of particles. Meanwhile, its SO_4^{2-} concentration was the lowest among the seven cities, at only 4.8 $\mu\text{g}/\text{m}^3$, indicating relatively weak input of acidic components.

To quantify the contribution of individual factors to particle pH changes during the dust episode, a single-factor substitution method was applied. Specifically, one parameter under clean-day conditions was replaced by its corresponding dust-day value, while all other parameters were kept unchanged. The modified inputs were then used in the ISORROPIA-II model to calculate the resulting pH, and the contribution of each factor to ΔpH was obtained. As shown in Fig. 6, the increase in Ca^{2+} concentration was the key factor driving the rise in particle pH on dust days. Ca^{2+} can combine with SO_4^{2-} to form poorly soluble CaSO_4 , which may precipitate from aerosol water, thereby reducing H^+ concentration and particle acidity (Ding et al., 2019; Karydis et al., 2021). In addition, increases in non-volatile cations such as Mg^{2+} and Na^+ during the dust episode further promoted the increase in particle pH, consistent with previous studies (Zhou et al., 2022; Lang et al., 2025). Besides non-volatile cations, the decrease in SO_4^{2-} concentration on dust days also reduced H^+ concentration and ALWC in



353 aerosol water, contributing to the increase in pH (Ding et al., 2019; Zhang et al., 2021). In contrast,
354 increased relative humidity did not cause large changes in particle pH, whereas increased temperature
355 led to a marked decrease in particle pH.

356 The contribution of Ca^{2+} to particle pH varied among cities. SMX and SQ were the most strongly
357 affected by Ca^{2+} during the dust episode, with Ca^{2+} increasing particle pH by approximately 1.01 and
358 0.91 units, respectively. However, their particle pH values remained relatively low because of the
359 suppressing effects of TNO_3 and temperature. Despite the marked pH increase associated with Ca^{2+}
360 input, SQ still exhibited the lowest particle pH among the seven cities during the dust episode, owing
361 to its relatively high initial acidity and limited buffering capacity under clean conditions. ZZ and PY
362 were also markedly affected by Ca^{2+} , with particle pH increasing by 0.81 and 0.69 units, respectively.
363 In addition, the decrease in SO_4^{2-} concentration was larger in ZZ and PY than in SMX and SQ, resulting
364 in higher dust-day particle pH in ZZ and PY. Notably, in JZ, the effects of Ca^{2+} and SO_4^{2-} on dust-day
365 pH were both weaker than those in ZZ, and the suppressing effect of TNO_3 was stronger. Nevertheless,
366 JZ maintained the highest particle pH on dust days because it had the lowest particle acidity and the
367 strongest buffering capacity on clean days. XX showed the smallest increase in particle pH, possibly
368 because of the relatively weak contribution of alkaline cation input. Overall, the increase in particle
369 pH during this dust episode was mainly driven by alkaline cations such as Ca^{2+} , while intercity
370 differences in pH resulted from the combined effects of long-range dust transport and differences in
371 local inorganic chemical composition.



3.4 The role of particle pH in secondary aerosol formation

Previous studies have shown that aqueous-phase reactions are a major pathway for sulfate formation in the North China region (Tao et al., 2020; Cai et al., 2024). In this study, sulfate formation rates through pH-dependent aqueous-phase pathways, including O_3 oxidation, TMI-catalyzed oxidation, and NO_2 oxidation of S(IV), were calculated for clean and dust days. The mean particle pH values for each city under clean and dust conditions are also marked in the figure. The results showed that TMI-catalyzed oxidation was consistently the dominant sulfate formation pathway under both clean and dust conditions. Its formation rates ranged from 2.5×10^{-1} to $2.0 \times 10^0 \mu\text{g m}^{-3} \text{h}^{-1}$ on clean days and from 1.3×10^{-2} to $5.0 \times 10^{-1} \mu\text{g m}^{-3} \text{h}^{-1}$ on dust days, much higher than those of the O_3 and NO_2 oxidation pathways. Compared with clean days, the TMI-catalyzed sulfate formation rates decreased markedly in all cities as particle pH increased during the dust episode, with decreases of approximately 2.2×10^{-2} – $1.5 \times 10^0 \mu\text{g m}^{-3} \text{h}^{-1}$, corresponding to reductions of 75.2%–97.1%. The decreases were particularly large in JZ, ZZ, and SMX, reaching 97.1%, 93.8%, and 93.3%, respectively. This may be attributed to the decrease in Fe(III) and Mn(II) concentrations at higher pH, which reduced the TMI-catalyzed reaction rate.

In contrast, the reaction rates of the O_3 and NO_2 oxidation pathways were higher on dust days than on clean days in all cities. The O_3 oxidation rate increased from 2.5×10^{-7} – $2.9 \times 10^{-6} \mu\text{g m}^{-3} \text{h}^{-1}$ on clean days to 3.2×10^{-6} – $6.7 \times 10^{-5} \mu\text{g m}^{-3} \text{h}^{-1}$ on dust days. The NO_2 oxidation rate increased from 6.2×10^{-7} – $6.3 \times 10^{-6} \mu\text{g m}^{-3} \text{h}^{-1}$ to 2.1×10^{-6} – $2.5 \times 10^{-5} \mu\text{g m}^{-3} \text{h}^{-1}$. These results indicate that the increase in particle pH during the dust episode favored sulfate formation through O_3 and NO_2 oxidation. However, because the TMI-catalyzed oxidation rate was much higher than those of the O_3 and NO_2



393 pathways, the increases in the O_3 and NO_2 oxidation rates were insufficient to offset the decrease in
394 TMI-catalyzed oxidation. As a result, the overall aqueous-phase sulfate formation rate decreased in all
395 cities. This result is generally consistent with the lower SOR values observed on dust days. This
396 indicates that although dust transport increased particle pH, the higher-pH conditions suppressed TMI-
397 catalyzed oxidation and reduced the efficiency of aqueous-phase sulfate formation.

398 The $\epsilon(NO_3^-)$ values in the seven cities of central China were already high on clean days, ranging
399 from 0.81 to 0.95, with the highest value in PY and the lowest in SQ. This indicates that nitrate gas-
400 particle partitioning was dominated by the particle phase even during non-dust periods. During the
401 dust episode, particle pH increased markedly in all cities, accompanied by a decrease in particle acidity,
402 and $\epsilon(NO_3^-)$ increased with increasing particle pH. The most pronounced increase was observed in SQ,
403 where $\epsilon(NO_3^-)$ increased from 0.81 to 0.95. ZZ and SMX also showed clear increases, from 0.88 to
404 0.97 and from 0.92 to 0.98, respectively. In contrast, the increases in PY, JZ, XX, and AY were
405 relatively small. Notably, the increase in $\epsilon(NO_3^-)$ during the dust episode was not fully consistent with
406 the increase in particle pH among cities. Although JZ showed the largest increase in particle pH, its
407 $\epsilon(NO_3^-)$ increased only from 0.94 to 0.99, with an increase of approximately 0.05. By contrast, SQ
408 exhibited a more pronounced increases in $\epsilon(NO_3^-)$, with an increase of 0.14. This difference was mainly
409 related to the response regime of $\epsilon(NO_3^-)$ corresponding to the clean-day pH. When the clean-day pH
410 was lower and located within the rapid-increase range of $\epsilon(NO_3^-)$, the dust-induced increase in pH more
411 effectively promoted nitrate partitioning into the particle phase, thereby enhancing the formation and
412 accumulation of particulate nitrate.



413 **4 Conclusions**

414 The regional dust episode markedly altered PM_{2.5} composition across the seven cities in central
415 China. The mean contribution of CM to PM_{2.5} increased to 53.2% on dust days. This contribution
416 exceeded the 45.6% reported for Beijing in April 2023 but remained below the 69% observed during
417 dust storms in the Horqin source region. Although the relative contribution of secondary inorganic
418 aerosols decreased, substantial nitrate and sulfate formation persisted during the episode. Beyond these
419 compositional changes, ISORROPIA-II simulations showed that particle pH increased in all cities,
420 primarily because of enhanced inputs of Ca²⁺, Mg²⁺, and other non-volatile cations. During the dust
421 episode, particle pH across the seven cities ranged from approximately 4.24 to 4.94. This range is
422 broadly consistent with previously calculated values for Henan (4.5–5.2) and is also close to the values
423 simulated over the southern North China Plain (4.4–5.7). Moreover, substantial intercity differences
424 in particle pH remained and were primarily governed by local inorganic composition and acid–base
425 buffering capacity. Transition-metal-ion-catalyzed oxidation remained the dominant aqueous-phase
426 sulfate formation pathway, but its rate decreased by 75.2%–97.1% as particle pH increased. Although
427 higher pH enhanced the O₃ and NO₂ oxidation pathways, these increases were insufficient to offset the
428 decrease in the dominant TMI-catalyzed pathway. In contrast, the increase in particle pH promoted the
429 partitioning of gaseous HNO₃ into the particle phase. In particular, for cities with relatively low clean-
430 day pH located within the rapid-increase range of $\epsilon(\text{NO}_3^-)$, even a pH increase that was not the largest
431 among the cities could lead to rapid particulate nitrate accumulation.

432 These findings should nevertheless be interpreted in light of several uncertainties associated with



the pathway calculations and thermodynamic modeling. In the absence of H_2O_2 observations, the H_2O_2 oxidation pathway was not included in the calculation, which may have resulted in an underestimation of the total aqueous-phase sulfate production rate under moderately acidic conditions. For the TMI-catalyzed oxidation pathway, Fe(III) and Mn(II) concentrations were estimated using fixed solubility products at 298 K and pH-dependent precipitation equilibria. Because ionic-strength effects were not considered, the calculated reaction rates also remain uncertain. In addition, because forward-mode ISORROPIA-II calculations are based on thermodynamic equilibrium and require complete gas- and particle-phase inputs, the modeled particle pH and ALWC are also subject to uncertainty. Future studies should therefore conduct simultaneous measurements of H_2O_2 and key gas-phase species to strengthen the quantitative constraints on sulfate formation pathways and particle acidity.

Currently, nitrate has become a major challenge for regional $\text{PM}_{2.5}$ control. Its formation is regulated not only by precursor emissions such as NO_x , but also by particle acidity. Dust-derived CM not only increase $\text{PM}_{2.5}$ mass as particulate components, but also modify nitrate gas-particle partitioning by increasing particle pH, thereby favoring particulate nitrate accumulation. Therefore, future nitrate control strategies should continue to prioritize NO_x emission reductions while also considering the long-term management of crustal material and fugitive dust sources, particularly in regions frequently affected by dust episodes or characterized by substantial local dust contributions.

Data availability

All data in this work are available by contacting the corresponding authors, Xiaohui Bi and



452 Shenbo Wang (bixh@nankai.edu.cn and shbwang@zzu.edu.cn).

453 **Supporting Information**

454 The Supporting Information contains additional data, figures, and tables, some of which are
455 referenced directly in the manuscript, as well as detailed descriptions of the field measurements and
456 samples.

457 **Author contributions**

458 X.B. designed this study. H.Z. and S.W. analyzed the data and prepared the manuscript with the
459 contributions of all coauthors. Y.Z. and Y.X. collected the data and contributed to the data analysis.
460 Q.D. revised the paper. Y.F. provided funding acquisition and supervision. All authors have read and
461 agreed to the published version of the manuscript.

462 **Competing interests**

463 The authors declare that they have no conflict of interest.

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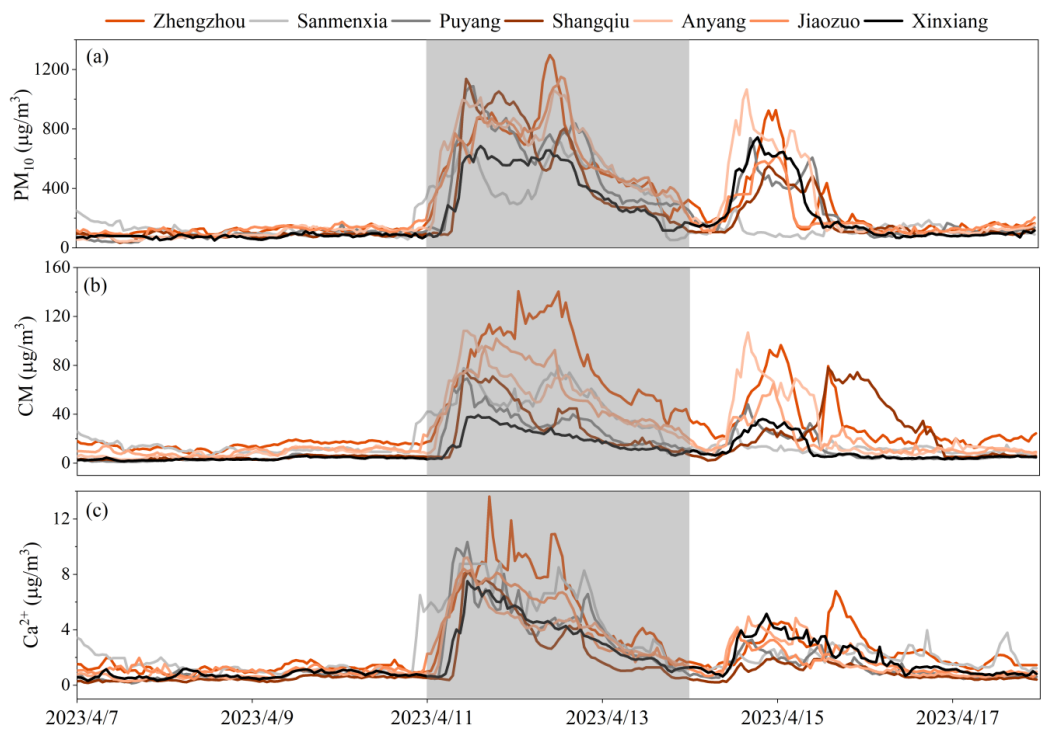
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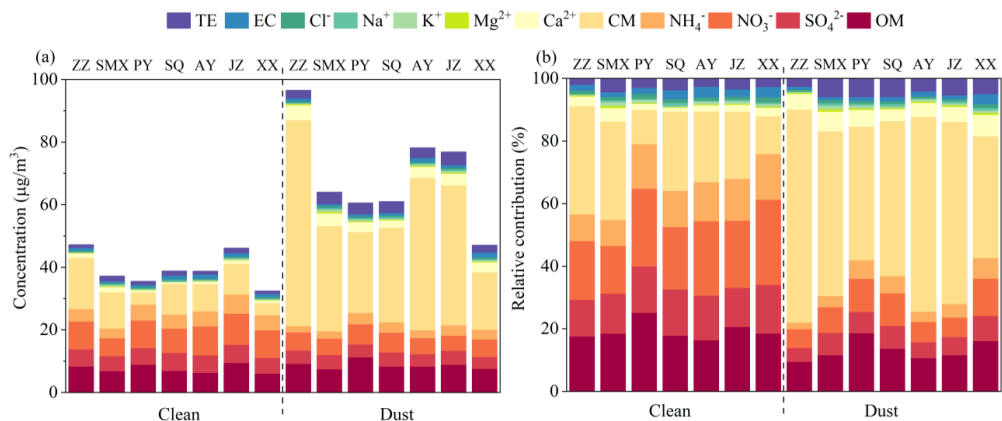
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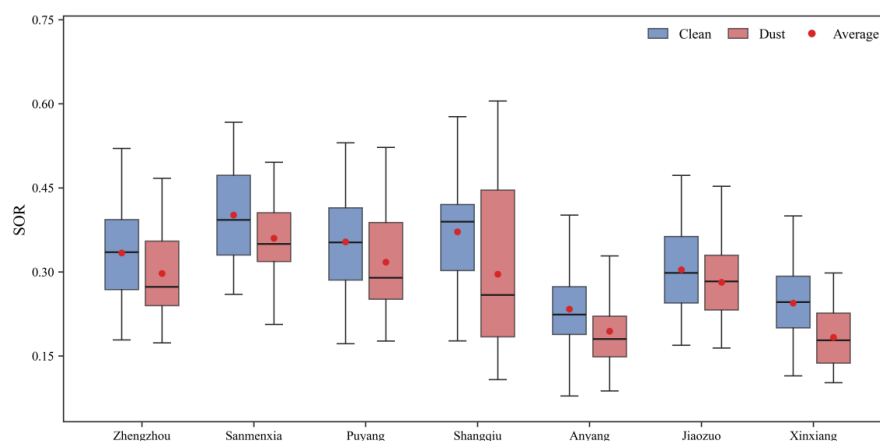
692 **Figures**



693
694 Figure 1. Time series of PM₁₀, crustal material (CM), and Ca²⁺ concentrations across seven cities in
695 central China during the observation period. The shaded area denotes the dust pollution episode.

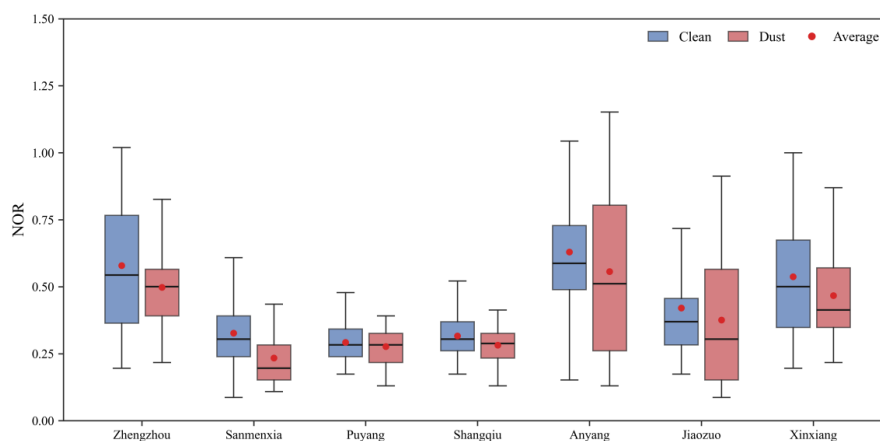


696
697 Figure 2. Chemical composition of PM_{2.5} across seven cities in central China. (a) mass concentrations
698 of individual components; (b) mass fractions of individual components in PM_{2.5}.



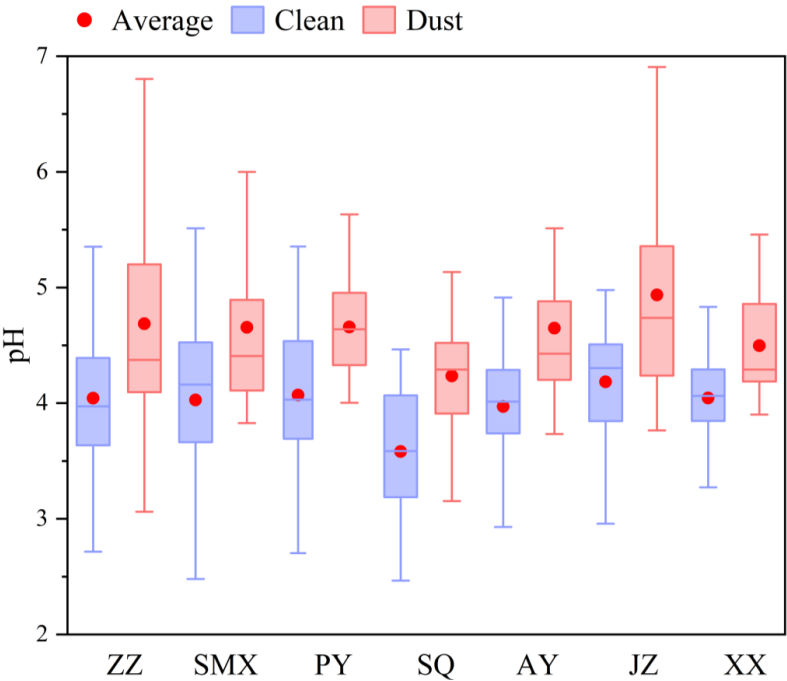
699

700 Figure 3. Box-and-whisker plots of SOR in each city on clean and dust days. Red dots and black
701 horizontal lines denote the mean and median, respectively. The boxes span the 25th–75th percentiles,
702 with the central line indicating the median, while the lower and upper whiskers represent the 10th and
703 90th percentiles, respectively.



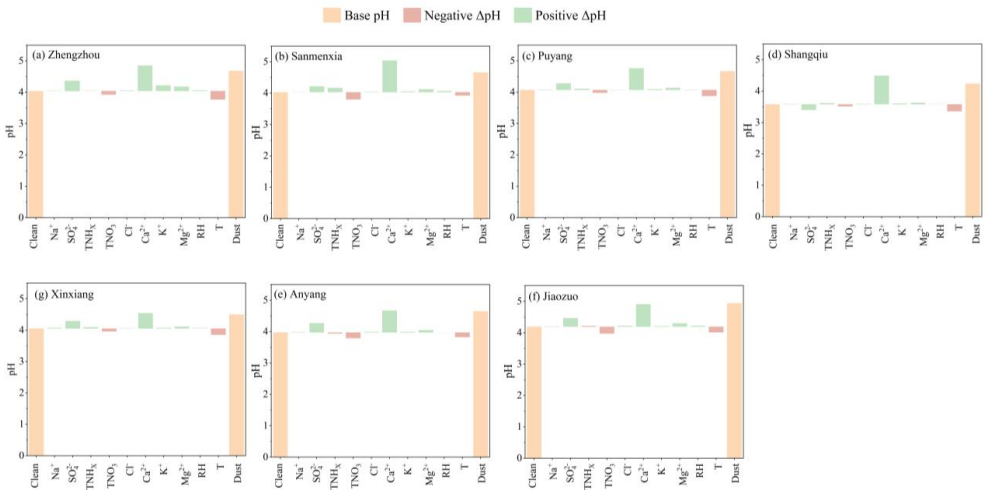
704

705 Figure 4. Box-and-whisker plots of NOR in each city on clean and dust days. Red dots and black
706 horizontal lines denote the mean and median, respectively. The boxes span the 25th–75th percentiles,
707 with the central line indicating the median, while the lower and upper whiskers represent the 10th
708 and 90th percentiles, respectively.



709

710 Figure 5. Box-and-whisker plots of particle pH across seven cities in central China during clean and
711 dust-polluted periods. Red dots denote the mean values, while the blue and red horizontal lines
712 indicate the median values for clean and dust-polluted periods, respectively. The boxes span the
713 25th–75th percentiles, while the lower and upper whiskers represent the 10th and 90th percentiles,
714 respectively.

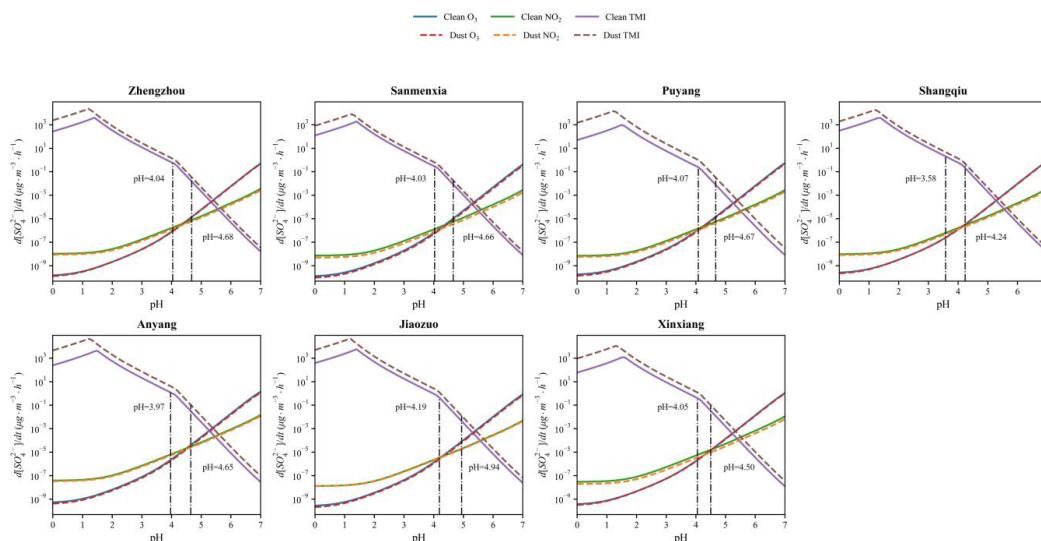


715

716 Figure 6. Factors driving particle pH variations during the dust episode in Zhengzhou (a), Sanmenxia

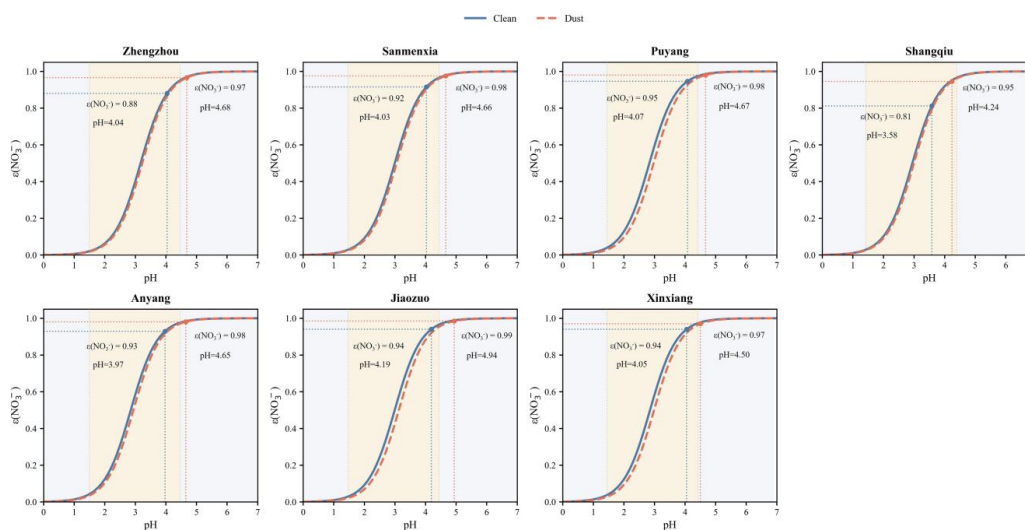


717 (b), Puyang (c), Shangqiu (d), Anyang (e), Jiaozuo (f), and Xinxiang (g). TNH_x represents NH_4^+ and
718 NH_3 , and TNO_3 represents NO_3^- .



719

720 Figure 7. Calculated sulfate production rates through individual aqueous-phase reaction pathways as a
721 function of particle pH across seven cities in central China under clean and dust conditions. Solid and
722 dashed curves represent clean and dust days, respectively. The left and right vertical lines indicate the
723 city-specific mean particle pH values on clean and dust days, respectively.



724

725 Figure 8. Variation in the particle-phase nitrate fraction, $\epsilon(NO_3^-)$, with particle pH across seven cities in



726 central China. Solid and dashed curves represent clean and dust days, respectively. The blue and orange
727 vertical lines indicate the city-specific mean particle pH values on clean and dust days, respectively,
728 while the corresponding horizontal dotted lines indicate the $\epsilon(\text{NO}_3^-)$ values at these pH levels. The
729 yellow-shaded region denotes the pH range over which $\epsilon(\text{NO}_3^-)$ increases rapidly, whereas the blue-
730 shaded regions on either side indicate ranges characterized by slower increases in $\epsilon(\text{NO}_3^-)$.

731



732 **Table**

733 Table 1. Average pollutant concentrations across seven cities in central China during clean and dust
734 periods ($\mu\text{g}/\text{m}^3$).

Cities	Substances	Clean	Dust
Zhengzhou	PM ₁₀	120.6 ± 20.5	510.5 ± 282.8
	PM _{2.5}	52.1 ± 14.3	134.5 ± 77.0
	CM	16.4 ± 4.0	65.8 ± 34.3
	Ca ²⁺	1.4 ± 0.5	4.8 ± 2.9
Sanmenxia	PM ₁₀	119.2 ± 41.9	313.1 ± 209.5
	PM _{2.5}	42.4 ± 10.4	97.6 ± 63.5
	CM	11.7 ± 4.7	33.7 ± 22.3
	Ca ²⁺	1.6 ± 0.9	4.0 ± 2.5
Puyang	PM ₁₀	92.7 ± 26.5	470.0 ± 259.3
	PM _{2.5}	28.7 ± 8.0	81.8 ± 34.2
	CM	3.9 ± 1.6	25.8 ± 15.3
	Ca ²⁺	0.7 ± 0.2	3.3 ± 2.3
Shangqiu	PM ₁₀	91.5 ± 20.4	414.5 ± 299.8
	PM _{2.5}	40.6 ± 14.8	106.5 ± 57.4
	CM	9.8 ± 13.8	30.3 ± 22.6
	Ca ²⁺	0.5 ± 0.2	2.3 ± 2.1
Anyang	PM ₁₀	101.6 ± 28.9	576.1 ± 288.3
	PM _{2.5}	35.6 ± 11.1	123.1 ± 56.8
	CM	8.8 ± 3.2	48.7 ± 27.3
	Ca ²⁺	0.8 ± 0.2	3.5 ± 1.8
Jiaozuo	PM ₁₀	123.0 ± 26.9	493.0 ± 272.6
	PM _{2.5}	50.2 ± 15.6	125.3 ± 55.1
	CM	9.9 ± 2.7	44.8 ± 26.3
	Ca ²⁺	1.0 ± 0.3	3.7 ± 2.3
Xinxiang	PM ₁₀	81.9 ± 13.2	377.4 ± 206.0
	PM _{2.5}	38.5 ± 10.8	104.3 ± 51.4
	CM	3.9 ± 1.1	18.3 ± 10.6
	Ca ²⁺	0.9 ± 0.4	3.2 ± 1.7

735