

**Measurement report: Observational insights into the impact of dust
transport on atmospheric dicarboxylic acids in ground region and free
troposphere**

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Abstract. This study investigated the vertical distribution of PM_{2.5} and size-segregated aerosols at the foot and top of Mount Hua in Northwest China, focusing on C₂ formation and its $\delta^{13}\text{C}$ characteristics influenced by dust transport. Under non-dust conditions, PM_{2.5} and diacids concentrations at the foot were 4.5 and 2.1 times higher than those at the top, with stronger local anthropogenic signals (C₉, 5.67 times higher) and pronounced ~~diurnal-day-night~~ differences for diacids variation. Higher C₂/C₄ (5.84 vs. 4.74), C₃/C₄ (1.04 vs. 0.56) ratios and $\delta^{13}\text{C}$ values (-21.5‰ vs. -27.6‰) at the top indicated photochemical aging during aerosol transport. C₂ concentration was positively correlated with aerosol liquid water content and its size distribution pattern matched with precursors, confirming aqueous-phase oxidation as the dominant formation pathway. During dust events, PM_{2.5} concentrations at the foot and top reached 457 $\mu\text{g m}^{-3}$ and 165 $\mu\text{g m}^{-3}$, but C₂ concentrations in PM_{2.5} decreased by 59% (foot) and 25% (top), while $\delta^{13}\text{C}$ values of C₂ exhibited a ~~significant~~ positive shift (foot: -27.6‰ to -23.9‰; top: -21.5‰ to -13.2‰), attributed to alkaline dust catalyzing ¹³C-enriched oxalate formation. mGly became the second most abundant acid due to enrichment on dust-particle surfaces. Size-segregated data revealed decreased fine-particle C₂ but increased coarse-particle C₂, elevating the coarse-to-fine particle ratio from 0.3~0.4 to 0.6~1.1. These findings offer valuable insights into the altitude-dependent transformation of SOA affected by dust transport, enhancing our understanding of mountain atmospheric chemistry and regional air quality.

Keywords: Dicarboxylic acids, dust particles, size distribution, stable carbon isotopes ($\delta^{13}\text{C}$), aqueous-phase oxidation, heterogeneous reactions

1 Introduction

Dust cycling is crucial to Earth's climate system (Maher et al., 2010; Liang et al., 2022). Mineral particles from dust storms absorb and scatter solar radiation (Kumar et al., 2014), altering regional heat balance and cloud properties, which in turn affects precipitation (Mahowald et al., 2014; Kok et al., 2023; Marx et al., 2024; Xu-Yang et al., 2025). They also act as effective ice-nucleating particles (INPs) in mixed-phase clouds, regulating ice formation and the radiation budget (Fan et al., 2016; Vergara-Temprado et al., 2018; Kawai, et al., 2021; Chen et al., 2024). Dust particles often contain high levels of salts, bacteria, and heavy metals, posing potential risks to human health and plant growth (Yamaguchi et al., 2016; Luo et al., 2024). Dust ~~not only degrades~~impairs air quality locally but also undergoes long-range near its source and can be transported over long distances by winds, ultimately impacting~~affecting both the climate on~~ hemispheric and global climate sealessystems (Pan et al., 2025). During transport, mineral dust may undergo heterogeneous reactions, forming secondary aerosols~~substances~~ that aid cloud formation (Wang et al., 2020; Bikkina et al., 2023). The large surface area of these particles facilitates reactions that alter radiation transfer and photolysis rates (Sullivan et al., 2007a).

Over the past 500 years, East Asia has been frequently hit by dust storms (Zhang et al., 2021; Wu et al., 2022). The Taklimakan and Gobi Deserts, the primary sources of East Asian dust, emit over 800 million tons of dust to downwind areas annually (Sullivan et al., 2007b; Wang et al., 2015; Ren et al., 2019; Zhu and Liu, 2024). Northwestern and northern China, frequently experience dusty weather due to these desert emissions (Gui et al., 2022; Liang et al., 2022). In spring 2023, Mongolia contributed over 42% of the dust concentration in northern China (Chen et al., 2023). Although dust storms are more common in China during spring (Sun et al., 2001), a large-scale, high-intensity dust storm hit northern China on January 10, 2021. This severe dust storm, originating from in southern Mongolia and ~~China's~~ western Inner Mongolia, triggered ~~it was intense and far-reaching, causing~~ rapid air quality deterioration across downwind regions in the affected areas (Figure S1). Our synchronized field observations of PM_{2.5} and size-segregated aerosols

at the top of Mount Hua and on the ground in the winter of 2021 successfully captured this large-scale dust event, as shown in Fig. S1, extensively covering Northern China and the Guanzhong Plain. Liu et al. (2024) compared and analyzed the concentrations and size distributions of water-soluble inorganic ions during dust and non-dust periods, finding Results show that the impact of dust storms have a less pronounced impact on ground aerosols in the Guanzhong Plain is weaker than on that in the free troposphere of the Guanzhong Plain (Liu et al., 2024). Nevertheless, the specific mechanisms through which dust affects organic components, particularly secondary organic aerosol (SOA) and their precursors but the specific effects of dust on organic matter in the ground and troposphere remain unclear and require further study.

To investigate these processes, this study focuses on dicarboxylic acids (diacids), which serve as key tracers for SOA (Xu et al., 2022). As important components of water-soluble organic carbon, diacids are widely distributed in the atmosphere from the surface layer to the free troposphere (Fu et al., 2008; Myriokefalitakis et al., 2011). Conventional theory suggests that aqueous-phase chemical reactions occur predominantly in submicron particles containing water or cloud droplets (Lim et al., 2010; Ervens et al., 2011; Lamkaddam et al., 2021). However, field observations have reported the coexistence of oxalate and nitrate in supermicron particles during dust events (Falkovich and Schkolnik, 2004; Sullivan et al., 2007a; Wang et al., 2015; Xu et al., 2020). To explain this, Wang et al. (2015) proposed that the reaction of nitric acid and/or nitrogen oxides with dust generates $\text{Ca}(\text{NO}_3)_2$, which absorbs water vapor to form an aqueous phase on the dust surface. This enables the partitioning of gas-phase water-soluble organic precursors into this aqueous phase, followed by their further oxidation to form oxalic acid (C_2). Research by Li et al. (2025) provides direct evidence for this mechanism, showing that aqueous nitrate coatings ($\text{Ca}(\text{NO}_3)_2$), due to their very low deliquescence relative humidity (absorbing water at atmospheric $\text{RH} > 8\%$), effectively promote the formation of aqueous secondary organic aerosols (aqSOA). Thus, aged dust surfaces provide critical reactive interfaces for aqSOA formation.

New research shows that nitrate-aged dust plays a crucial role in secondary organic aerosol (SOA) formation through aqueous-phase reactions (Li et al., 2025). As important SOA constituents, dicarboxylic acids (diacids) are ubiquitously distributed from the surface layer to the free troposphere. Due to their low gas-phase reactivity, these compounds are primarily removed via dry deposition, wet deposition, and other heterogeneous processes. Their photochemical reactivity on mineral dust surfaces exhibits distinct carbon chain length dependence, significantly influencing atmospheric organic transformations and aerosol properties (Ponczek et al., 2019). In Asian dust systems, diacids form insoluble complexes with divalent cations (e.g., Ca^{2+}) and undergo photocatalytic reactions with transition metals (e.g., $\text{Fe}^{2+}/\text{Fe}^{3+}$), generating radical oxidants that promote oxalate oxidation and iron reduction (Deguillaume et al. 2005).

Oxalic acid (C_2), a major atmospheric diacid, has a global tropospheric burden of 0.2–0.3 Tg and comprises 5–9% of atmospheric water-soluble organic carbon (Myriokefalitakis et al., 2011). Alkaline mineral particles, like Asian dust, adsorb acidic C_2 readily, while acidic carbonaceous particles are resistant to further adsorption (Yu et al., 2019). The daily variation of C_2 in East Asian mineral dust outflow indicates that its mixture originates from the photochemical transformation of volatile organic compounds (VOCs) (Sullivan et al., 2007b). Mochida et al. (2003) investigated the size distribution of C_2 , malonic acid (C_3) and succinic acid (C_4) in aerosols from the Western Pacific and East Asian coasts. They discussed the adsorption of gas-phase diacids on sea salt particles and the possible formation of solid oxalates, which may enhance the absorption of gaseous C_2 by aerosol particles.

Tropospheric aerosols in high mountain areas are significantly influenced by long-range transport of surface pollutants, making them more representative of regional atmospheric quality. Our previous study (Shen et al., 2023), ~~we investigated the vertical distribution of diacids and the formation mechanisms of C_2 in $\text{PM}_{2.5}$ at different elevations of Mount Hua during the summer. The results demonstrated that~~ summer daytime valley winds on Mount Hua ~~can~~ transport organic acids ~~emitted~~ from the foot to ~~the~~ top, significantly thereby altering the chemical composition of the free troposphere and establishing distinct formation pathways of

C₂ at different altitudes. , while distinct C₂ formation pathways were observed between the top and foot sites. This study examines aerosol vertical distribution characteristics in winter. Low temperatures cause a significant reduction in the boundary layer height over Mount Hua, which inhibits the diffusion of local pollutants to the top. Consequently, the top remains in a free tropospheric environment where aerosols originate primarily from long-range transport from dust source regions (Liu et al., 2024). During the observational period, we documented a major dust event in which particles from dust source regions were directly transported to the top. There, they mixed with local anthropogenic pollutants, triggering complex atmospheric chemical reactions that resulted in notable vertical differences in aerosol chemical properties. However, the influence of heterogeneous reactions on dust aerosol surfaces on the generation of organic acids, particularly their role in modifying C₂ formation mechanisms at different altitudes remains unclear. Therefore, using Here, we further analyzed the observational data from a typical dust event thein winter -of 20201, during which a typical dust event occurred. Tthis study focuses on examining aims to explore the impacts of winter dust transport events on the molecular distribution, particle size characteristics of diacids, and the formation mechanisms of C₂. Our findings will provide new insights The work aims to elucidate the key into the important role of heterogeneous chemical reactions on the surfaces of dust aerosols in the formation of secondary organic aerosolsSOA, providing new insights into regional atmospheric chemical processes during dust events.

2 Experimental and methods

2.1 Sample collection

Samples were collected simultaneously at the free troposphere and the ground surface during December 17, 2020 to January 12, 2021. The sampling site at the ground surface is located on Yinquan Road, Huayin City, Weinan (34°31'N, 110°04'E; ~500 m a.s.l) (referred to as “Foot”), while alpine sampling site is located at the summit of the west peak of Mount Hua (34°28'N, 110°05'E; ~2065 m a.s.l) (referred to as “Top”) (Fig. 1). PM_{2.5} aerosol samples was collected using medium-flow sampler (HC-1010, China Qingdao Company, China) at a flow rate of 100 L min⁻¹ with a duration of 11 hours for each sample during the day (from 08:00 to

19:00) and night (from 20:00 to 07:00 the next day). A total of 54 samples were collected at both the alpine region and ground. The size-segregated samples were collected for ~71 h in each set using an Andersen multi-stage impactor (Andersen, Thermo electronic, USA) at a flow rate of 28.3 L min^{-1} with 9 size bins as < 0.4 , $0.4\text{--}0.7$, $0.7\text{--}1.1$, $1.1\text{--}2.1$, $2.1\text{--}3.3$, $3.3\text{--}4.7$, $4.7\text{--}5.8$, $5.8\text{--}9.0$ and $> 9.0 \text{ }\mu\text{m}$, respectively. In a total of 9 sets of size-segregated samples were collected. All the samples were collected onto pre-combusted (450°C for 6 h) quartz fiber filters produced by Whatman, UK. After sampling, the filters were stored in -18°C until analysis.

2.2 Laboratory Analysis

2.2.1 Determination of carbonaceous species and water-soluble inorganic ions

The concentrations of organic carbon (OC) and elemental carbon (EC) in $\text{PM}_{2.5}$ samples were determined/quantified using a DRI Model 2001 carbon analyzer (Atmoslytic Inc., USA), following the IMPROVE thermal/optical reflectance (TOR) protocol (Cao et al., 2007). A 0.526 cm^2 filter punch was heated stepwise in pure helium (at 120°C , 250°C , 450°C , and 550°C) followed by heating in a 2% oxygen/helium atmosphere (at 550°C , 700°C , 800°C). The method detection limits were $0.41 \text{ }\mu\text{g cm}^{-2}$ for OC and $0.03 \text{ }\mu\text{g cm}^{-2}$ for EC.

Water-soluble components were extracted from a quarter of each filter using 40 mL of ultrapure water (Milli-Q, $18.2 \text{ M}\Omega$, Merck, France) via a combined process of 1-hour ultrasonication and 1-hour mechanical shaking. After filtration through a $0.45 \text{ }\mu\text{m}$ membrane, the extracts were preserved at 4°C for subsequent analysis. Water-soluble inorganic ions (NH_4^+ , K^+ , Mg^{2+} , Ca^{2+} , Cl^- , NO_3^- , and SO_4^{2-}) were analyzed by ion chromatography (Metrohm 940, Switzerland). Anions and cations were separated using an IonPac AS23 column and an IonPac CS12A column, with $9.0 \text{ mM Na}_2\text{CO}_3$ and 20 mM methanesulfonic acid as eluents, respectively (Zhang et al., 2011). Concurrently, water-soluble organic carbon (WSOC) was determined using a total organic carbon (TOC) analyzer (TOC-L CPH, Shimadzu, Japan) (Li et al., 2019). The detection limits for inorganic ions ranged from 0.008 to $0.022 \text{ }\mu\text{g m}^{-3}$, while those for total carbon (TC) and inorganic carbon (IC) were 0.07 mg L^{-1} and 0.08 mg L^{-1} , respectively.

Aerosol liquid water content (ALWC) was calculated using the ISORROPIA-II model (Fountoukis and Nenes, 2007), based on the concentrations of water-soluble inorganic ions (SO_4^{2-} , NH_4^+ , Cl^- , NO_3^- , Ca^{2+} , K^+ , and Mg^{2+}) and meteorological parameters including fields (i.e., relative humidity (RH) and temperature (T).) in the ISORROPIA-II model developed by Fountoukis and Nenes (2007). Meteorological data for the top and the foot were obtained from the Mount Hua Meteorological Station and the Huayin Meteorological Bureau, respectively. All statistical analyses were performed using SPSS.

2.2.2 Determination of dicarboxylic acids and related compounds

The analysis of diacids, keto-carboxylic acids, and α -dicarbonyls in analysis, $\text{PM}_{2.5}$ and size-segregated aerosols samples, were was conducted based on an established derivatization method (Shen et al., 2022). A quarter of each filter was extracted by ultrasonicing ~~one quarter of each filter~~ in ultrapure water (Milli-Q, 18.2 M Ω , Merck, France) for three sequential 15-minute intervals. To ~~maximize~~ optimize the recovery of low-molecular-weight acids (e.g., C_2), the extracts were alkalized ~~were adjusted~~ to pH 8.5–9.0 using with 0.1 M KOH prior to the concentration step before drying. The aqueous extracts were concentrated to near-dryness under vacuum in a water bath maintained at 55 °C (evaporation was halted immediately after the disappearance of the last drop of solvent). The ~~Dried dried~~ residues were derivatized ~~underwent derivatization~~ with 14% BF_3 /n-butanol at 100°C for 1 hour to convert ~~transform~~ carboxyl groups into dibutyl esters and oxo groups into dibutoxyacetals. After the reaction, the derivatives were sequentially dissolved in n-hexane, acetonitrile, and pure water, followed by triple extraction via vortex mixing to remove water-soluble inorganic substances. The aqueous lower layer was removed using a Pasteur pipette. Organic derivatives were purified via triple water washing of the The n-hexane layer was concentrated by rotary evaporation and N_2 blow-down, ~~and~~ reconstituted in 100 μL n-hexane, and finally analyzed for by GC-FID ~~analysis~~ (HP 6890, Agilent Technologies, USA) (Wang et al., 2012). The recovery of the target compounds was 83% for C_2 and ranged from 87% to 110% for other diacids.

Stable carbon isotopic compositions ($\delta^{13}\text{C}$) of ~~oxalic acid~~ C_2 were determined using gas chromatography–isotope ratio mass spectrometry (GC-IRMS; Delta V Advantage, Thermo

Fisher Scientific, Franklin, MA, USA) following established protocols (Kawamura and Watanabe, 2004). To ensure analytical precision (standard deviation < 0.2‰), derivatized samples were analyzed in triplicate. Final $\delta^{13}\text{C}$ values of free oxalic acid were calculated via mass-balance correction to account for isotopic contributions from the $\text{BF}_3/\text{n-butanol}$ derivatizing agent.

3 Results and discussion

3.1 Vertical differences in $\text{PM}_{2.5}$ chemical composition at the foot and top of Mount Hua during non-dust periods

During ~~winter~~ non-dust periods, significant vertical differences were observed in the chemical composition of $\text{PM}_{2.5}$ ~~chemical composition~~ between the foot and top of Mount Hua. The average $\text{PM}_{2.5}$ concentration at the foot ~~was~~ $(127 \pm 48 \mu\text{g m}^{-3})$ was 4.5 times higher than that at the top $(28 \pm 14 \mu\text{g m}^{-3})$, with a mean difference of $99 \mu\text{g m}^{-3}$ (95% CI: 86 to 113; $t(61.05) = 14.60$, $p < 0.001$) (Supplementary Table S1 and Table S3a). approximately 4.5 times higher than the top concentration of $28 \pm 14 \mu\text{g m}^{-3}$ (Supplementary Table S1).

~~This vertical gradient was apparent not only in $\text{PM}_{2.5}$ mass concentrations but also in their chemical composition, as carbonaceous~~ Carbonaceous aerosols ~~components~~ and water-soluble ions were significantly enriched ~~accumulated in significantly higher amounts~~ at the foot (all $p < 0.001$; Table S3a). ~~Diacids and their derivatives, which are~~ As key components ~~constituents~~ of ~~water-soluble organic carbon (WSOC)~~ (Kawamura et al., 2016; Yang et al., 2020), ~~showed particularly strong spatial variation.~~ the total concentrations of diacids at the foot $(1808 \pm 1280 \text{ ng m}^{-3})$ ~~were~~ was approximately 2.1 times higher than those at the top $(860 \pm 534 \text{ ng m}^{-3})$, with a mean difference of 718 ng m^{-3} (95% CI: 453 to 983; $t(69.15) = 5.40$, $p < 0.001$; Table S3b). The spatial difference was most pronounced for azelaic acid (C_9), a biomarker of biomass burning (Kalogridis et al., 2018; Shen et al., 2022) ~~showed the difference~~, with the concentration at the foot $(153 \pm 110 \text{ ng m}^{-3})$ being 5.7 times higher than that ~~and~~ at the top $(27 \pm 18 \text{ ng m}^{-3})$, corresponding to a mean difference of 126 ng m^{-3} (95% CI: 95 to 156; $t(54.77) = 8.24$, $p < 0.001$; Table S3b). ~~, indicating a 5.7 fold higher concentration at the foot.~~

Consequently, ~~the contribution ratios of C₉ to total diacids~~ was substantially greater at the foot (at the two sites were 8.5%) than at the top and (3.2%) (Fig. 2b), respectively.

Correlation analysis of PM_{2.5} and its chemical components revealed no significant relationship between the two sites during winter, contrasting with the positive correlations observed in summer (Shen et al., 2023). Combined with ion composition data and back-trajectory analysis from Liu et al. (2024), these findings indicate that pollutants at the top primarily originated from regional transport ~~in from~~ the northwestern direction with minimal vertical mixing, while ~~the~~ components at the foot of the mountain were mainly controlled by local emission sources. ~~This conclusion was further supported by diurnal variation patterns.~~

Day-night differences provide further evidence supporting this conclusion. At the foot of the mountain, PM_{2.5}, ionic components, and carbonaceous components (except for NO₃⁻ and NH₄⁺) all showed significant day-night differences ($p < 0.05$; Table S4a). Among the diacids, methylglyoxal (mGly) exhibited ~~the most pronounced significant~~ day-night differences, ~~with methylglyoxal (mGly) showing the most pronounced variation~~ (daytime: $128 \pm 49 \text{ ng m}^{-3}$, 47% higher than nighttime: $87 \pm 42 \text{ ng m}^{-3}$, $p < 0.001$; Fig. S2, Table S4b). ~~As the terminal product of diacids photo-oxidation (Kawamura and Sakaguchi, 1999) Oxalic acid (C₂), as the terminal product of diacids (Kawamura and Sakaguchi, 1999), C₂~~ also displayed marked day-night differences ~~diurnal variation~~ (daytime: $766 \pm 552 \text{ ng m}^{-3}$ vs. nighttime: $585 \pm 497 \text{ ng m}^{-3}$, $p = 0.023$), reflecting strong anthropogenic influence on ground-level photochemistry. In contrast, C₂ at the top of the Mount Hua (daytime: $312 \pm 224 \text{ ng m}^{-3}$ vs. nighttime: $299 \pm 186 \text{ ng m}^{-3}$; $p = 0.941$) and its precursors ~~concentrations~~ showed no ~~significant~~ clear day-night differences ~~diurnal fluctuation~~ ($p = 0.341\text{--}0.917$; Table S4c), consistent with the patterns of PM_{2.5} ($p = 0.979$), OC ($p = 0.766$), and other major components (Table S4a). ~~indicating that the high-altitude environment is primarily dominated by regional transport with relatively limited local photochemical contributions. Such stability is a typical characteristic of high-altitude sites located above the planetary boundary layer, primarily governed by regional transport processes (Fu et al., 2008; Li et al., 2012; Meng et al., 2014), indicating that aerosol processes in the free troposphere differ from those at ground level. In this high-altitude environment, in-cloud~~

processes represent a key pathway for aqSOA formation. Studies have shown that C₂ mainly originates from the in-cloud oxidation of precursors such as glyoxal (Gly) and isoprene (Warneck, 2003; Lim et al., 2005; Carlton et al., 2006), a mechanism supported by global model simulations (Myriokefalitakis et al., 2011). Additionally, the photochemical decomposition of C₂ following its association with Fe-containing particles in clouds (Zhang et al., 2019) also contributes to the stable distribution of organic acid concentrations at high altitudes.

Aerosol aging indicators further confirmed distinct oxidation processes between different altitudes. According to existing research, succinic acid (C₄) can be via hydroxylation to generate C₂ and malonic acid (C₃), while C₃ can be further converted to C₂ through intermediates such as hydroxymalonic acid or ketomalonic acid (Kawamura et al. 1993; Kunwar and Kawamura, 2014; Hoque et al., 2017). Therefore, the C₂/C₄ and C₃/C₄ ratios are widely used as effective indicators for assessing the extent of photochemical aging in organic aerosols (Kawamura et al., 2016; Meng et al., 2018; Shen et al., 2022). In this study, ~~the top of Mount Hua,~~ the C₂/C₄ ratio at the top of Mount Hua ~~reached was~~ 5.84 ± 0.32 , ~~while and the~~ C₃/C₄ ratio was 1.04 ± 0.08 , both ~~significantly~~ higher than the corresponding values at the foot (4.74 ± 0.28 and 0.56 ± 0.05 , respectively; Fig. 2c). These results are consistent with reports from high-altitude areas such as Mount Tai (Wang et al., 2009; Meng et al., 2018) and Mount Hua (Meng et al., 2014), collectively confirming that the atmosphere undergoes more significant photochemical aging during long-range transport at high altitudes. ~~—elevated molecular ratios clearly indicate more advanced photochemical aging processes occurring during high-altitude atmospheric transport (Meng et al., 2018).—~~

This study compared the correlations between C₂ and its key precursors at the foot and top of Mount Hua (Fig. 3a–d, 3i–l), revealing differences in atmospheric oxidation pathways across altitudes. Building on this, the synergistic effects of inorganic ions and ALWC on C₂ formation were investigated (Fig. 3e–h, 3m–p), thereby elucidating the aqueous-phase reaction mechanisms and the role of environmental factors such as humidity in C₂ generation. Results showed that ~~were observed between~~ C₂ exhibited the strongest correlation with C₂ and its major

precursors at both sites, with glycolic acid (ωC_2) exhibiting the strongest relationship ($R^2 = 0.88$ at the foot, $R^2 = 0.95$ at the top, $p < 0.01$; Fig. 3 d, l). Other key precursors included Gly ($R^2 = 0.57-0.67$, $p < 0.01$; Fig. 3b, 3j), mGly ($R^2 = 0.58-0.85$, $p < 0.01$; Fig. 3a, 3i), and pyruvic acid (Pyr: $R^2 = 0.80-0.81$, $p < 0.01$; Fig. 3c, 3k). These correlation characteristics confirm that aqueous-phase oxidation serves as the primary formation pathway for C_2 under non-dust conditions (Deshmukh et al., 2017; Du et al., 2022). The consistently higher correlation coefficients at the top further substantiate the enhancing effect of prolonged atmospheric processes on secondary organic aerosol formation.

Contrary to previous studies (Meng et al., 2018; Wang et al., 2012; Meng et al., 2018), we found that C_2 correlated more strongly with NO_3^- ($R^2 = 0.79$, $p < 0.01$; Fig. 3f) than with SO_4^{2-} ($R^2 = 0.71$, $p < 0.01$; Fig. 3e) at the foot, reflecting the promoting effect of local anthropogenic emissions on nitrate formation. The difference mainly stems from strong influences of local anthropogenic emissions (e.g., traffic and industrial activities) at the foot, which provide abundant NO_x and lead to an increased proportion of NO_3^- in $\text{PM}_{2.5}$. Since NO_3^- is more hygroscopic than SO_4^{2-} , its elevated concentration further enhances ALWC, thereby promoting aqSOA formation and intensifying heterogeneous reaction processes (Huang et al., 2025). In contrast, at the top of Mount Hua, C_2 exhibited a higher showed stronger correlation with SO_4^{2-} ($R^2 = 0.63$, $p < 0.01$; Fig. 3m) than with NO_3^- ($R^2 = 0.37$, $p < 0.01$; Fig. 3n). This phenomenon is closely related to the active in-cloud processes in the free troposphere mentioned above. SO_4^{2-} at the top primarily originates from the oxidation of SO_2 within cloud droplets (Yermakov et al., 2023), a process dominated by heterogeneous reactions (Wang et al., 2025). Meanwhile, cloud droplets also provide key reaction media for the aqueous-phase photooxidation of C_2 precursors such as Gly and ωC_2 (Warneck, 2003).

Aerosol Liquid water content (ALWC) exhibited a humidity-dependent threshold effect on C_2 formationthe oxidation process. Under $\text{RH} < 75\%$ During the sampling period, ALWC was positively correlated with C_2 concentration ($R^2 = 0.36-0.44$, $p < 0.01$; Fig. 3g, 3o), reflecting the promotion of precursor dissolution and oxidation by the expansion of the aqueous phase. However, when relative humidity (RH) exceeded 75%, supersaturation shifted the gas-

particle partitioning equilibrium, causing C_2 concentrations to decrease with increasing ALWC. After excluding high-humidity data, the correlation between ALWC and C_2 significantly strengthened ($R^2 = 0.59$, $p < 0.01$; Fig. 3h, 3p), confirming that aqueous-phase oxidation is the primary pathway for C_2 formation during non-dust periods. This finding complements the research by Yang et al. (2022) on the synergistic effects of humidity and pH, jointly revealing the complex regulatory mechanisms of C_2 formation under different humidity conditions.

3.2 Impact of dust transport on $PM_{2.5}$ chemical composition

On January 10, 2021, an extensive and intense dust storm, driven by successive cold fronts and sustained high-velocity winds, swept across northern China, triggering a dramatic surge in $PM_{2.5}$ concentrations (Fig. S3). At the foot of Mount Hua, $PM_{2.5}$ concentrations rapidly rose from $95 \mu g m^{-3}$ to $457 \mu g m^{-3}$ within 24 hours, reaching 3.4 times the non-dust average ($127 \pm 48 \mu g m^{-3}$). Concurrently at the top, $PM_{2.5}$ climbed from 46 to $165 \mu g m^{-3}$, representing a 5.9-fold rise compared to typical conditions ($28 \pm 14 \mu g m^{-3}$). The cleaner atmospheric environment at the top amplified the relative impact of dust transport, while at the foot, existing local pollution partially masked the dust contribution. This contrast highlights the altitude-dependent response to dust events, with the top showing greater sensitivity due to its lower background pollution levels.

HYSPLIT trajectory analysis revealed the dust originated from the Inner Mongolia-Gansu arid region and was transported along a northwest path to the study area (Fig. S4). Dust transport was closely linked to atmospheric circulation, especially in the troposphere, where changes in wind speed play a key role in dust dispersion (Yang et al., 2017). During the dust period, wind speeds at the foot increased from 2.0 to $5.3 m s^{-1}$, and high-altitude wind speeds reached $12.2 m s^{-1}$, significantly higher than the average wind speed during non-dust periods ($5.4 \pm 3.0 m s^{-1}$).

Although $PM_{2.5}$ absolute concentrations rose during the dust period, component concentration changes at the two sites differed markedly. At the foot of the Mount Hua, elemental carbon (EC) concentrations remained relatively stable (4.5 ± 2.1 during non-dust vs. $4.8 \pm 1.8 \mu g m^{-3}$ during dust; Table S2). Organic carbon (OC) showed a modest increase from

17 ± 8.0 to 19 ± 4.6 μg m⁻³, but its mass fraction in PM_{2.5} decreased substantially from 13.4% to 4.4%, reflecting the overwhelming contribution of mineral dust. The top exhibited ~~significantly~~ different characteristics, with both EC and OC concentrations doubling from 0.9 to 1.8 μg m⁻³ and 4.7 to 9.4 μg m⁻³ respectively. OC maintained a higher mass fraction of 6.5% compared to 4.4% at the foot. These patterns indicate efficient mixing of dust with anthropogenic carbonaceous aerosols during long-range transport, coupled with more vigorous secondary formation processes in the free troposphere (Wang et al., 2023; Zheng et al., 2024). ~~The elevated OC fraction at higher altitudes likely results from enhanced photochemical reactions and gas-particle conversions under these atmospheric conditions.~~

Concentrations of mineral components like calcium and magnesium ions (Ca²⁺ and Mg²⁺) rose ~~significantly~~ (foot: 1.8 to 7.7 μg m⁻³; top: 0.7 to 3.2 μg m⁻³), confirming their established role as reliable tracers of dust emissions (Li et al., 2016; Liu et al., 2024). SO₄²⁻ concentrations also increased ~~significantly~~ at both sites (foot: 5.8 to 10.0 μg m⁻³; top: 3.8 to 8.7 μg m⁻³). This ~~increase can be attributed to both the release of inherent sulfate species in dust (e.g., CaSO₄) (Wu et al., 2012) was mainly due to and~~ heterogeneous ~~chemical~~ reactions on dust particle surfaces, ~~with where~~ transition metals ~~like such as~~ Fe (III) and Mn (II) ~~act as catalysts to promote catalyze~~ the conversion of ~~atmospheric sulfur dioxide (SO₂)~~ to SO₄²⁻ (Harris et al., 2013; Myriokefalitakis et al., 2022).

Dust transport ~~significantly~~ altered the concentrations and molecular distribution of diacids and their precursors. Although C₂ remained the most concentrated acidic molecule during dust periods, its absolute concentration decreased noticeably. At the foot of Mount Hua, C₂ concentrations dropped from 674 ± 528 ng m⁻³ (non-dust periods) to 276 ± 20 μg m⁻³ (dust periods), ~~a decrease of by~~ 59%. At the top of Mount Hua, C₂ concentrations decrease from 306 ± 204 ng m⁻³ to 229 ± 45 ng m⁻³, a reduction of 25%. Severe ozone (O₃) pollution was present in this dust storm event, and the particulate eruption promoted the generation and dispersion of O₃ pollutants. O₃ concentrations at the foot increased sharply from 15 μg m⁻³ to 62 μg m⁻³ (Fig. S3), much higher than the non-dust average of 26 ± 19 μg m⁻³. The ~~dust's dust's~~ extinction effect likely reduced aerosol optical thickness, enhancing surface

UV radiation. Combined with a local temperature rise ($\Delta T = +5.8^{\circ}\text{C}$), this probably triggered free-radical chain reactions, promoting the heterogeneous oxidation of C_2 on mineral surfaces (Usher, et al., 2003; Lu et al., 2023; ~~Usher, et al., 2003~~). Notably, the proportion of C_2 in total diacids ~~exhibited contrasting~~ ~~showed different~~ trends at the two sites, decreasing from 37.3% to 32.2% at the foot and increasing from 35.5% to 42.8% at the top (Fig. 4c). This divergence could be closely related to the humidity levels at the two sites. During the dust events, the lower relative humidity at the foot ~~experienced low relative humidity~~ ($\text{RH} = 24 \pm 8.5\%$), suppressed inhibiting aqueous liquid-phase oxidation, ~~whereas~~ while the higher humidity at ~~altitude~~ the top ($\text{RH} = 44 \pm 11\%$) favored ~~secondary~~- C_2 formation through such reactions. Additionally, variations in aerosol sources, transport pathways, aging processes, and potential contributions from other chemical reactions may also have influenced C_2 generation.

As a major atmospheric keto acid and key precursor of C_2 (Kawamura et al., 2012; 2013), ωC_2 ranked second among the acids detected at both sites during non-dust periods, with concentrations of $214 \pm 199 \text{ ng m}^{-3}$ at the foot and $77 \pm 52 \text{ ng m}^{-3}$ at the top of Mount Hua. However, during dust periods, mGly became the second most abundant acid at both sites due to its enrichment on dust-particle surfaces. At the foot, mGly concentration reached $116 \pm 34 \text{ ng m}^{-3}$ (13.6% of total diacids), up from 5.9% in non-dust periods. At the top, mGly concentration was $38 \pm 9 \text{ ng m}^{-3}$ (7.1% of total diacids) (Fig. 4b), slightly higher than the non-dust 5.6%. Phthalic acid (Ph), a photo-oxidation product of naphthalene and other aromatic hydrocarbons, primarily originates from industrial processes and incomplete combustion of coal in heavy and diesel vehicles (Ho et al., 2006). At both foot and top of Mount Hua, the proportion of Ph remains relatively stable, accounting for approximately 4% during both dust and non-dust periods, indicating that its sources are stable and closely related to regional industrial activities and traffic emissions. During dust periods, the C_2/C_4 ratios at the foot and top of Mount Hua rose to 5.24 and 7.75 (Fig. 4c), showing stronger aerosol aging. However, the C_3/C_4 ratio at top of Mount Hua dropped to 0.70, this might result from the combined effects of selective adsorption of C_2 onto dust particles and enhanced photolysis of C_3 on mineral dust surfaces.

3.3 Size distribution characteristics of diacids and related compounds during non-dust and dust periods

During non-dust periods, the size distribution of C₂ at both the foot and top of Mount Hua exhibits a distinct bimodal ~~distribution pattern~~ (Fig. 5a and 5(a)), characterized by a ~~primary major~~ peak in the fine particle mode (0.4-1.1 μm) and a ~~secondary minor~~ peak in the coarse particle mode (4.7-5.8 μm). Fine particles, with their greater specific surface area and hygroscopic nature, provide a conducive liquid-phase environment that enhances the oxidation of precursors such as mGly and Pyr, leading to C₂ formation (Ervens et al., 2011; Wang et al., 2015). The high correlation between C₂ and secondary inorganic ions (SO₄²⁻, NO₃⁻, NH₄⁺) (with R² values of 0.92-0.95 at the foot and 0.48-0.92 at the top, p < 0.01; as shown in Fig. 6) supports this mechanism, confirming that C₂ formation during non-dust periods primarily relies on liquid-phase oxidation reactions on fine particle surfaces. The lower R² values at the top may be due to the greater influence of long-range transport at high-altitude sites, resulting in more complex sources of precursors. C₂ in the coarse particle mode likely originates from direct adsorption of biogenic emissions (such as plant waxes) or heterogeneous oxidation of gas-phase precursors on mineral dust surfaces (Wang et al., 2012).

The distribution patterns of short-chain diacids, such as C₃ and C₄ are similar to that of C₂ (Fig. 5b-c and 5(b)-(c)), with ~~primary main~~ peaks at 0.4-1.1 μm and secondary peaks at 4.7-5.8 μm, indicating that these acids mainly come from fine particles. In contrast, glutaric (C₅) acid shows distinct distribution characteristics at the two sites. At the foot, it exhibits a ~~unimodal distribution single peak~~ in the coarse particles (4.7-5.8 μm) (Fig. 5d), while at the top, it displays a bimodal distribution (0.4-1.1 μm and 4.7-5.8 μm) (Fig. 5(d)). This significant difference in modal structure suggests that different atmospheric processes govern the behavior of C₅ at different altitudes. The C₅ at the foot likely mainly comes from local emissions (Hatakeyama et al., 1985), such as the oxidation of cyclopentene in vehicle exhaust, whereas the high-altitude area is influenced by a combination of long-range transport and local processes (Zhao and Fu, 2018). Adipic (C₆) acid shows a bimodal distribution at both sites, probably resulting from the oxidation of cyclohexene (in the fine mode) or adsorption of gas-

phase precursors on coarse particle surfaces (Deshmukh et al., 2016). Azelaic (C₉) acid is ~~significantly~~ enriched only in fine particles at the foot, which may be closely related to the oxidation of unsaturated fatty acids emitted from biomass burning in northern regions during winter (Deshmukh et al., 2016). The coarse particle fraction of Ph likely forms through gas-phase adsorption, consistent with the strong adsorption characteristics of coarse particles reported by Kanellopoulos et al. (2021).

The particle size distributions of mGly (Fig. 5g and 5(g)) and ωC₂ (Fig. 5j and 5(j)) showed remarkable consistency with that of C₂ (Fig. 5a and 5(a)), providing direct evidence that aqueous-phase oxidation serves as the predominant formation pathway for C₂. Pyr exhibited distinct altitudinal variation in its distribution characteristics. At the top of Mount Hua, Pyr displayed a similar size distribution pattern to C₂, indicating their shared photochemical origin. In contrast, at the foot, Pyr demonstrated ~~significant~~ enrichment in coarse particles (4.7-5.8 μm), likely attributable to heterogeneous reactions of gaseous precursors from local coal combustion on mineral dust surfaces. Gly also exhibits a peak in coarse particles, likely due to its strong adsorption and chemical stability on particle surfaces. The diacids concentrations are consistently lower at the top compared to the foot, which is closely tied to substantial local emissions at the lower elevation. As shown in Fig. 6, the coal combustion and biomass burning tracer Cl⁻ demonstrates an exceptionally strong correlation with C₂ ($R^2 = 0.88$, $p < 0.01$) at the foot.

The dust transport process ~~significantly impacted the size distributions~~ ~~altered the particle size distribution characteristics~~ of diacids in aerosols (Fig. 5). At the foot of the mountain, the C₂ concentration in fine particles (≤ 2.1 μm) decreased ~~markedly~~ from 8662 ng m⁻³ to 4880 ng m⁻³ (a reduction of 43.7% ~~reduction~~) during dust events, while the concentration in coarse particle (> 2.1 μm) ~~concentration showed a modest~~ increased from 2718 ng m⁻³ to 2843 ng m⁻³ (an increase of 4.6%) (Table 3). A more pronounced change was observed at the high-altitude top of Mount Hua, where the C₂ concentration in coarse particles (2301 ng m⁻³) exceeded that in fine particles (2161 ng m⁻³) during dust events, indicating a shift in the dominant particle size distribution from fine to coarse modes. This shift in particle size distribution can be

453 attributed to the formation of $\text{Ca}(\text{NO}_3)_2$ coatings resulting from the reaction between calcium
454 carbonate and NO_3^- during dust aging (Li and Shao, 2009; Zhi et al., 2025). These hygroscopic
455 coatings create favorable conditions for the adsorption and oxidation of gaseous organic
456 compounds, thereby promoting the formation of SOA on the surfaces of coarse particles.
457 Research by Li et al. (2025) further confirms that aqSOA formed on dust surfaces can
458 effectively enhance SOA production and drive a transition in the size distribution from the
459 submicron to the supermicron range, which is highly consistent with the observational results
460 of this study. Size-segregated ion data (Fig. S5) provide direct evidence for the above
461 mechanism. Ca^{2+} was primarily present in the coarse mode (3.3–5.8 μm), while during dust
462 events, NO_3^- at the top migrated from the fine mode (0.4–1.1 μm) to the coarse mode (3.3–5.8
463 μm) and coexisted with Ca^{2+} in the same size range, strongly supporting the formation of
464 $\text{Ca}(\text{NO}_3)_2$ coatings on dust particle surfaces. In contrast, at the foot of the mountain, although
465 the concentration of NO_3^- decreased, it remained predominantly in the fine mode. This spatial
466 difference may stem from more thorough aging and reactions of aerosols at the top due to
467 longer transport times. Meanwhile, the foot is influenced by local pollution, resulting in higher
468 background NO_3^- concentrations and competitive reactions with components such as SO_4^{2-} ,
469 which may collectively delay the distinct shift of NO_3^- to the coarse mode. These findings show
470 excellent agreement with the research results of Li et al. (2025), which demonstrated that
471 calcium carbonate reacts with nitric acid during dust aging processes to form calcium nitrate
472 ($\text{Ca}(\text{NO}_3)_2$). The resulting hygroscopic surfaces provide favorable conditions for the adsorption
473 and heterogeneous oxidation of gaseous organic compounds. Furthermore, aqueous secondary
474 organic aerosols (aqSOA) formed on dust surfaces promote SOA formation and drive the
475 transition of particle size distribution from submicron to supermicron ranges.

476 Analysis of the dust/non-dust concentration ratio ($R_{\text{D/N}}$) revealed $R_{\text{D/N}}$ values of 0.3 and
477 0.6 for fine and coarse particles at the foot, respectively, while these values reached 0.4 and 1.1
478 at the top, indicating that dust processes have a more significant impact on the particle size
479 distribution of diacids at high-altitude regions. Further investigations confirmed significant
480 enrichment of C_2 precursors (mGly, Pyr, and ωC_2) in the coarse particle fraction ($>2.1 \mu\text{m}$).

Observational data from the top of Mount Hua revealed that the concentration ratio ($R_{D/N}$) of these precursors in coarse particles during dust periods reaches 0.8-1.4 during dust periods, higher than the 0.5-0.7 ratio in fine particles ($\leq 2.1 \mu\text{m}$), demonstrating the crucial contribution of heterogeneous oxidation on dust particle surfaces to C_2 formation. Throughout dust episodes, the particle size distribution patterns of diacids (C_2 - C_6) consistently displayed a pronounced shift from fine to coarse particles (4.7 - $5.8 \mu\text{m}$) (Fig. 5). Notably, concentrations of the biomass burning tracer C_9 decreased during dust episodes, while Ph and isophthalic acid (iPh) showed distinct peaks in the coarse particle mode. This phenomenon indicates that dust particles effectively scavenge gaseous pollutants through strong adsorption, thereby suppressing the formation of local fine-mode ~~secondary organic aerosols (SOA)~~. However, these gaseous precursors adsorbed onto coarse particle surfaces can still undergo heterogeneous oxidation reactions to form SOA.

During dust events, the correlation between C_2 and mineral ions (Ca^{2+} , Mg^{2+}) showed significant enhancement (with R^2 values of 0.33 - 0.30 ; $p < 0.05$ at the foot and 0.65 - 0.39 ; $p < 0.01$ at the top). This finding shows excellent agreement with the recent research results of Li et al. (2025), who similarly observed stronger correlations between Ca^{2+} and ~~oxalate~~ C_2 ($R^2 = 0.46$ - 0.95) in the coarse particle phase. This arises as organic acids like ~~oxalic acid~~ C_2 in aged carbonate-containing dust particles react with carbonates to form stable salts (Ervens et al., 2008; Lim et al., 2010). This process inhibits the volatility of organic acids and stabilizes them in the coarse particle phase. Furthermore, due to differences in rock types and weathering processes, Asian dust particles inherently contain higher concentrations of alkaline metal elements (Ca and Mg) compared to dust from other regions (Yu et al., 2025). The size-resolved correlations between C_2 , Ca^{2+} , and Mg^{2+} (Fig. 6), further support this conclusion. During dust events, the concentrations of C_2 , Ca^{2+} , and Mg^{2+} exhibited synchronous increases with increasing particle size. In contrast, during non-dust periods, the peak concentration of C_2 occurred in the fine particle size range, while mineral ion concentrations did not show corresponding increases. As described in Section 3.2, dust events led to ~~significant~~ reductions in C_2 concentrations in $\text{PM}_{2.5}$ (foot: $674 \pm 528 \text{ ng m}^{-3}$ during non-dust periods vs. $276 \pm 20 \text{ ng m}^{-3}$ during dust periods).

m⁻³ during dust periods; top: 306 ± 204 ng m⁻³ during non-dust periods vs. 229 ± 45 ng m⁻³ during dust periods).

Overall, the transformation mechanisms of C₂ and its precursors underwent ~~significant~~ alterations during dust events, shifting from aqueous-phase oxidation dominated in fine particles during non-dust periods to heterogeneous oxidation on coarse particle surfaces as the primary pathway during dust episodes. Regional comparative analysis further revealed that atmospheric chemical processes at the mountain foot were mainly influenced by local emission sources such as coal combustion and biomass burning, whereas the summit site more clearly reflected the complex interactions between long-range dust transport and regional atmospheric processes.

3.4 Stable carbon isotopes (δ¹³C) of oxalic acid

Synchronized observations at the foot and top of Mount Hua (2060 m asl) revealed an inverse correlation between C₂ concentration and δ¹³C values in PM_{2.5} (Fig. 7a and 7b). When C₂ concentration at the foot increased to 2424 ng m⁻³, its δ¹³C value decreased to -34.5‰, while at the top, a concentration of 917 ng m⁻³ corresponded to a δ¹³C of -24.7‰. Conversely, during low concentration periods, δ¹³C at the foot rose to -21.9‰ (258 ng m⁻³) and at the top to -17.2‰ (63 ng m⁻³). This systematic variation provides clear evidence for ~~significant~~ kinetic carbon isotope fractionation during atmospheric aqueous-phase oxidation processes. Specifically, volatile organic compounds (VOCs) and semi-volatile organic compounds (SVOCs) with lower δ¹³C values preferentially react to form aqSOA, resulting in ¹³C-depleted products (Xu et al., 2022).

Spatially, the δ¹³C values of C₂ in PM_{2.5} at the top of Mount Hua (-28.4‰ to -12.8‰, mean -21.5‰) were ~~significantly~~ higher than those at the foot (-36.2‰ to -14.9‰, mean -27.6‰). This vertical gradient primarily results from long-range transport of aerosols at high altitudes coupled with deep oxidation processes. The higher C₂/C₄ ratio observed at Mount Hua (5.84 vs. 4.74 at the foot) indicates more pronounced atmospheric aging characteristics. This distribution pattern originates from prolonged photochemical oxidation during long-range transport, where preferential cleavage of ¹²C-¹²C bonds (due to their lower bond energy) leads

to relative ^{13}C enrichment in residual C_2 . In contrast, surface aerosols dominated by local fresh emissions undergo shorter oxidation periods and exhibit weaker isotope fractionation effects. Moreover, $\delta^{13}\text{C}$ can provide insights into the sources of aerosols, Pavuluri and Kawamura (2016) found that biogenic aerosols had higher mean $\delta^{13}\text{C}$ values (-15.8‰) than anthropogenic sources (-19.5‰). Our study shows that foot aerosols were mainly influenced by anthropogenic sources (biomass burning and coal combustion), while the top was more affected by natural sources due to richer vegetation, with long-range transport potentially weakening local isotope fractionation effects.

During dust events, C_2 concentrations in $\text{PM}_{2.5}$ showed ~~significant~~ decreasing trends, declining from $674 \pm 528 \text{ ng m}^{-3}$ to $276 \pm 20 \text{ ng m}^{-3}$ at the foot and from $306 \pm 204 \text{ ng m}^{-3}$ to $229 \pm 45 \text{ ng m}^{-3}$ at the top of Mount Hua. Concurrently, the $\delta^{13}\text{C}$ values of C_2 exhibited distinct positive shifts, increasing from -27.6‰ to -23.9‰ at the foot and from -21.5‰ to -13.2‰ at the top. This phenomenon reveals key chemical transformation mechanisms during dust transport: alkaline mineral surfaces promote heterogeneous catalytic oxidation of ~~oxalate- C_2~~ precursors, with coarse-mode mineral components ($\text{Ca}^{2+}/\text{Mg}^{2+}$ etc.) preferentially combining with ^{13}C -labeled C_2 to form stable compounds like calcium oxalate. This mechanism is strongly supported by observational data during dust events, both oxalic acid and $\text{Ca}^{2+}/\text{Mg}^{2+}$ concentrations showed ~~significant~~ increases with growing particle size, while exhibiting high correlations ($R^2 = 0.30\text{-}0.65$) during dust periods. Simultaneously, ^{12}C -enriched C_2 produced on fine particle surfaces moves to coarse particles through gas-particle conversion or coagulation, resulting in ^{13}C -enriched residues remaining in fine particles. As demonstrated by the aforementioned research findings, the concentration of oxalic acid in coarse particles showed a marked increase during dust events, with this variation being particularly pronounced at the top. These two synergistic processes collectively altered both aerosol size distribution and isotopic composition characteristics.

4. Conclusions

This study provides compelling evidence for the dual role of dust transport in modulating oxalic acid formation and isotopic composition in atmospheric aerosols. Through

comprehensive analysis of PM_{2.5} and size-segregated samples, we demonstrate that dust events trigger a fundamental shift in oxalic acid production pathways from predominant aqueous-phase oxidation in fine particles during non-dust periods (showing strong humidity dependence below 75% RH) to mineral heterogeneous chemistry in coarse particles during dust episodes (Fig. 8). The dust induced transformations are particularly pronounced at high-altitude sites like Mount Hua, where we observed: (1) enhanced aerosol aging indicators (C₂/C₄ ratio increasing to 7.75); (2) ~~significant~~ $\delta^{13}\text{C}$ enrichment (+8.3‰ compared to non-dust periods); and (3) preferential partitioning of ~~C₂oxalate~~ to coarse-mode particles (coarse/fine ~~particle~~ ratio reaching 1.1). These changes result from synergistic effects of surface-catalyzed oxidation favoring ¹³C retention and metal-oxalate complexation. Conversely, surface sites exhibited stronger local anthropogenic influences, with higher ~~C₂oxalate~~ concentrations (674 ± 528 ng m⁻³) but lower $\delta^{13}\text{C}$ values (-27.6‰), reflecting less processed aerosols. The identified inverse ~~oxalateC₂~~ $\delta^{13}\text{C}$ relationship and altitude-dependent response to dust transport provide new quantitative tools for evaluating aerosol aging processes and improving regional air quality models, particularly in dust-prone regions under changing climate conditions.

Data availability

The data in this study are available at <https://zenodo.org/doi/10.5281/zenodo.15788834> (Shen et al., 2025).

Author contributions

Jianjun Li conceived and designed the study. Minxia Shen conducted the literature search, performed sample and data analysis, and wrote the manuscript. Jianjun Li and Qiyuan Wang contributed to manuscript revision. Weining Qi, Yali Liu, Yifan Zhang, Wenting Dai, Lu Li, Xiao Guo, Yue Cao, Yingkun Jiang, Qian Wang and Shicong Li collected particulate samples and supervised the experiments. All authors provided critical feedback on the manuscript and approved the final version.

Conflict of Interest

The authors declare no conflicts of interest relevant to this study.

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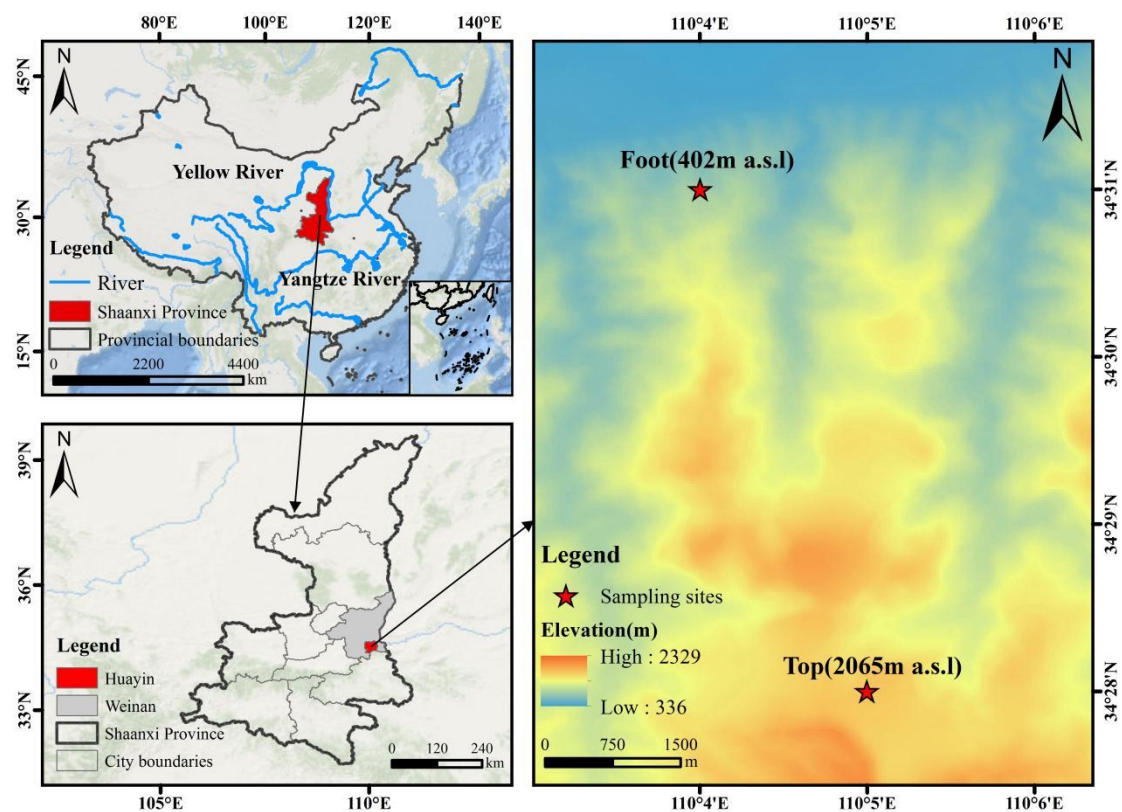


Figure 1 Distribution of aerosol sampling sites at the foot and top of Mount Hua (December 2020 to January 2021)

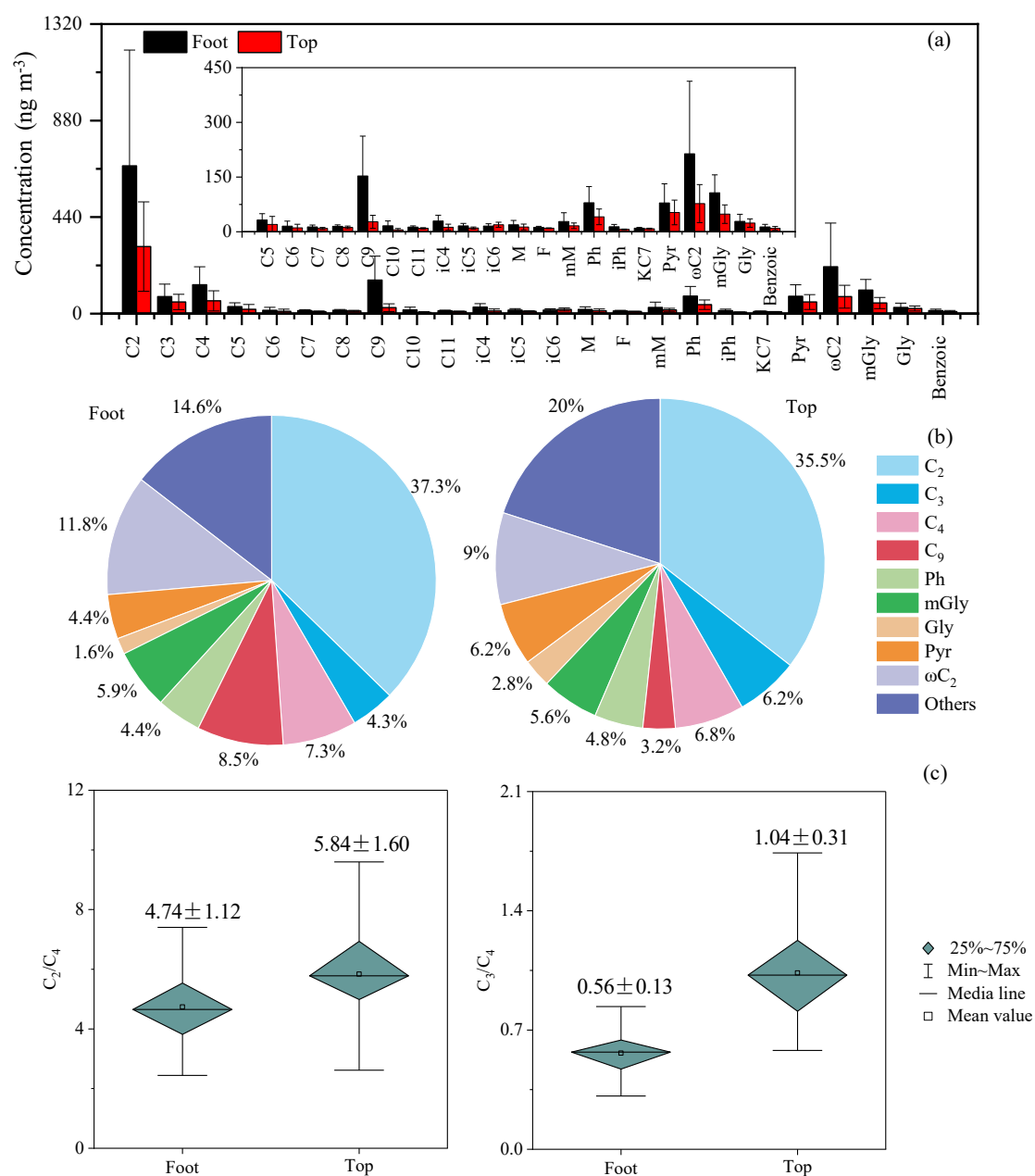


Figure 2 Molecular distribution of dicarboxylic acids and related Compounds (a), relative percentages of major dicarboxylic acids (b), and ratios of C₂/C₄ and C₃/C₄ (c) at the foot and top of Mount Hua during non-dust periods

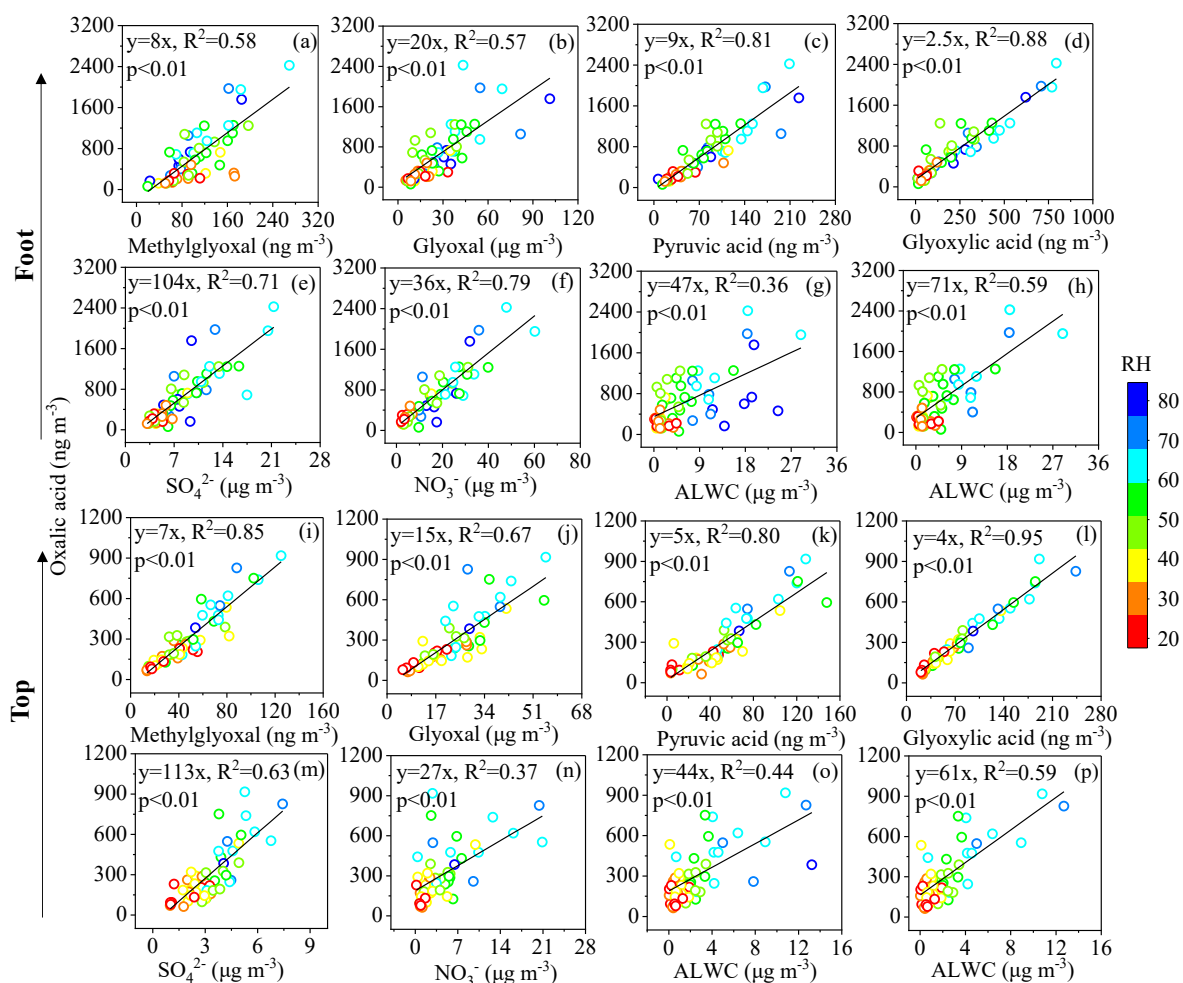


Figure 3 Correlation between C_2 and its key precursors, sulfate (SO_4^{2-}), nitrate (NO_3^-), and aerosol liquid water content (ALWC) at the foot and top of Mount Hua with varying relative humidity (RH)

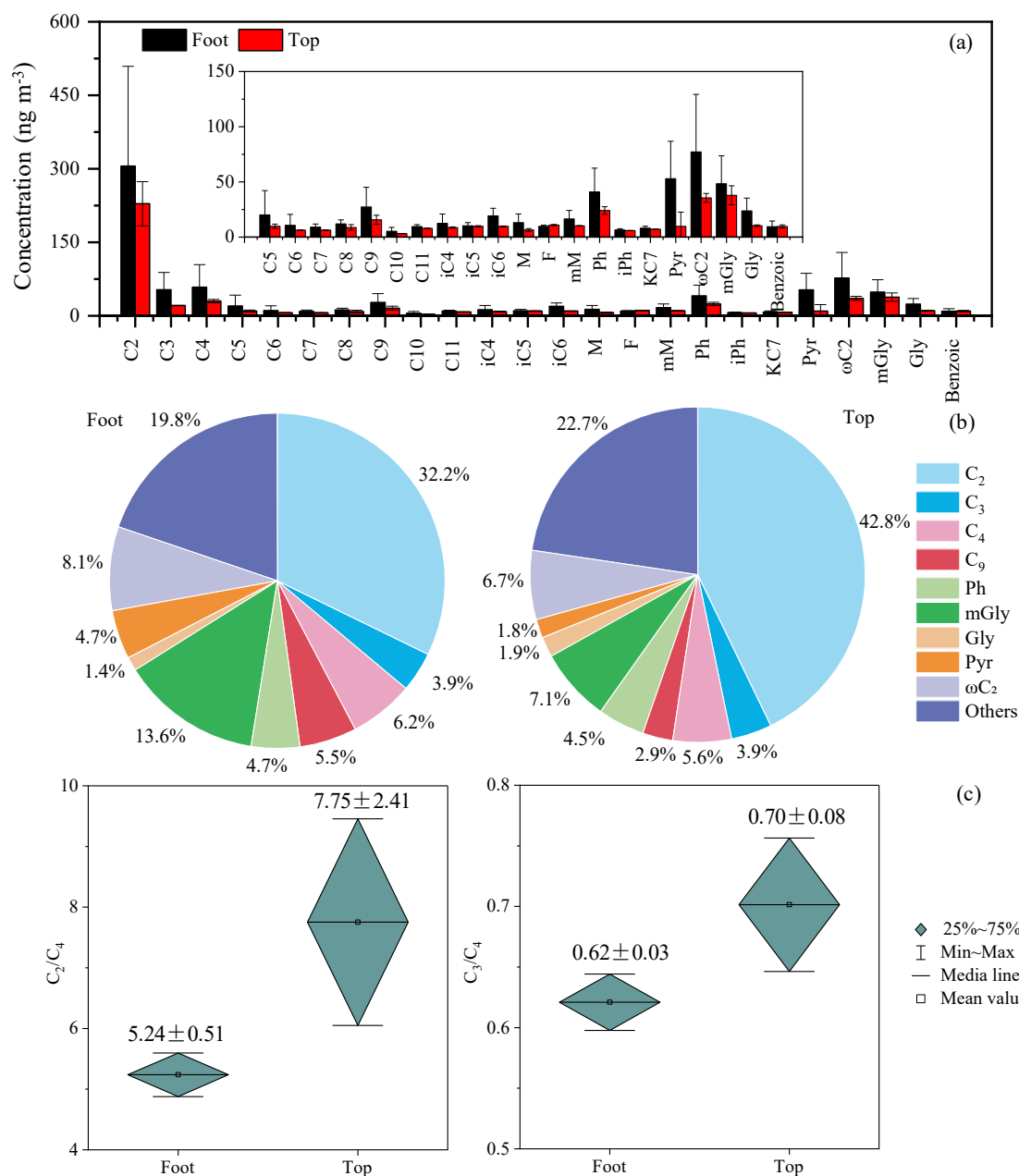


Figure 4 Molecular distribution of dicarboxylic acids and related compounds (a), relative percentages of major dicarboxylic acids (b), and ratios of C₂/C₄ and C₃/C₄ (c) at the foot and top of Mount Hua during dust events

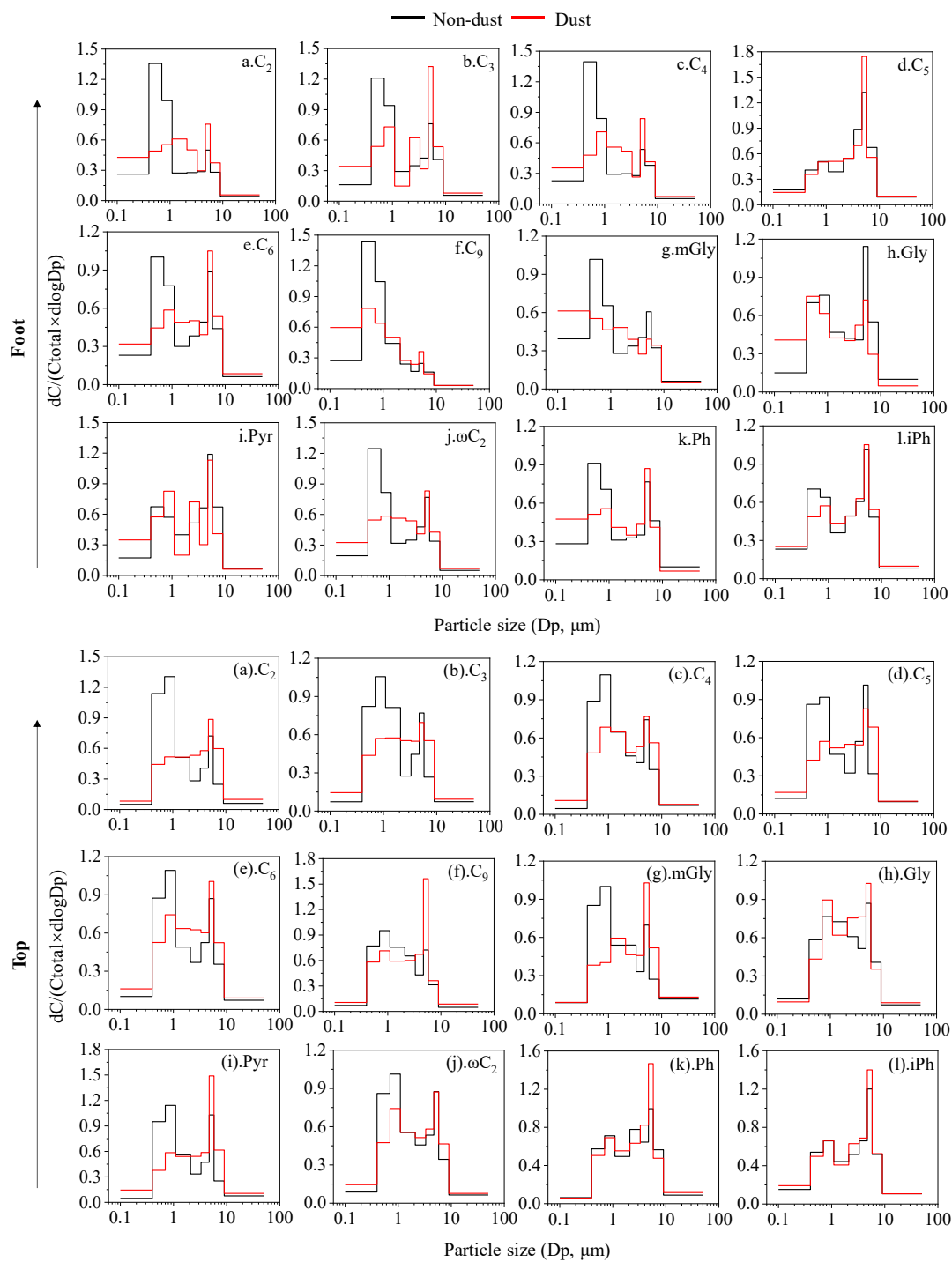


Figure 5 Size Distribution-distribution patterns of dicarboxylic acids at the foot and top of Mount Hua during non-dust and dust periods

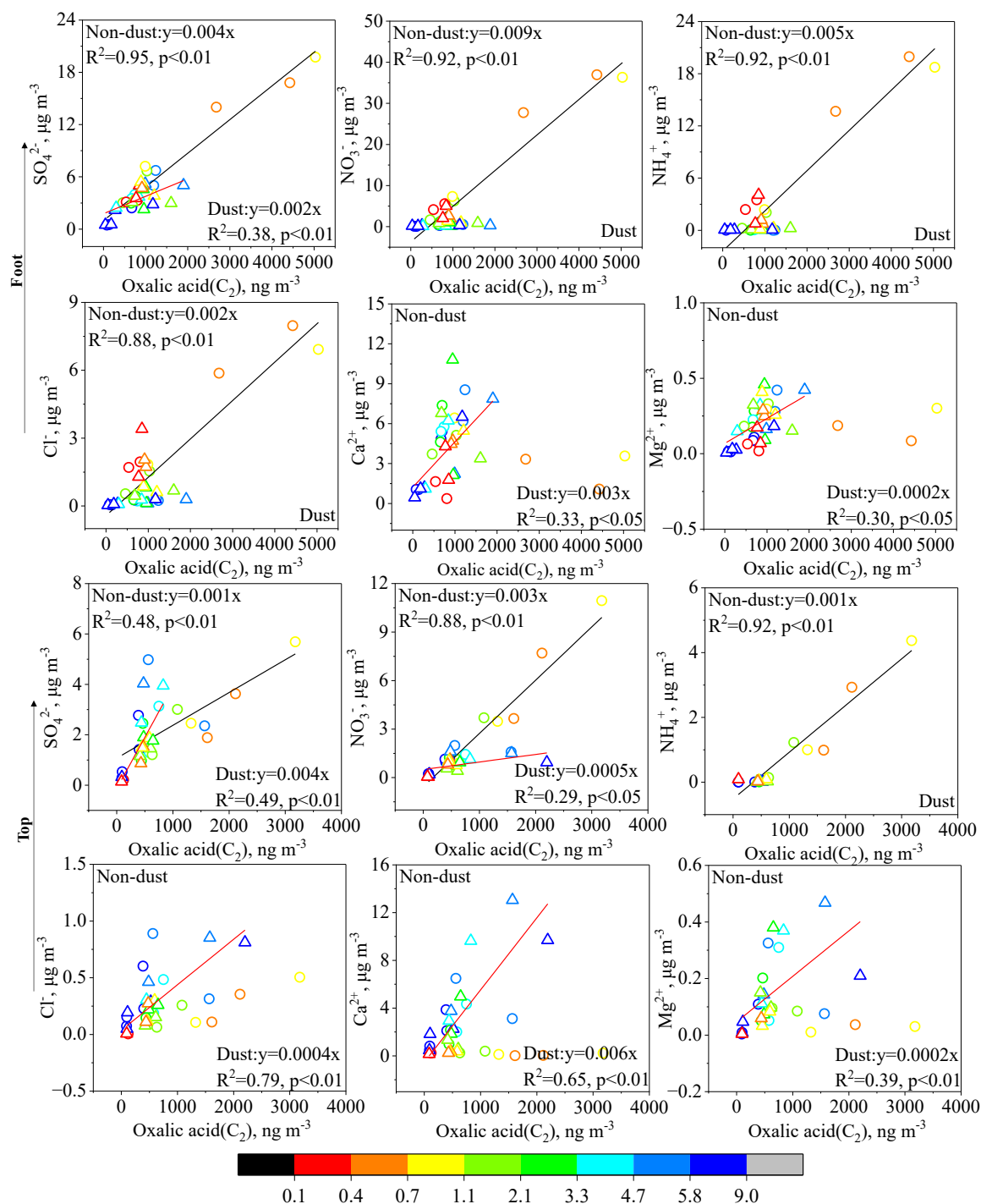


Figure 6 Correlation of C_2 with water-soluble ions at the foot and top of Mount Hua during non-dust and dust periods (Circles in the figure represent non-dust periods, and triangles represent dust periods)

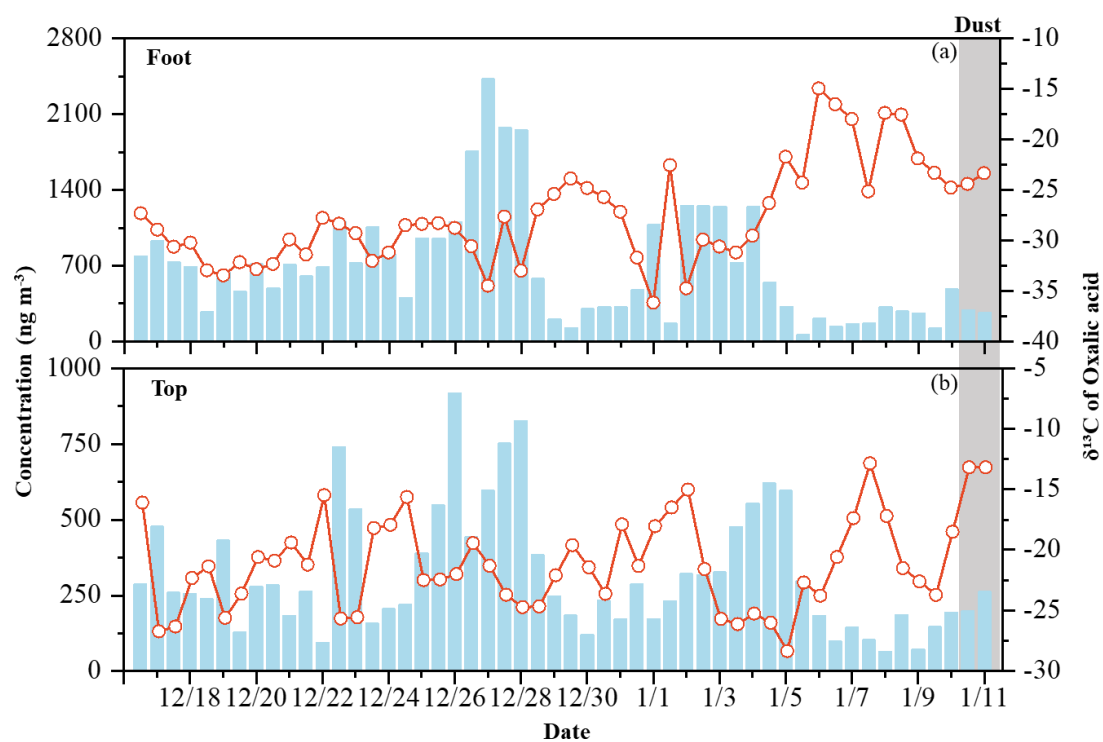


Figure 7 Stable carbon isotopes ($\delta^{13}\text{C}$) of C_2 in $\text{PM}_{2.5}$ at the foot and top of Mount Hua

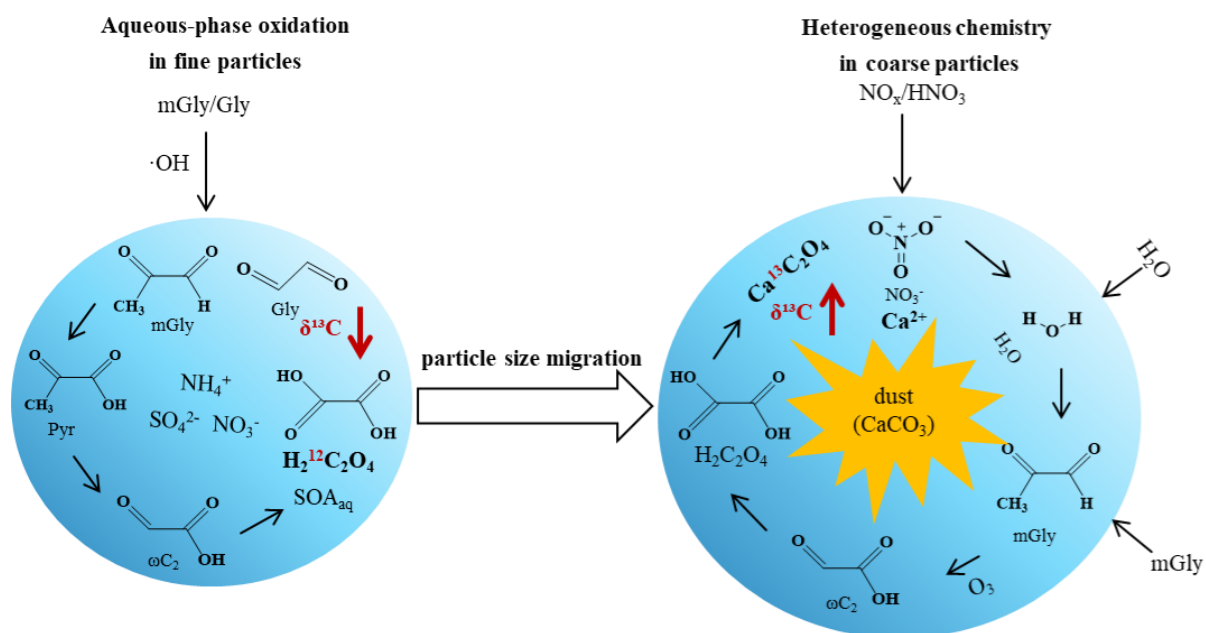


Figure 8 Mechanism diagram of dust-driven particle-size migration and formation pathways of C₂

Table 1 Average concentrations of dicarboxylic acids and related compounds in $PM_{2.5}$ at the foot and top of Mount Hua during non-dust periods

Compound	Foot			Top			R _{F/T}
	Daytime (<i>N</i> = 26)	Nighttime (<i>N</i> = 27)	Whole (<i>N</i> = 53)	Daytime (<i>N</i> = 26)	Nighttime (<i>N</i> = 27)	Whole (<i>N</i> = 53)	
Dicarboxylic acids							
Oxalic, C ₂	766 ± 552	585 ± 497	674 ± 528	312 ± 224	299 ± 186	306 ± 204	2.2
Malonic, C ₃	86 ± 58	71 ± 55	78 ± 56	54 ± 39	53 ± 32	53 ± 35	1.5
Succinic, C ₄	152 ± 83	112 ± 77	132 ± 82	60 ± 47	57 ± 46	58 ± 46	2.3
Glutaric, C ₅	38 ± 19	27 ± 14	32 ± 17	22 ± 27	18 ± 16	20 ± 22	1.6
Adipic, C ₆	17 ± 15	14 ± 13	15 ± 14	12 ± 13	10 ± 6.3	11 ± 9.8	1.4
Pimelic, C ₇	15 ± 5.0	12 ± 5.0	13 ± 5.1	9.2 ± 3.3	8.9 ± 2.3	9.0 ± 2.8	1.4
Suberic, C ₈	16 ± 4.1	14 ± 3.4	15 ± 3.8	13 ± 3.7	12 ± 3.5	12 ± 3.6	1.3
Azelaic, C ₉	175 ± 110	131 ± 106	153 ± 110	28 ± 19	27 ± 17	27 ± 18	5.7
Sebacic, C ₁₀	19 ± 15	14 ± 11	16 ± 14	5.3 ± 3.5	5.3 ± 3.7	5.3 ± 3.5	3.0
Undecanedioic, C ₁₁	14 ± 3.8	12 ± 2.5	13 ± 3.3	9.9 ± 1.8	9.2 ± 1.7	9.6 ± 1.8	1.4
Methylmalonic, iC ₄	36 ± 17	24 ± 13	29 ± 16	13 ± 9.4	12 ± 7.6	13 ± 8.5	2.2
Mehtylsuccinic, iC ₅	16 ± 5.4	16 ± 7.9	16 ± 6.7	10 ± 3.2	10 ± 2.4	10 ± 2.8	1.6
Methylglutaric, iC ₆	18 ± 6.7	14 ± 5.4	16± 6.3	19 ± 7.7	19 ± 6.2	19 ± 6.9	0.8
Maleic, M	22 ± 12	17 ± 11	19 ± 12	13 ± 10	13 ± 5.7	13 ± 7.9	1.5
Fumaric, F	13 ± 3.4	11 ± 2.4	12 ± 3.0	10 ± 1.2	10 ± 1.0	9.7 ± 1.1	1.2
Methylmaleic, mM	29 ± 18	27 ± 29	28 ± 24	17 ± 9.1	16 ± 6.2	16 ± 7.7	1.8
Phthalic, Ph	89 ± 43	71 ± 45	80 ± 44	44 ± 23	39 ± 20	41 ± 22	2.0
Isophthalic, iPh	15 ± 5.4	13 ± 7.0	14 ± 6.3	6.5 ± 1.2	6.5 ± 0.8	6.5 ± 1.0	2.2
Ketopimelic, kC ₇	10.2 ± 2.3	9.4 ± 2.1	9.8 ± 2.2	8.3 ± 1.9	8.0 ± 1.7	8.1 ± 1.8	1.2
Ketocarboxylic acids							
Pyruvic, Pyr	85 ± 45	73 ± 59	79 ± 53	52 ± 38	54 ± 30	53 ± 34	1.5
Glyoxylic, ωC ₂	230 ± 201	198 ± 200	214 ± 199	79 ± 58	75 ± 47	77 ± 52	2.8
α-Dicarbonyls							
Glyoxal, Gly	29 ± 15	29 ± 23	29 ± 19	23 ± 13	24 ± 10	24 ± 12	1.2
Methylglyoxal, mGly	128 ± 49	87 ± 42	107 ± 49	49 ± 28	48 ± 23	48 ± 25	2.2
Others							
Benzoic, Ha	15 ± 7.1	12 ± 6.7	13 ± 6.9	9.6 ± 5.4	9.3 ± 5.1	9.4 ± 5.2	1.4
Total detected (ng m ⁻³)	2029 ± 1294	1594 ± 1238	1807 ± 1280	879 ± 591	842 ± 481	860 ± 534	2.1

Table 2 Average concentrations of dicarboxylic acids and related compounds in PM_{2.5} at the foot and top of Mount Hua during dust events

Compound	Foot			Top		
	Daytime (N = 1)	Nighttime (N = 1)	Whole (N = 2)	Daytime (N = 1)	Nighttime (N = 1)	Whole (N = 2)
Oxalic, C ₂	289	262	276 ± 20	261	197	229 ± 45
Malonic, C ₃	28	38	33 ± 7.3	21	21	21 ± 0.1
Succinic, C ₄	47	59	53 ± 8.9	28	33	30 ± 3.5
Glutaric, C ₅	15	16	15 ± 0.5	8.4	11	10 ± 1.9
Adipic, C ₆	9.8	10.2	10 ± 0.3	6.2	6.4	6.3 ± 0.2
Pimelic, C ₇	7.6	9.2	8.4 ± 1.1	6.3	6.3	6.3 ± 0.0
Suberic, C ₈	9.2	11	10 ± 1.3	6.9	11	8.7 ± 2.6
Azelaic, C ₉	41	54	47 ± 9.4	13	19	16 ± 4.1
Sebacic, C ₁₀	4.7	5.6	5.1 ± 0.7	3.2	3.2	3.2 ± 0.0
Undecanedioic, C ₁₁	9.3	10	9.8 ± 0.7	8.0	7.9	8.0 ± 0.0
Methylmalonic, iC ₄	15	16	16 ± 0.3	8.2	9.1	8.7 ± 0.6
Mehtylsuccinic, iC ₅	12	11	12 ± 0.9	9.4	10.2	9.8 ± 0.6
Methylglutaric, iC ₆	15	16	16 ± 0.3	9.5	10.0	9.8 ± 0.3
Maleic, M	7.8	10	9.1 ± 1.8	7.3	5.6	6.4 ± 1.3
Fumaric, F	13	15	14 ± 1.4	10.5	11.3	10.9 ± 0.6
Methylmaleic, mM	12	13	13 ± 0.2	10.3	10.2	10.3 ± 0.0
Phthalic, Ph	39	41	40 ± 1.3	22	27	24 ± 3.5
Isophthalic, iPh	9.5	12	11 ± 1.6	5.9	5.9	5.9 ± 0.0
Ketopimelic, kC ₇	7.5	7.9	7.7 ± 0.3	7.2	7.1	7.2 ± 0.1
Pyruvic, Pyr	29	52	40 ± 17	0.6	19	9.8 ± 13
Glyoxylic, ωC ₂	51	87	69 ± 26	33	38	36 ± 4.0
Glyoxal, Gly	14	8.7	12 ± 4.1	9.8	10.9	10.4 ± 0.7
Methylglyoxal, mGly	92	140	116 ± 34	32	44	38 ± 8.6
Benzoic, Ha	12	17	15 ± 3.8	8.7	10.7	9.7 ± 1.4
Total detected (ng m ⁻³)	761	949	855 ± 142	535	533	534 ± 92

Table 3 Comparison of concentrations of dicarboxylic acids and related compounds in particulate matter of different particle size ranges ($\leq 2.1 \mu\text{m}$ and $> 2.1 \mu\text{m}$) at the foot and top of Mount Hua during non-dust and dust periods

Compound	Foot						Top					
	Non-dust		Dust		$R_{D/N}$		Non-dust		Dust		$R_{D/N}$	
	$\leq 2.1 \mu\text{m}$	$> 2.1 \mu\text{m}$	$\leq 2.1 \mu\text{m}$	$> 2.1 \mu\text{m}$	$\leq 2.1 \mu\text{m}$	$> 2.1 \mu\text{m}$	$\leq 2.1 \mu\text{m}$	$> 2.1 \mu\text{m}$	$\leq 2.1 \mu\text{m}$	$> 2.1 \mu\text{m}$	$\leq 2.1 \mu\text{m}$	$> 2.1 \mu\text{m}$
<u>Oxalic</u> , C ₂	8662	2718	4880	2843	0.3	0.6	5512	2217	2161	2301	0.4	1.1
<u>Malonic</u> , C ₃	750	388	361	371	0.5	1.0	757	359	305	253	0.5	0.8
<u>Succinic</u> , C ₄	1505	594	829	505	0.4	0.6	986	488	371	294	0.5	0.8
<u>Glutaric</u> , C ₅	238	350	199	297	1.5	1.5	557	390	334	324	0.7	1.0
<u>Adipic</u> , C ₆	427	277	347	292	0.6	0.8	457	275	290	240	0.6	0.8
<u>Azelaic</u> , C ₉	2313	371	1097	304	0.2	0.3	646	302	379	396	0.5	1.0
<u>Methylglyoxal</u> , mGly	237	119	217	90	0.5	0.4	146	67	49	55	0.5	1.1
<u>Glyoxal</u> , Gly	312	268	237	145	0.9	0.6	192	129	140	114	0.7	0.8
<u>Pyruvic</u> , Pyr	729	800	687	513	1.1	0.7	1022	521	451	633	0.5	1.4
<u>Glyoxylic</u> , ωC ₂	2181	1171	979	663	0.5	0.7	1138	678	658	545	0.6	0.8
<u>Phthalic</u> , Ph	1018	652	499	386	0.6	0.8	564	481	302	356	0.9	1.2
<u>Isophthalic</u> , iPh (ng m^{-3})	206	184	157	163	0.9	1.0	162	173	146	169	1.1	1.2