

Dear Editor,

We have carefully revised our manuscript (MS No.: egusphere-2025-3094) entitled “Measurement report: Observational insights into the impact of dust transport on atmospheric dicarboxylic acids in ground region and free troposphere” in response to the comments from the two reviewers. Our point-by-point responses and the corresponding modifications are detailed in the attached file.

Should you have any further questions regarding our paper, please do not hesitate to contact me at lijj@ieecas.cn.

Thank you for your time and consideration.

Best regards,

Jianjun Li

On behalf of all co-authors

Oct 2, 2025

Reviewer comments:

Reviewer #1:

General comment:

This study conducted simultaneous observations at the foot and top of Mount Hua, systematically analyzing the distribution characteristics of dicarboxylic acids (with a focus on oxalic acid, C₂) in PM_{2.5} and size-segregated samples, along with their $\delta^{13}\text{C}$ composition during both non-dust and dust periods. The research data demonstrate novelty and provide important evidence for understanding the transformation processes of SOA and the interactions between dust and anthropogenic emissions in mountainous environments. While the manuscript is fundamentally sound, certain aspects require further clarification and refinement.

Response: We sincerely appreciate the reviewer's positive assessment and constructive comments, which are very helpful for improving our manuscript. Our point-by-point responses and corresponding revisions are detailed below.

1. The introduction would benefit from smoother transitions between paragraphs. Currently, there's a noticeable jump from discussing research gaps (end of paragraph 2) to introducing nitrate-aged dust effects (paragraph 3) without adequate transitional phrasing.

Response: We thank the reviewer for this constructive suggestion. We have improved the logical flow by adding a transitional sentence at the beginning of the third paragraph. This sentence connects the research gap mentioned at the end of the second paragraph (unclear mechanisms of dust-organic interactions) to the specific focus of our study (dicarboxylic acids as key tracers). The revised text reads as follows:

Lines 70-73: 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).

2. Line 64-66: "Results show that dust storms have a less pronounced impact on ground aerosols than on the free troposphere of the Guanzhong Plain (Liu et al., 2024) ..." Please specify the exact chemical species (e.g., water-soluble ions, carbonaceous fractions) and the particle-size ranges examined by Liu et al., 2024, and clarify whether their sampling sites coincide with those of the present study to avoid any potential confusion.

Response: We thank the reviewer for this suggestion. We have clarified that Liu et al. (2024) was conducted concurrently at our study sites, analyzing water-soluble inorganic ions in PM_{2.5} and size-segregated aerosols. The relevant sentence now reads:

Lines 64-67: Liu et al. (2024) compared and analyzed the concentrations and size distributions of water-soluble inorganic ions during dust and non-dust periods, finding that the impact of dust on ground aerosols in the Guanzhong Plain is weaker than that in the free troposphere.

3. Further refinement of language and optimization of sentence structure are needed. For instance, line 46-47, "Dust degrades air quality near its source and can be transported over long distances by winds, impacting the climate on hemispheric and global scales" could be improved to: "Dust not only impairs air quality locally but also undergoes long-range transport, ultimately

affecting both hemispheric and global climate systems”; Line 60-62: “Originating in southern Mongolia and China’s western Inner Mongolia, it was intense and far-reaching, causing rapid air quality deterioration in the affected areas” can be revised as “This severe dust storm, originating from southern Mongolia and western Inner Mongolia, triggered rapid air quality deterioration across downwind regions”.

Response: We thank the reviewer for these constructive suggestions to improve the clarity and conciseness of our manuscript. Following the reviewer's specific recommendations, we have revised the two sentences accordingly:

The sentence in Lines 46-47 has been revised to: “Dust not only impairs air quality locally but also undergoes long-range transport, ultimately affecting both hemispheric and global climate systems.”

The sentence in Lines 60-62 has been revised to: “This severe dust storm, originating from southern Mongolia and western Inner Mongolia, triggered rapid air quality deterioration across downwind regions.”

In addition, we have performed a comprehensive language check throughout the manuscript to refine phrasing and optimize clarity.

4. Line 62: The introduction merely cites Figure S1 without clarifying its relevance. Revise the sentence so that specific elements of the figure are explicitly linked to the research questions.

Response: We thank the reviewer for this suggestion. We have revised the sentence to explicitly link Figure S1 to the spatial extent of the dust event. The updated text now reads:

Lines 61-64: 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.

5. Maintain terminological consistency: “secondary organic aerosols” was abbreviated as “SOA” in line 68; please use “SOA” throughout the manuscript (including lines 102 and 360) and avoid alternating with the full term.

Response: We thank the reviewer for pointing this out. The abbreviation “SOA” has been used consistently throughout the revised manuscript, including in Lines 102 and 360.

6. Unify citation format: for example, line 38 “(Maher et al. 2010; Liang et al., 2022)” should be corrected to “(Maher et al., 2010; Liang et al., 2022)” with the comma added; please check the entire manuscript.

Response: We appreciate the reviewer’s careful attention to detail. We have corrected the citation format (e.g., in Line 38) and have thoroughly checked the entire manuscript to ensure that all in-text citations now follow the consistent format of “(Author et al., Year)”.

7. Line 121-139: The analytical methods lack sufficient detail. Please provide the exact column model, detection limits for target ions (e.g., Cl⁻, NO₃⁻, SO₄²⁻) and for OC/EC, together with a full description of the drying protocol for dicarboxylic acid samples, the hexane-wash purification of their organic derivatives, and the nitrogen blow-down concentration procedure.

Response: We sincerely thank the reviewer for this constructive suggestion. We have now expanded the Methods section to provide comprehensive details regarding the analytical

procedures, as requested. The modifications are presented in the revised manuscript (Section 2.2.1 and 2.2.2) and summarized below:

Lines 125-173: **2.2.1 Determination of carbonaceous species and water-soluble inorganic ions**

The concentrations of organic carbon (OC) and elemental carbon (EC) in PM_{2.5} were determined 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 cm² 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 µg cm⁻² for OC and 0.03 µg 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 MΩ, Merck, France) via a combined process of 1-hour ultrasonication and 1-hour mechanical shaking. After filtration through a 0.45 µm membrane, the extracts were preserved at 4 °C for subsequent analysis. Water-soluble inorganic ions (NH₄⁺, K⁺, Mg²⁺, Ca²⁺, Cl⁻, NO₃⁻, and SO₄²⁻) 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 mM Na₂CO₃ 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 µg 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 and meteorological parameters including relative humidity (RH) and temperature (T). 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 PM_{2.5} and size-segregated aerosols was conducted based on an established derivatization method (Shen et al., 2022). A quarter of each filter was extracted by ultrasonication in ultrapure water (Milli-Q, 18.2 MΩ, Merck, France) for three sequential 15-minute intervals. To maximize the recovery of low-molecular-weight acids (e.g., C₂), the extracts were alkalized to pH 8.5–9.0 with 0.1 M KOH prior to the concentration step. 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 residues were derivatized with 14% BF₃/n-butanol at 100 °C for 1 hour to convert 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. The n-hexane layer was concentrated by rotary evaporation and N₂ blow-down, reconstituted in 100 µL n-hexane, and finally analyzed by GC-FID (HP 6890, Agilent Technologies, USA) (Wang et al., 2012). The recovery of the target compounds was 83% for C₂ and ranged from 87% to 110% for other diacids.

Stable carbon isotopic compositions ($\delta^{13}\text{C}$) of 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.

8. Line 129: Please specify the exact source(s) of the relative humidity and temperature data.

Response: We thank the reviewer for this comment. The requested clarification has been added to the manuscript. The revised text now reads: “**Meteorological data for the top and the foot were obtained from the Mount Hua Meteorological Station and the Huayin Meteorological Bureau, respectively.**”

9. The discussion of results does not adequately compare with key reference. For example, although the study points out that oxalic acid (C_2) at the foot shows significant diurnal variations, while the top shows no significant differences, it does not compare these findings with reference on photochemical processes in similar topographical studies. Moreover, the study uses the C_2/C_4 ratio of 5.84 (at the top) as an indicator of photochemical aging but fails to cite the classic study by Kawamura and Ikushima (1993), who first proposed that this ratio can indicate the degree of aging. It is recommended to supplement the study with references to Kawamura and Ikushima (1993) and other relevant studies in high-altitude regions to further substantiate the applicability of the C_2/C_4 ratio as an aging indicator.

Response: We fully agree with the reviewer’s comment. Following the suggestion, we have now cited the study by Kawamura and Ikushima (1993) when discussing the C_2/C_4 and C_3/C_4 ratios as aging indicators. Additionally, we have incorporated comparative analyses with relevant studies from high-altitude areas such as Mount Hua and Mount Tai, which further validate the reliability of our findings. The revised text now reads:

Lines 220-232: Aerosol aging indicators further confirmed distinct oxidation processes between different altitudes. According to existing research, succinic acid (C_4) can be via hydroxylation to generate C_2 and malonic acid (C_3), while C_3 can be further converted to C_2 through intermediates such as hydroxymalonic acid or ketomalonic acid (Kawamura et al. 1993; Kunwar and Kawamura, 2014; Hoque et al., 2017). Therefore, the C_2/C_4 and C_3/C_4 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 C_2/C_4 ratio at the top of Mount Hua was 5.84 ± 0.32 , and the C_3/C_4 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.

10. Please provide an explanation for the notable discrepancy observed between the nitrate-dominated C_2 correlation (with an R^2 value of 0.79) that you have observed and the previously

reported sulfate-dominated mechanisms by Meng et al. (2018) and Wang et al. (2012).

Response: Thank you for your valuable suggestion. We have added an explanation for this discrepancy in the discussion section of the manuscript, as detailed below:

Lines 246-259: Contrary to previous studies (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. 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 $PM_{2.5}$. 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 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).

11. Line 265-268: the authors mention “the proportion of C_2 decreasing from 37.3% to 32.2% at the foot and increasing from 35.5% to 42.8% at the top” Is it sufficient to explain this solely based on humidity? Additionally, they mention that high humidity promotes the “secondary formation of C_2 ” Is this also referring to an aqueous-phase reaction?

Response: We thank the reviewer for this important comment. We agree that explaining the trend based solely on humidity is insufficient, and we confirm that the “secondary formation of C_2 ” in our context primarily refers to aqueous-phase reactions. We have revised the manuscript accordingly to provide a more comprehensive explanation.

Lines 322-326: During the dust event, the lower relative humidity at the foot ($RH = 24 \pm 8.5\%$) suppressed aqueous-phase oxidation, whereas the higher humidity at the top ($RH = 44 \pm 11\%$) favored 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.

12. It is recommended to divide the 16 subplots of Figure 3 into two groups for discussion: the correlations between precursors and C_2 at the foot and summit sites (Figures 3a-d, i-l), and the influences of inorganic ions and ALWC on C_2 (Figures 3e-h, m-p).

Response: We thank the reviewer for this constructive suggestion. The discussion of Fig. 3 has been reorganized into two distinct sections as recommended the correlations between C_2 and its key precursors at the foot and summit sites (Fig. 3a-d, i-l), and the influences of inorganic ions and ALWC on C_2 formation (Fig. 3e-h, m-p). The corresponding text has been revised to reflect this clearer structure and now reads:

Lines 233-237: This study compared the correlations between C_2 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.

13. Please explicitly cite the relevant figure numbers in the text, for example by adding “(Figure 4c)” after the mention of “C₂/C₄ ratios” on line 282, to strengthen the linkage between the narrative and the figures.

Response: We thank the reviewer for this suggestion. We have now added explicit citations to the relevant figure panels throughout the manuscript to strengthen the link between the narrative and the data. For example, as suggested, we have added “(Fig. 4c)” following the mention of the “C₂/C₄ ratios” on line 282. A thorough check has been performed to ensure all relevant figures are properly cited.

14. Line 340-341: the authors need to clarify whether the conclusion that “aqueous secondary organic aerosols (aqSOA) formed on dust surfaces promote SOA formation and drive the transition of particle size distribution from submicron to supermicron ranges” is based on the findings of Li et al. (2025) or represents original findings from this study. If it is a reference to the literature, it is recommended to clearly indicate the citation.

Response: We thank the reviewer for this important clarification. We have revised the sentence in question by explicitly citing Li et al. (2025) to attribute the conclusion that aqSOA formed on dust surfaces promotes SOA production and drives the size-distribution transition. The original general statement has been replaced with the following text:

Lines 398-401: Research by Li et al. (2025) further confirms that aqSOA formed on dust surfaces can effectively enhance SOA production and drive a transition in the size distribution from the submicron to the supermicron range, which is highly consistent with the observational results of this study.

15. Line 364-366: Please ensure consistency in verb tense, and it is recommended to replace “oxalate” with “C₂” for consistency in terminology usage.

Response: We thank the reviewer for this careful observation. We have corrected the verb tense inconsistency in Lines 364–366. Regarding terminology, we have conducted a thorough review and now use “C₂” as the primary term throughout the manuscript to ensure consistency. The term “oxalate” is retained exclusively in contexts that specifically describe its salt form with metal cations (e.g., calcium oxalate) to maintain chemical accuracy.

16. Please add the p-values for all correlation coefficients in Figures 3 and 6 so that readers can properly assess their statistical significance. Additionally, standardize the notation for dicarboxylic acids in Table 3 to match that used in Tables 1 and 2.

Response: We thank the reviewer for these valuable suggestions. We have added the p-values for all correlation coefficients in Fig. 3 and 6 to allow for a proper assessment of their statistical significance and have standardized the notation for dicarboxylic acids in Table 3 to ensure consistency with the format used in Tables 1 and 2.

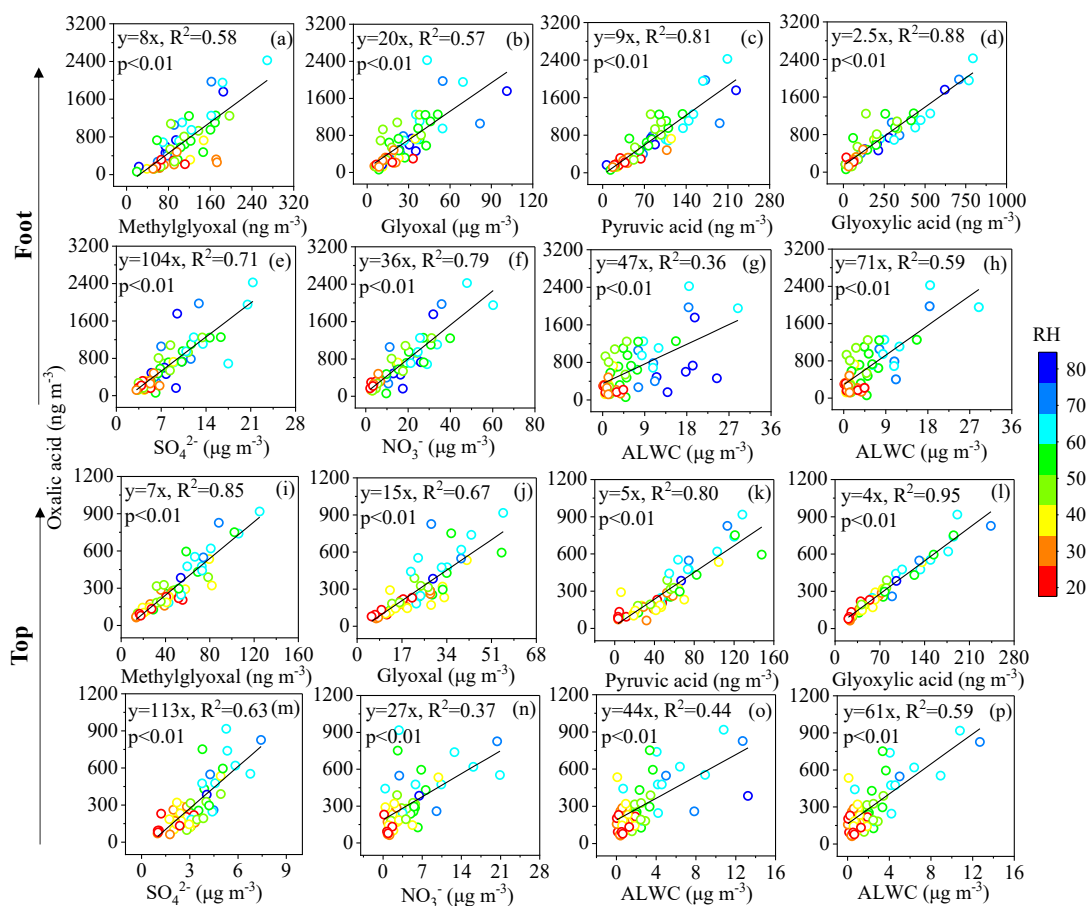


Figure 3 Correlation between C₂ and its key precursors, sulfate (SO₄²⁻), nitrate (NO₃⁻), and aerosol liquid water content (ALWC) at the foot and top of Mount Hua with varying relative humidity (RH)

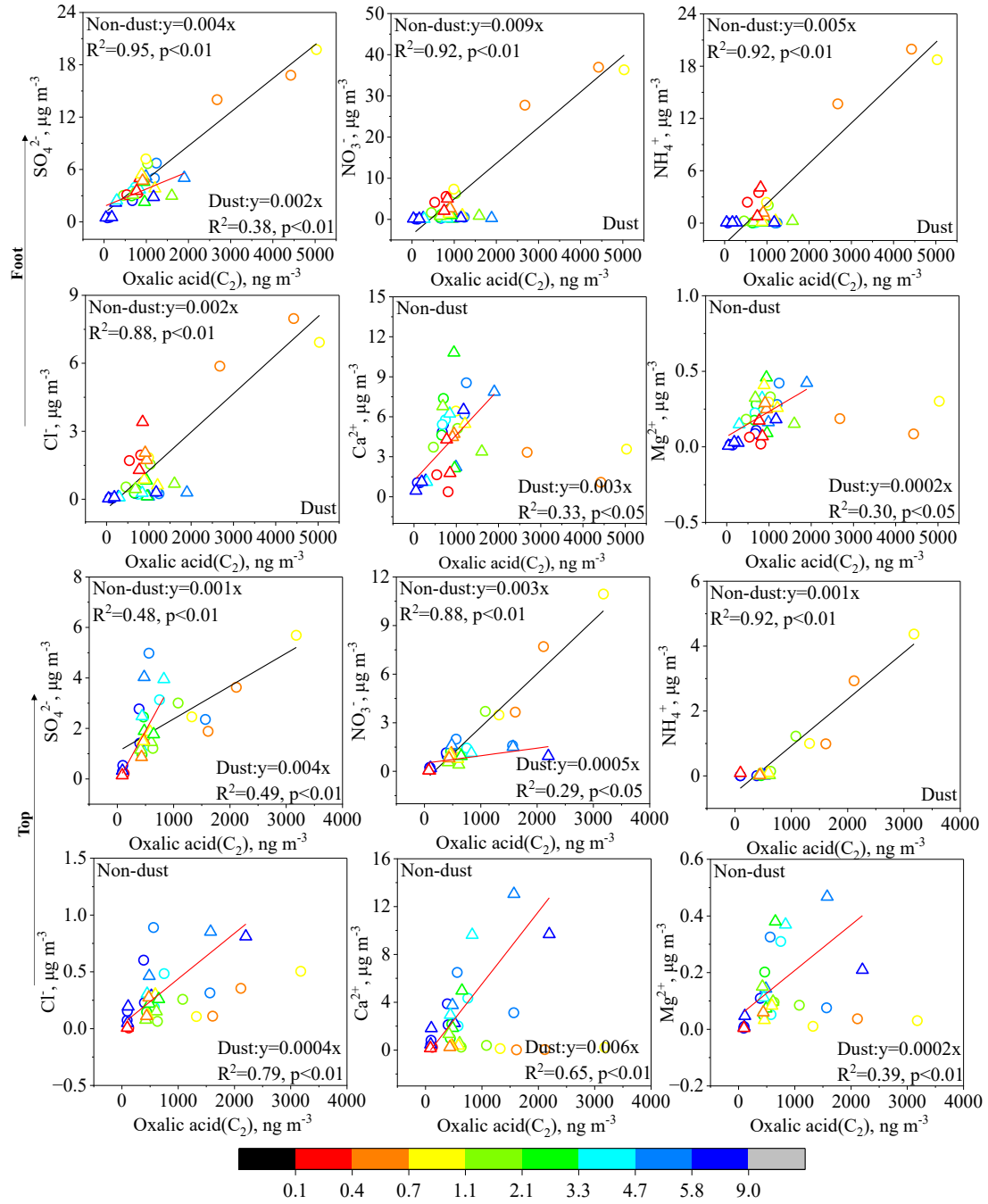


Figure 6 Correlation of C_2 with water-soluble ions at the foot and top of Mount Hua during non-dust and dust periods (Circles in the figure represent non-dust periods, and triangles represent dust periods)

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		$R_{D/N}$	
	$\leq 2.1 \mu\text{m}$	$> 2.1 \mu\text{m}$	$\leq 2.1 \mu\text{m}$	$> 2.1 \mu\text{m}$	$\leq 2.1 \mu\text{m}$	$> 2.1 \mu\text{m}$	$\leq 2.1 \mu\text{m}$	$> 2.1 \mu\text{m}$	$\leq 2.1 \mu\text{m}$	$> 2.1 \mu\text{m}$	$\leq 2.1 \mu\text{m}$	$> 2.1 \mu\text{m}$
Oxalic, C_2	8662	2718	4880	2843	0.3	0.6	5512	2217	2161	2301	0.4	1.1
Malonic, C_3	750	388	361	371	0.5	1.0	757	359	305	253	0.5	0.8
Succinic, C_4	1505	594	829	505	0.4	0.6	986	488	371	294	0.5	0.8
Glutaric, C_5	238	350	199	297	1.5	1.5	557	390	334	324	0.7	1.0
Adipic, C_6	427	277	347	292	0.6	0.8	457	275	290	240	0.6	0.8
Azelaic, C_9	2313	371	1097	304	0.2	0.3	646	302	379	396	0.5	1.0
Methylglyoxal, mGly	237	119	217	90	0.5	0.4	146	67	49	55	0.5	1.1
Glyoxal, Gly	312	268	237	145	0.9	0.6	192	129	140	114	0.7	0.8
Pyruvic, Pyr	729	800	687	513	1.1	0.7	1022	521	451	633	0.5	1.4
Glyoxylic, ωC_2	2181	1171	979	663	0.5	0.7	1138	678	658	545	0.6	0.8
Phthalic, Ph	1018	652	499	386	0.6	0.8	564	481	302	356	0.9	1.2
Isophthalic, iPh (ng m^{-3})	206	184	157	163	0.9	1.0	162	173	146	169	1.1	1.2

17. Line 442: “coarse/fine ratio” should be revised to “coarse/fine particle ratio” for greater precision.

Response: We thank the reviewer for this suggestion to improve terminological precision. We have revised “coarse/fine ratio” to “coarse/fine particle ratio” in Line 442 as recommended.

References

Fu, T. M., Jacob, D. J., Wittrock, F., Burrows, J. P., Vrekoussis, M., and Henze, D. K.: Global budgets of atmospheric glyoxal and methylglyoxal, and implications for formation of secondary organic aerosols. J. Geophys. Res. 113. D15303, <https://doi.org/10.1029/2007JD009505>, 2008.

Hoque, M. M. M., Kawamura, K., and Uematsu, M.: Spatio temporal distributions of dicarboxylic acids, ω -oxocarboxylic acids, pyruvic acid, α -dicarbonyls and fatty acids in the marine aerosols from the North and South Pacific, Atmos. Res., 185, 158–168,

<https://doi.org/10.1016/j.atmosres.2016.10.022>, 2017.

Huang, R. J., Li, Y. J., Chen, Q., Zhang, Y. L., Lin, C. S., Chan, C. K., Yu, J. Z., Gouw, J., Tong, S. R., Jiang, J. K., Wang, W. G., Ding, X., Wang, X. M., Ge, M. F., Zhou, W. J., Worsnop, D., Boy, M., Bilde, M., Dusek, U., Carlton, A. G., Hoffmann, T., McNeill, V. F., and Glasius M.: Secondary organic aerosol in urban China: A distinct chemical regime for air pollution studies, *Science*, 389, eadq2840. <https://doi.org/10.1126/science.adq2840>, 2025.

Kawamura, K., and Ikushima, K.: Seasonal changes in the distribution of dicarboxylic acids in the urban atmosphere, *Environ. Sci. Technol.*, 27, 2227-2235, <https://doi.org/10.1021/es00047a033>, 1993.

Li, W. J., and Shao L. Y.: Observation of nitrate coatings on atmospheric mineral dust particles. *Atmos. Chem. Phys.*, 9(6), 1863–1871. <https://doi.org/10.5194/acp-10-10521-2010>, 2009.

Meng, J. J., Wang, G. H., Li, J. J., Cheng, C. L., Ren, Y. Q., Huang, Y., Cheng, Y. B., Cao, J. J., and Zhang, T.: Seasonal characteristics of oxalic acid and related SOA in the free troposphere of Mt. Hua, central China: Implications for sources and for formation mechanisms, *Sci. Total Environ.*, 493, 1088–1097, <https://doi.org/10.1016/j.scitotenv.2014.04.086>, 2014.

Wang, G. H., Kawamura, K., Umemoto, N., Xie, M. J., Hu, S. Y., and Wang, Z. F.: Water-soluble organic compounds in PM_{2.5} and size-segregated aerosols over Mount Tai in North China Plain. *J. Geophys. Res.* 114, 1–10. <https://doi.org/10.1029/2008jd011390>. D19208, 2009.

Wang, G. H., Zhang, S., Wu, C., Zhu, T., Xu, X. B., Ge, S. S., Sun, H. T., Sun Z. R., Wang, J. X., Ji, Y. M., Gao, J., Ren, Y. Q., Li, H., Zhang, F., Wang, Y., and Seinfeld, J. H.: Atmospheric sulfate aerosol formation enhanced by interfacial anions. *PNAS Nexus*, 4, pgaf058. <https://doi.org/10.1093/pnasnexus/pgaf058>, 2025.

Warneck, P.: In-cloud chemistry opens pathway to the formation of oxalic acid in the marine atmosphere. *Atmos. Environ.* 37(17), 2423–2427, [https://doi.org/10.1016/S1352-2310\(03\)00136-5](https://doi.org/10.1016/S1352-2310(03)00136-5), 2003.

Yermakov, A. N., Aloyan, A. E., Arutyunyan, V. O., and Pronchev, G. B.: On the mechanism of sulfur dioxide oxidation in cloud drops. *Izv. Atmos. Oceanic Phys.*, 59(5), 538–547. <https://doi.org/10.1134/S0001433823050055>, 2023.

Zhi, M. K., Wang, G. C., Xu, L., Li, K. I., Nie, W., Niu, H. Y., Shao, L. Y., Liu, Z. R., Yi, Z. W., Wang, Y. T., Shi, Z. B., Ito, A., Zhai, S. X., and Li, W. J.: How acid iron dissolution in aged dust particles responds to the buffering capacity of carbonate minerals during Asian dust storms. *Environ. Sci. Technol.*, 59(12): 6167–6178. <https://doi.org/10.1021/acs.est.4c12370>, 2025.

Reviewer #2:

The manuscript examines vertical gradients of PM_{2.5} and size-segregated aerosols at the foot and summit of Mount Hua, emphasizing oxalic acid (C₂) formation and $\delta^{13}\text{C}$ signatures under dust events. The dataset has clear value and the topic—dust impacts on diacids and SOA pathways across altitude—is important. However, there are substantial questions and the English requires substantial polishing.

Response: We thank the reviewer for their thoughtful assessment of our manuscript and their valuable suggestions for improvement. We have carefully considered all the points raised and have revised the manuscript accordingly. Our point-by-point responses are detailed below.

Specific comments

1. 88–91: **Wang et al. (2015) discovered that nitric acid and nitrogen oxides react with dust to form calcium nitrate, which absorbs water vapor and creates a liquid phase on the dust surface. This process promotes the oxidation of water-soluble organic precursors to C₂, with nitrate playing a crucial role.** You cite nitrate's role but omit critical evidence that water-containing nitrate coatings (Ca(NO₃)₂) facilitate aqueous-phase SOA formation due to their very low deliquescence RH (Ca(NO₃)₂) absorbs water under atmospheric RH > 8%). Li et al. (National Science Review, 12, 10.1093/nsr/nwaf221, 2025) provide direct evidence for this mechanism; that work explicitly discussed where nitrate coatings are invoked.

Response: We sincerely thank the reviewer for this insightful comment and for highlighting the importance of explicitly citing the role of aqueous nitrate coatings in facilitating SOA formation. We fully agree that the mechanism of Ca(NO₃)₂ induced aqueous-phase reactions under low-RH conditions is critical to our discussion. We have revised the manuscript accordingly to incorporate the suggested reference and to more clearly articulate the deliquescence behavior and its implications for SOA formation. The modified text now reads as follows:

Lines 70-86: 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 (Ca(NO₃)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 (C₂). Research by Li et al. (2025) provides direct evidence for this mechanism, showing that aqueous nitrate coatings (Ca(NO₃)₂), due to their very low deliquescence relative humidity (absorbing water at atmospheric RH > 8%), effectively promote the formation of

aqueous secondary organic aerosols (aqSOA). Thus, aged dust surfaces provide critical reactive interfaces for aqSOA formation.

2. Statistical support for “most striking differences” throughout your manuscript.

Wherever you describe significant differences (e.g., foot vs. top; dust vs. non-dust; day vs. night), please perform and report statistical tests (e.g., two-sample t-tests with assumptions stated). The manuscript currently lacks the statistical support needed to substantiate claims of significance.

Response: Thank you very much for your valuable comments. In response to your suggestions, we have now supplemented the manuscript with complete statistical analyses for all comparative conclusions to provide solid statistical support. Specifically, independent samples t-tests were used for the comparison between the foot and top of the mountain during non-dust periods, with detailed results provided in Supplementary Tables S3a and S3b; paired samples t-tests were used for the day-night variation comparisons, with detailed results shown in Tables S4a, S4b, and S4c. All descriptions such as “most striking” or “significantly higher” now explicitly cite the specific statistical results (including t-value, degrees of freedom, p-value, and 95% confidence interval). Furthermore, for the dust events with a limited sample, statistical testing was not performed, and the relevant descriptions have been adjusted accordingly.

Lines 177-191: During non-dust periods, significant vertical differences were observed in the chemical composition of PM_{2.5} between the foot and top of Mount Hua. The average PM_{2.5} concentration at the foot ($127 \pm 48 \mu\text{g m}^{-3}$) 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). Carbonaceous aerosols and water-soluble ions were significantly enriched at the foot (all $p < 0.001$; Table S3a). As key components of WSOC (Kawamura et al., 2016; Yang et al., 2020), the total concentration of diacids at the foot ($1808 \pm 1280 \text{ ng m}^{-3}$) 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 (C₉), a biomarker of biomass burning (Kalogridis et al., 2018; Shen et al., 2022), with the concentration at the foot ($153 \pm 110 \text{ ng m}^{-3}$) being 5.7 times higher than that 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). Consequently, the contribution of C₉ to total diacids was substantially greater at the foot (8.5%) than at the top (3.2%) (Fig. 2b).

Lines 198-209: 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 day-night differences (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), C₂ also displayed marked day-night differences (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 showed no clear day-

night differences ($p = 0.341\text{--}0.917$; Table S4c), consistent with the patterns of $\text{PM}_{2.5}$ ($p = 0.979$), OC ($p = 0.766$), and other major components (Table S4a).

Table S3a. Independent samples t-test results for $\text{PM}_{2.5}$, major chemical components, and meteorological parameters between the foot and top of Mount Hua during non-dust events

Parameter	Mean difference (Foot-top)	95% CI	t-value	df	p-value
$\text{PM}_{2.5}$ ($\mu\text{g m}^{-3}$)	99	[86, 113]	14.60	61.05	<0.001
OC ($\mu\text{g m}^{-3}$)	12	[10, 15]	10.83	60.59	<0.001
EC ($\mu\text{g m}^{-3}$)	3.6	[3.1, 4.2]	12.44	57.00	<0.001
WSOC ($\mu\text{g m}^{-3}$)	6.2	[4.5, 7.9]	7.34	64.41	<0.001
SO_4^{2-} ($\mu\text{g m}^{-3}$)	5.2	[4.0, 6.4]	8.40	64.57	<0.001
NO_3^- ($\mu\text{g m}^{-3}$)	12	[8.3, 16]	6.38	65.11	<0.001
Cl^- ($\mu\text{g m}^{-3}$)	3.0	[2.6, 3.4]	13.51	55.09	<0.001
Ca^{2+} ($\mu\text{g m}^{-3}$)	1.3	[1.0, 1.7]	7.06	94.60	<0.001
K^+ ($\mu\text{g m}^{-3}$)	0.6	[0.4, 0.7]	9.96	57.41	<0.001
Mg^{2+} ($\mu\text{g m}^{-3}$)	0.05	[0.03, 0.08]	4.12	102.3	<0.001
NH_4^+ ($\mu\text{g m}^{-3}$)	1.4	[1.1, 1.7]	8.07	82.95	<0.001
O_3 ($\mu\text{g m}^{-3}$)	-22	[-28, -16]	-6.94	90.69	<0.001
T ($^{\circ}\text{C}$)	5.9	[3.8, 7.9]	5.60	92.06	<0.001
RH (%)	7.6	[0.8, 14]	2.23	103.71	0.028
WS (m s^{-1})	-4.1	[-5.0, -3.3]	-9.69	56.94	<0.001

Table S3b. Independent samples t-test results of dicarboxylic acids and related compounds in PM_{2.5} at the foot and top of Mount Hua during non-dust events

Compound	Mean difference (Foot-top)	95% CI	t-value	df	p-value
Oxalic, C ₂	368	[213, 523]	4.74	67.16	<0.001
Malonic, C ₃	25	[6.6, 43]	2.71	87.37	0.008
Succinic, C ₄	73	[48, 99]	5.70	82.10	<0.001
Glutaric, C ₅	12	[4.5, 20]	3.16	98.72	0.002
Adipic, C ₆	4.6	[-0.001, 9.3]	1.99	93.82	0.05
Pimelic, C ₇	4.4	[2.8, 6.0]	5.48	81.2	<0.001
Suberic, C ₈	3.1	[1.7, 4.5]	4.28	103.6	<0.001
Azelaic, C ₉	126	[95, 156]	8.24	54.77	<0.001
Sebacic, C ₁₀	11	[7.2, 15]	5.79	59.20	<0.001
Undecanedioic, C ₁₁	3.3	[2.3, 4.3]	6.40	79.53	<0.001
Methylmalonic, iC ₄	17	[12, 22]	6.81	78.91	<0.001
Methylsuccinic, iC ₅	5.8	[3.8, 7.8]	5.83	69.66	<0.001
Methylglutaric, iC ₆	-3.6	[-6.1, -1.0]	-2.77	103.13	0.007
Maleic, M	6.1	[2.2, 10]	3.11	90.69	0.002
Fumaric, F	2.2	[1.3, 3.1]	5.01	66.42	<0.001
Methylmaleic, mM	12	[4.8, 19]	3.39	62.64	0.001
Phthalic, Ph	39	[25, 52]	5.72	75.35	<0.001
Isophthalic, iPh	7.4	[5.7, 9.2]	8.47	54.78	<0.001
Ketopimelic, kC ₇	1.7	[0.9, 2.4]	4.29	99.63	<0.001
Pyruvic, Pyr	26	[9.1, 43]	3.05	88.86	0.003
Glyoxylic, ωC ₂	137	[80, 193]	4.84	59.09	<0.001
Glyoxal, Gly	4.7	[-1.5, 11]	1.52	84.57	0.133
Methylglyoxal, mGly	59	[44, 74]	7.72	77.83	<0.001
Benzoic	4.0	[1.6, 6.3]	3.34	96.15	0.001
Category Summary					
Dicarboxylic acids	718	[453, 983]	5.40	69.15	<0.001
Ketocarboxylic acids	163	[91, 234]	4.53	63.76	<0.001
α-Dicarbonyls	63	[44, 83]	6.48	82.19	<0.001

Note: All concentrations are in ng m⁻³.

Table S4a. Paired samples t-test results of day-night differences for PM_{2.5}, major chemical components, and meteorological parameters at the foot and top of Mount Hua during non-dust events

Parameter	Sites	Mean difference (Day-night)	95% CI	t-value	p-value
PM _{2.5} (μg m ⁻³)	Foot	24	[4.0, 45]	2.46	0.021
	Top	-0.09	[-7.4, 7.2]	-0.026	0.979
OC (μg m ⁻³)	Foot	4.4	[1.4, 7.4]	2.99	0.006
	Top	-0.1	[-1.0, 0.7]	-0.30	0.766
EC (μg m ⁻³)	Foot	1.5	[0.7, 2.3]	3.96	0.001
	Top	0.03	[-0.2, 0.2]	0.27	0.793
WSOC (μg m ⁻³)	Foot	2.7	[0.7, 4.6]	2.84	0.009
	Top	0.1	[-0.6, 0.9]	0.30	0.770
SO ₄ ²⁻ (μg m ⁻³)	Foot	1.7	[0.1, 3.2]	2.23	0.035
	Top	0.3	[-0.2, 0.9]	1.37	0.184
NO ₃ ⁻ (μg m ⁻³)	Foot	2.6	[-1.1, 6.3]	1.44	0.163
	Top	0.2	[-2.0, 2.3]	0.16	0.878
Cl ⁻ (μg m ⁻³)	Foot	0.6	[0.2, 1.0]	2.89	0.008
	Top	0.1	[-0.03, 0.20]	1.59	0.123
Ca ²⁺ (μg m ⁻³)	Foot	0.8	[0.4, 1.2]	3.78	0.001
	Top	0.03	[-0.4, 0.4]	-0.14	0.889
K ⁺ (μg m ⁻³)	Foot	0.2	[0.1, 0.3]	3.31	0.003
	Top	-0.01	[-0.05, 0.03]	-0.39	0.703
Mg ²⁺ (μg m ⁻³)	Foot	0.04	[0.01, 0.07]	3.05	0.005
	Top	0.01	[-0.02, 0.04]	-0.53	0.602
NH ₄ ⁺ (μg m ⁻³)	Foot	0.8	[-0.3, 0.4]	0.47	0.640
	Top	-0.02	[-0.2, 0.2]	-0.16	0.875
O ₃ (μg m ⁻³)	Foot	18	[11, 26]	4.95	<0.001
	Top	1.9	[0.4, 3.5]	2.53	0.019
T (°C)	Foot	4.2	[3.1, 5.4]	7.34	<0.001
	Top	1.8	[0.6, 3.0]	3.00	0.006
RH (%)	Foot	-18	[-21, -14]	-10.89	<0.001
	Top	-6.0	[-11, -0.9]	-2.40	0.024
WS (m s ⁻¹)	Foot	0.4	[0.1, 0.7]	2.60	0.015
	Top	-0.8	[-2.1, 0.4]	-1.34	0.192

Table S4b. Paired samples t-test results for day-night differences of dicarboxylic acids and related compounds in PM_{2.5} at the foot of Mount Hua during non-dust events

Compound	Mean difference (Foot-top)	95% CI	t-value	p-value
Oxalic, C ₂	154	[23, 284]	2.41	0.023
Malonic, C ₃	12	[-2.3, 26]	1.71	0.098
Succinic, C ₄	34	[11, 58]	3.02	0.006
Glutaric, C ₅	10	[4.1, 16]	3.50	0.002
Adipic, C ₆	2.0	[-2.1, 6.1]	1.00	0.326
Pimelic, C ₇	1.8	[0.2, 3.6]	2.27	0.031
Suberic, C ₈	1.4	[0.01, 2.8]	2.07	0.049
Azelaic, C ₉	38	[-2.0, 77]	1.95	0.061
Sebacic, C ₁₀	4.0	[-1.3, 9.3]	1.56	0.132
Undecanedioic, C ₁₁	1.4	[-0.6, 3.4]	1.41	0.171
Methylmalonic, iC ₄	11	[4.9, 17]	3.77	0.001
Methylsuccinic, iC ₅	-1.1	[-4.0, 1.8]	-0.75	0.459
Methylglutaric, iC ₆	-3.0	[0.6, 5.5]	2.52	0.018
Maleic, M	3.8	[0.1, 7.4]	2.11	0.044
Fumaric, F	0.6	[-1.0, 2.3]	0.81	0.426
Methylmaleic, mM	1.0	[-6.0, 8.0]	0.29	0.776
Phthalic, Ph	15	[-3.4, 33]	1.68	0.106
Isophthalic, iPh	1.3	[-1.1, 3.7]	1.09	0.287
Ketopimelic, kC ₇	0.4	[-0.6, 1.4]	0.84	0.409
Pyruvic, Pyr	8.4	[-9.4, 26]	0.97	0.341
Glyoxylic, ωC ₂	23	[-38, 84]	0.77	0.448
Glyoxal, Gly	-1.0	[-8.8, 6.9]	-0.26	0.797
Methylglyoxal, mGly	36	[18, 53]	4.13	<0.001
Benzoic	1.7	[-1.0, 4.4]	1.33	0.196
Category Summary				
Dicarboxylic acids	293	[50, 536]	2.47	0.020
Ketocarboxylic acids	31	[-45, 107]	0.85	0.404
α-Dicarbonyls	35	[13, 56]	3.33	0.003

Note: All concentrations are in ng m⁻³.

Table S4c. Paired samples t-test results for day-night differences of dicarboxylic acids and related compounds in PM_{2.5} at the top of Mount Hua during non-dust events

Compound	Mean difference (Foot-top)	95% CI	t-value	p-value
Oxalic, C ₂	1.9	[-51, 55]	0.075	0.941
Malonic, C ₃	-1.2	[-13, 10]	-0.22	0.828
Succinic, C ₄	0.01	[-12, 12]	0.002	0.998
Glutaric, C ₅	3.0	[-4.1, 10]	0.87	0.390
Adipic, C ₆	1.4	[-2.4, 5.3]	0.78	0.443
Pimelic, C ₇	-0.1	[-1.1, 0.9]	-0.13	0.897
Suberic, C ₈	0.5	[-1.2, 2.2]	0.61	0.550
Azelaic, C ₉	0.2	[-6.1, 6.5]	0.062	0.951
Sebacic, C ₁₀	-0.2	[-1.3, 0.9]	-0.36	0.720
Undecanedioic, C ₁₁	0.4	[-0.6, 1.3]	0.76	0.457
Methylmalonic, iC ₄	0.4	[-2.6, 3.4]	0.28	0.782
Mehtylsuccinic, iC ₅	-0.1	[-1.7, 1.5]	-0.14	0.892
Methylglutaric, iC ₆	-0.6	[-3.9, 2.8]	-0.35	0.733
Maleic, M	0.0	[-3.3, 3.3]	0.00	1.000
Fumaric, F	-0.1	[-1.0, 0.7]	-0.30	0.770
Methylmaleic, mM	-0.4	[-3.2, 2.5]	-0.27	0.792
Phthalic, Ph	3.4	[-2.9, 9.8]	1.12	0.274
Isophthalic, iPh	-0.2	[-0.9, 0.4]	-0.71	0.481
Ketopimelic, kC ₇	0.004	[-0.7, 0.7]	0.011	0.992
Pyruvic, Pyr	-3.2	[-14, 7.8]	-0.60	0.553
Glyoxylic, ωC ₂	1.4	[-12, 15]	0.21	0.834
Glyoxal, Gly	-1.8	[-5.5, 2.0]	-0.97	0.341
Methylglyoxal, mGly	-0.5	[-9.2, 8.3]	-0.11	0.917
Benzoic	-0.1	[-2.0, 1.9]	-0.075	0.941
Category Summary				
Dicarboxylic acids	17	[-87, 122]	-0.343	0.734
Ketocarboxylic acids	-0.9	[-24, 22]	-0.082	0.935
α-Dicarbonyls	-1.4	[-13, 11]	-0.24	0.813

Note: All concentrations are in ng m⁻³.

3. Introduction section — Scientific motivation for foot vs. top comparison. You reference prior work (Shen et al., 2023) that already found distinct C₂ formation pathways at different elevations of Mount Hua during the summer. Please state explicitly: (i) the knowledge gap left by your previous studies, (ii) what new question or hypothesis this study addresses beyond the prior work.

Response: We thank the reviewer for this critical comment. We have revised the Introduction to explicitly state the knowledge gap remaining after our previous study (Shen et al., 2023) and the new scientific hypothesis addressed in this work. The modified text now reads as follows:

Lines 87-107: 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) demonstrated that summer daytime valley winds on Mount Hua transport organic acids from the foot to top, thereby altering the chemical composition of the free troposphere and establishing distinct formation pathways of C₂ at different altitudes. 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 observational data from a typical dust event in winter 2021, this study focuses on examining the impacts of dust transport on the molecular distribution, particle size characteristics of diacids, and the formation mechanisms of C₂. The work aims to elucidate the key role of heterogeneous chemical reactions on the surfaces of dust aerosols in the formation of SOA, providing new insights into regional atmospheric chemical processes during dust events.

4. 173–179 — Interpretation of diurnal behavior at altitude. The statement that oxalic acid (C₂) and its precursors show no significant diurnal fluctuation at the summit, implying limited local photochemical contributions aloft, is presented in the manuscript without supporting references or mechanistic discussion. This interpretation seems to be unclear and potentially inconsistent with established understanding. Cloud and in-cloud processes are widely recognized as important pathways for aqSOA formation in the free troposphere, and oxalate is known to associate preferentially with Fe-containing particles during in-cloud processing (e.g., Zhang et al., 2019, DOI: 10.1021/acs.est.8b05280). Fe is also known to catalyze the photochemical decomposition of oxalic acid. In this context, the claim that high-altitude C₂ shows no significant diurnal variation requires further justification and supporting references, as it appears difficult to reconcile with existing literature.

Response: We thank the reviewer for their valuable comments. In response to the points raised, we have made the following revisions to the manuscript. First, to ensure terminological accuracy, we have systematically replaced “diurnal variation” with “day-night differences”

throughout the text. This revision better aligns with our defined sampling periods, daytime (08:00–19:00) and nighttime (20:00–07:00 the next day). Second, the absence of clear day-night differences is consistent with the environmental characteristics of the high-altitude free troposphere. Such stability is typical for high-altitude sites located above the planetary boundary layer and is primarily governed by regional transport processes (Fu et al., 2010; Li et al., 2012; Meng et al., 2014). We fully agree with the reviewer’s insight regarding the importance of in-cloud processes and have accordingly expanded the relevant section in the manuscript. The revised text now emphasizes that in-cloud processes including the in-cloud oxidation of precursors and the iron-catalyzed decomposition of C₂ represent key mechanisms regulating the stable distribution of organic acid concentrations at high altitudes. Supporting references, such as Zhang et al. (2019), have been added accordingly.

Lines 198-219: 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 day-night differences (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), C₂ also displayed marked day-night differences (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 showed no clear day-night differences ($p = 0.341$ – 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). 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.

5. 241: You attribute higher OC fraction to “enhanced photochemical reactions and gas-particle conversion,” but no direct evidence is shown. Earlier text in your manuscript also suggests mixing with anthropogenic carbonaceous aerosols during long-range transport, and you report elevated azelaic acid (biomass burning tracer) at the summit. The manuscript does not provide clear evidence to distinguish between secondary formation and primary biomass-burning/transport contributions as explanations for the higher OC fraction.

Response: We thank the reviewer for this critical comment. We agree that attributing the increase in the OC fraction specifically to “enhanced photochemical reactions and gas-particle conversion” lacked direct evidence and was speculative. Therefore, we have removed this statement from the manuscript. In the revised version, we have retained the description of “efficient mixing of dust with anthropogenic carbonaceous aerosols during long-range transport,

coupled with more vigorous secondary formation processes in the free troposphere.” We believe this statement is a reasonable interpretation of the observed phenomena, as it is based on actual observational characteristics such as the doubling of OC concentration at the summit and is supported by relevant literature. Furthermore, the concentration peak of the biomass burning tracer C_9 did not coincide with the rise in OC, indicating a limited contribution from primary emissions. This evidence further supports the conclusion that transported and secondarily processed aerosols are the main factor responsible for the observed changes in OC.

6. 246: You ascribe higher SO_4^{2-} mainly to heterogeneous reactions on dust particle surfaces. An alternative pathway—direct contribution from dust-borne sulfate species such as $CaSO_4$ —receives insufficient consideration in the text. The current discussion does not adequately address this alternative or reconcile it with the observations.

Response: Thank you for this constructive suggestion. We have revised the discussion to incorporate the potential contribution of dust-borne sulfate species. The updated text in Line 246 now reads as follows:

Lines 301-306: SO_4^{2-} concentrations also increased at both sites (foot: 5.8 to 10.0 $\mu g\ m^{-3}$; top: 3.8 to 8.7 $\mu g\ m^{-3}$). This increase can be attributed to both the release of inherent sulfate species in dust (e.g., $CaSO_4$) (Wu et al., 2012) and heterogeneous reactions on dust particle surfaces, where transition metals such as Fe (III) and Mn (II) catalyze the conversion of SO_2 to SO_4^{2-} (Harris et al., 2013; Myriokefalitakis et al., 2022).

7. 307–310 “The C_5 at the foot likely mainly comes from local emission, 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. The text does not present the necessary observational support and data analysis to substantiate this source attribution.

Response: We thank the reviewer for this critical comment. We agree that the original statement regarding the sources of C_5 was speculative and lacked sufficient supporting evidence. Following the reviewer's suggestion, we have removed the speculative attribution. The revised text now focuses on the objective observational fact by stating: “This difference in modal structure suggests that different atmospheric processes govern the behavior of C_5 at different altitudes.”

8. 332–337 — Coarse vs. fine C_2 and the nitrate-coating hypothesis. The manuscript reports a shift from fine- to coarse-mode C_2 during dust events and links this to nitrate coatings formed via $CaCO_3 + HNO_3 \rightarrow Ca(NO_3)_2$. However, size-resolved evidence for Ca^{2+} and NO_3^- is not presented in the manuscript, so the causal link to a water-containing nitrate coating is not demonstrated. Relevant literature (e.g., W. J. Li & L. Y. Shao, *ACP*, 2009; Zhi et al., *ES&T*, 2025) that discusses the role of Ca-rich coatings during dust aging should be integrated into the discussion.

Response: We thank the reviewer for this suggestion. As recommended, we have integrated the relevant literature (Li and Shao, 2009; Zhi et al., 2025) and added our size-resolved ion measurements (Fig. S5) to support the discussion on nitrate coatings. The revised text now includes this evidence and clarifies the association between coarse-mode C_2 and $Ca(NO_3)_2$ formation during dust events.

Lines 387-411: The dust transport process impacted the size distributions of diacids in aerosols (Fig. 5). At the foot of the mountain, the C_2 concentration in fine particles ($\leq 2.1 \mu\text{m}$) decreased from 8662 ng m^{-3} to 4880 ng m^{-3} (a reduction of 43.7%) during dust events, while the concentration in coarse particle ($> 2.1 \mu\text{m}$) 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 C_2 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 attributed to the formation of $\text{Ca}(\text{NO}_3)_2$ coatings resulting from the reaction between calcium carbonate and NO_3^- during dust aging (Li and Shao, 2009; Zhi et al., 2025). These hygroscopic coatings create favorable conditions for the adsorption and oxidation of gaseous organic compounds, thereby promoting the formation of SOA on the surfaces of coarse particles. Research by Li et al. (2025) further confirms that aqSOA formed on dust surfaces can effectively enhance SOA production and drive a transition in the size distribution from the submicron to the supermicron range, which is highly consistent with the observational results of this study. Size-segregated ion data (Fig. S5) provide direct evidence for the above mechanism. Ca^{2+} was primarily present in the coarse mode ($3.3\text{--}5.8 \mu\text{m}$), while during dust events, NO_3^- at the top migrated from the fine mode ($0.4\text{--}1.1 \mu\text{m}$) to the coarse mode ($3.3\text{--}5.8 \mu\text{m}$) and coexisted with Ca^{2+} in the same size range, strongly supporting the formation of $\text{Ca}(\text{NO}_3)_2$ coatings on dust particle surfaces. In contrast, at the foot of the mountain, although the concentration of NO_3^- decreased, it remained predominantly in the fine mode. This spatial difference may stem from more thorough aging and reactions of aerosols at the top due to longer transport times. Meanwhile, the foot is influenced by local pollution, resulting in higher background NO_3^- concentrations and competitive reactions with components such as SO_4^{2-} , which may collectively delay the distinct shift of NO_3^- to the coarse mode.

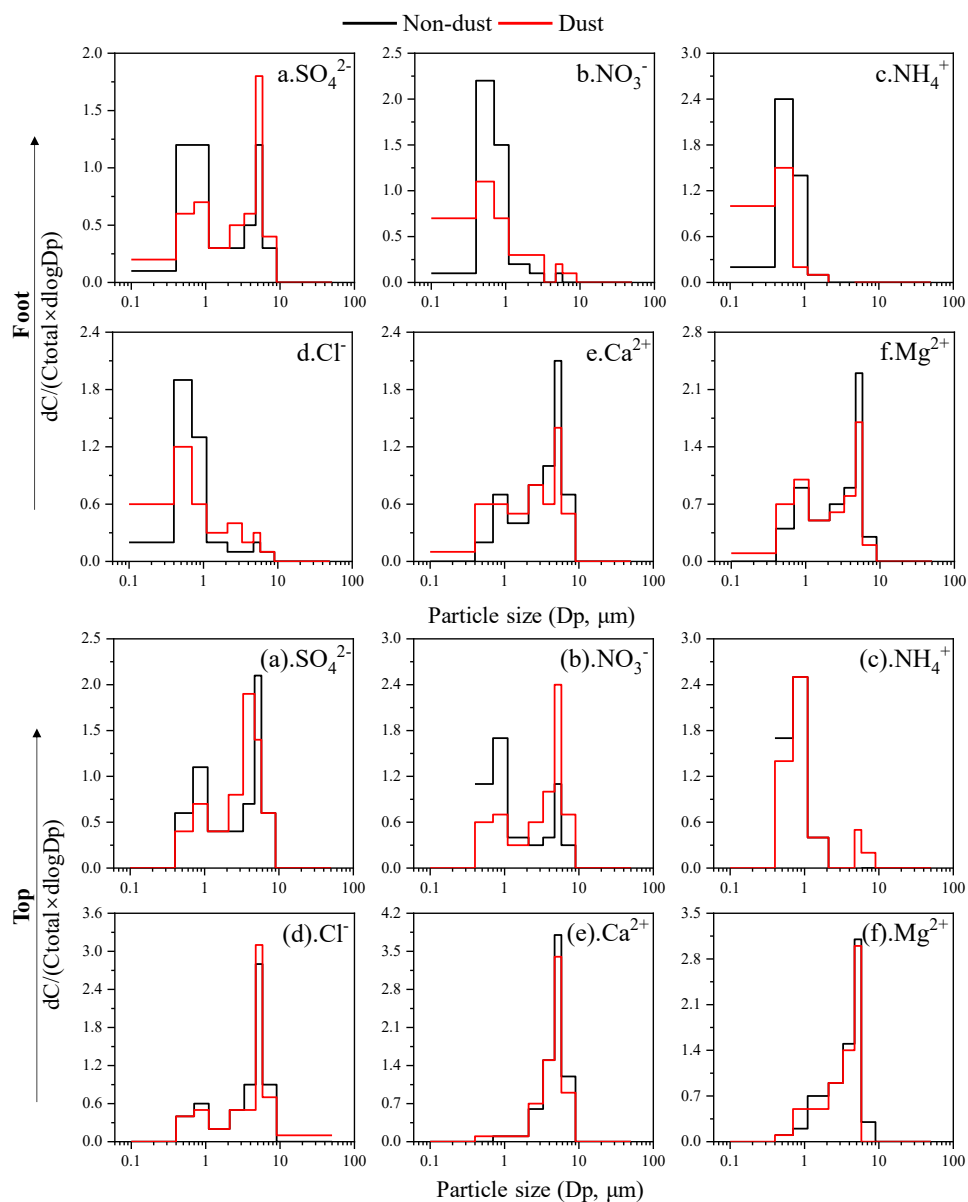


Figure S5 Size distribution of water-soluble inorganic ions at the foot and top of Mount Hua during non-dust and dust periods

English polishing and wording precision

9. 49: “secondary substances” → “secondary aerosols”.

Response: “secondary substances” has been replaced with “secondary aerosols”.

10. 75: “photocatalytic reactions” → “photochemical reactions”.

Response: The original phrasing has been eliminated as part of a broader revision of the introduction.

11. 163: “The contribution ratios of C₉ at the two sites were 8.5% and 3.2%, respectively” → Specify “contribution to what?” (e.g., total diacids, total organic acids, or OC). Define the denominator.

Response: We thank the reviewer for this important clarification. The sentence has been revised to: “Consequently, the contribution of C₉ to total diacids was substantially greater at the foot (8.5%) than at the top (3.2%) (Fig. 2b)”.

12. 233: Avoid “modest increase.” Provide exact statistics and confidence intervals; define “modest” quantitatively.

Response: This is a valid point. We have removed the subjective term “modest” and replaced it with the exact quantitative data: “...increased from 2718 ng m⁻³ to 2843 ng m⁻³ (an increase of 4.6%)”.

13. 255: “a decrease of 59%” → “decrease by 59%”.

Response: “a decrease of 59%” has been corrected to “decrease by 59%”.

14. 305: “single peak” → “unimodal distribution”.

Response: “single peak” has been replaced with the more precise “unimodal distribution”.

15. 331: “altered the particle size distribution characteristics” → “impacted the size distributions of diacids in aerosols”.

Response: The text has been revised to “impacted the size distributions of diacids in aerosols”.

References

- Carlton, A. G., Turpin, B. J., Lim, H. J., Altieri, K. E., and Seitzinger, S.: Link between isoprene and secondary organic aerosol (SOA): Pyruvic acid oxidation yields low volatility organic acids in clouds, *Geophys. Res. Lett.*, 33, L06822, <https://doi.org/10.1029/2005GL025374>, 2006.
- Falkovich, A. H., and Schkolnik, G.: Adsorption of organic compounds pertinent to urban environments onto mineral dust particles. *J. Geophys. Res. Atmos.*, 109, D02208. <https://doi.org/10.1029/2003JD003919>, 2004.
- Fu, P. Q., Kawamura K., Kanaya, Y., and Wang, Z. F.: Contributions of biogenic volatile organic compounds to the formation of secondary organic aerosols over Mt. Tai, Central East China. *Atmos. Environ.*, 44, 4817–4826. <https://doi.org/10.1016/j.atmosenv.2010.08.040>, 2010.
- Fu, T. M., Jacob, D. J., Wittrock, F., Burrows, J. P., Vrekoussis, M., and Henze, D. K.: Global budgets of atmospheric glyoxal and methylglyoxal, and implications for formation of secondary organic aerosols. *J. Geophys. Res.* 113. D15303, <https://doi.org/10.1029/2007JD009505>, 2008.
- Lamkaddam, H., Dommen, J., Ranjithkumar, A., Gordon, H., Wehrle, G., Krechmer, J., Majluf, F., Salionov, D., Schmale, J., Bjelić, S., Carslaw, K. S., Haddad, I. E., and Baltensperger, U.: Large contribution to secondary organic aerosol from isoprene cloud chemistry. *Sci. Adv.*, 7, eabe2952. <https://doi.org/10.1126/sciadv.abe2952>, 2021.
- Li, J. J., Wang, G. H., Zhou, B. H., Cheng, C. L., Cao, J. J., Shen, Z. X., and An, Z. S.: Airborne particulate organics at the summit (2060 m, a.s.l.) of Mt. Hua in central China during winter:

Implications for biofuel and coal combustion. *Atmos. Res.*, 106, 108–119, <https://doi.org/10.1016/j.atmosres.2011.11.012>, 2012.

Li, W. J., and Shao L. Y.: Observation of nitrate coatings on atmospheric mineral dust particles. *Atmos. Chem. Phys.*, 9(6), 1863–1871. <https://doi.org/10.5194/acp-10-10521-2010>, 2009.

Lim, H. J., Carlton, A. G., and Turpin, B. J.: Isoprene forms secondary organic aerosol through cloud processing: model simulations. *Environ. Sci. Technol.* 39(12), 4441–4446. <https://doi.org/10.1021/es048039h>, 2005.

Meng, J. J., Wang, G. H., Li, J. J., Cheng, C. L., Ren, Y. Q., Huang, Y., Cheng, Y. B., Cao, J. J., and Zhang, T.: Seasonal characteristics of oxalic acid and related SOA in the free troposphere of Mt. Hua, central China: Implications for sources and for formation mechanisms, *Sci. Total Environ.*, 493, 1088–1097, <https://doi.org/10.1016/j.scitotenv.2014.04.086>, 2014.

Warneck, P.: In-cloud chemistry opens pathway to the formation of oxalic acid in the marine atmosphere. *Atmos. Environ.* 37(17), 2423–2427, [https://doi.org/10.1016/S1352-2310\(03\)00136-5](https://doi.org/10.1016/S1352-2310(03)00136-5), 2003.

Wu, F., Zhang, D. Z., Cao, J. J., Xu, H. M., and An, Z. S.: Soil-derived sulfate in atmospheric dust particles at Taklimakan desert, *Geophys. Res. Lett.*, 39(24), L24803, <https://doi.org/10.1029/2012GL054406>, 2012.

Xu, W. Y., Kuang, Y., Liang, L. L., He, Y., Cheng, H. B., Bian, Y. X., Tao, J. C., Zhang, G., Zhao, P. S., Ma, N., Zhao, H. R., Zhou, G. S., Su, H., Cheng, Y. F., Xu, X. B., Shao, M., and Sun, Y. L.: Dust-dominated coarse particles as a medium for rapid secondary organic and inorganic aerosol formation in highly polluted air. *Environ. Sci. Technol.*, 54(24):15710–15721. <https://doi.org/10.1021/acs.est.0c07243>, 2020.

Zhang, G. H., Lin, Q. H., Peng, L., Yang, Y. X., Jiang, F., Liu, F. X., Song, W., Chen, D. H., Cai, Z., Bi, X. H., Miller, M., Tang, M. J., Huang, W. L., Wang, X. M., Peng, P. A., and Sheng, G. Y.: Oxalate formation enhanced by Fe-containing particles and environmental implications. *Environ. Sci. Technol.*, 53(3):1269–1277. <https://doi.org/10.1021/acs.est.8b05280>, 2019.

Zhang, R. F., Gen, M. S., Liang, Z. C., Li, Y. J., and Chan, C. K.: Photochemical reactions of glyoxal during particulate ammonium nitrate photolysis: brown carbon formation, enhanced glyoxal decay, and organic phase formation. *Environ. Sci. Technol.*, 56(3), 1605–1614. <https://doi.org/10.1021/acs.est.1c07211>, 2022.

Zhi, M. K., Wang, G. C., Xu, L., Li, K. I., Nie, W., Niu, H. Y., Shao, L. Y., Liu, Z. R., Yi, Z. W., Wang, Y. T., Shi, Z. B., Ito, A., Zhai, S. X., and Li, W. J.: How acid iron dissolution in aged dust particles responds to the buffering capacity of carbonate minerals during Asian dust storms. *Environ. Sci. Technol.*, 59(12): 6167–6178. <https://doi.org/10.1021/acs.est.4c12370>, 2025.