Dear Editor,

We have now revised our manuscript (MS No.: egusphere-2025-3094) according to your final revision request. The abstract and conclusions have been carefully updated in an effort to align with the current ACP style guide. Specifically, we have rewritten the beginning of the abstract to include a brief introduction to the topic for the benefit of non-specialist readers.

Thank you for your guidance and support throughout the review process. Should you have any further questions, please do not hesitate to contact me at lijj@ieecas.cn.

Best regards,
Jianjun Li
On behalf of all co-authors
Oct 10, 2025

Editor comments:

Public justification (visible to the public if the article is accepted and published):

I have read the reviews and the authors' responses and I am satisfied that the reviewers' comments have been adequately addressed and that the manuscript can go to publication. However before it does, I would ask that the abstract and conclusions be revised so that they conform better to ACP's current style guides: https://www.atmospheric-chemistry-and-physics.net/policies/guidelines_for_authors.html. In particular, I would ask that the abstract starts with a brief introduction to the topic and scientific need for the benefit of a nonspecialist reader.

Response: We thank the editor for this final guidance. We have thoroughly revised the abstract and conclusions sections to comply with the ACP style guide. Specifically, as requested, we have rewritten the opening of the abstract to provide a clear introduction to the topic and its scientific significance for a nonspecialist audience.

Abstract: Dust transport significantly affects downwind aerosol formation and regional climate, yet the evolutionary mechanisms of SOA during this process remain poorly understood. Here, we conducted vertical observations of $PM_{2.5}$ and size-segregated aerosols at the foot and top of Mount Hua, focusing on C_2 formation and its $\delta^{13}C$ signatures influenced by dust transport. Under non-dust conditions, $PM_{2.5}$ and diacid concentrations at the foot were 4.5 and 2.1 times higher than those at the top, indicating stronger anthropogenic influence at lower elevations. Aerosols at the top revealed enhanced photochemical aging, with higher C_2/C_4 (5.84 vs. 4.74), C_3/C_4 ratios (1.04 vs. 0.56), and more positive $\delta^{13}C$ values (-21.5‰ vs. -27.6‰). The positive correlation of C_2 with ALWC and its consistent size distribution with precursors confirm aqueous-phase oxidation as the dominant formation pathway. During dust events, although $PM_{2.5}$ concentrations increased, C_2 concentrations in $PM_{2.5}$ decreased by 59% at the foot and 25% at the top. Concurrently, the $\delta^{13}C$ values of C_2 showed a positive shift, particularly at the top (from -21.5‰ to -13.2‰), suggesting that alkaline dust catalyzes the formation of ^{13}C-enriched oxalate. Size-segregated data revealed a shift of C_2 from the fine to the coarse mode, with the coarse-to-fine ratio increasing from 0.3–0.4 to 0.6–1.1. These findings demonstrate that under dust influence, the primary formation pathway of C_2 shifts from aqueous-phase oxidation in fine particles to heterogeneous reactions on coarse-particle surfaces. Moreover, this shift is accompanied by a positive shift in the $\delta^{13}C$ signature of C_2 and is more pronounced at higher altitudes.

Conclusions: Based on synchronous observations of $PM_{2.5}$ and size-segregated aerosols from surface to mountain sites, this study reveals the influence of dust transport on the formation pathways and $\delta^{13}C$ signature of C_2 at different altitudes. The results indicate that during non-dust periods, C_2 is primarily generated through aqueous-phase oxidation in fine particles, whereas during dust periods, its formation shifts to heterogeneous chemical reactions on coarse-particle surfaces (Fig. 8).

At high-altitude sites, the influence of dust on aerosols was particularly pronounced, as evidenced by enhanced aerosol aging indicators (C_2/C_4 ratio increased to 7.75), enriched $\delta^{13}C$ (+8.3‰ compared to non-dust periods), and a distinct shift of C_2 from the fine to the coarse mode. This redistribution resulted in a higher concentration of C_2 in coarse particles (2301 ng

m^{-3}) than in fine particles (2161 ng m^{-3}), producing a coarse-to-fine particle ratio of 1.1. The shift in particle size distribution was closely associated with the formation of $Ca(NO_3)_2$ coatings during dust aging. These hygroscopic coatings provide active interfaces for the adsorption and oxidation of gaseous precursors, thereby promoting the formation of C_2 and other SOA on coarse particle surfaces. The migration of NO_3^- from the fine mode (0.4–1.1 μm) to the coarse mode (3.3–5.8 μm) in the presence of Ca^{2+} provides direct evidence for the formation of such coatings. The pronounced enrichment of $\delta^{13}C$ further supports this reaction pathway, which is attributed to the synergistic effects of surface-catalyzed oxidation (favoring ^{13}C retention) and metal-oxalate complexation. In contrast, aerosols at the low-altitude site were dominated by local anthropogenic emissions, exhibiting a lower degree of aging, as reflected by higher C_2 concentrations (674 ± 528 ng m^{-3}) and lower $\delta^{13}C$ values (-27.6‰). The higher background concentration of NO_3^- and competitive reactions with components such as SO_4^{2-} collectively delayed the shift of NO_3^- to the coarse mode.

This study clarifies how dust alters the formation pathways, particle-size distribution, and $\delta^{13}C$ signature of C_2, highlighting the significant altitudinal variation of these effects. Our findings provide critical insights into the altitude-dependent transformation of SOA during dust transport, thereby enhancing the understanding of mountain atmospheric chemistry and regional climate effects. Limited by field observations, the microkinetic parameters of the aforementioned surface reactions remain unquantified. Future work should integrate laboratory experiments with model simulations to fully elucidate the microscopic mechanisms and regional climate impacts of this heterogeneous process.