******Response to referee comments from Yaping Shao, 28 Dec 2025******

General comments (overall quality):

This study analyzes how calcium-containing dust particles from Asian deserts react with water, finding that a majority of particles lose soluble components when exposed to moisture. Particle size, mineral composition, and surface coatings determine the dissolution rate of calcium. These findings are important for modeling how mineral dust neutralizes atmospheric acidity and mitigates ocean acidification.

The impact of mineral dust on the climate system depends on its size-resolved mineral composition and mixing state. The authors identify saltation bombardment as a key mechanism for enhancing dust emissions and enriching soluble calcium. Using laboratory dust abrasion, water dialysis, and electron microscopy, this study tracks the evolution of calcium-containing dust at the single-particle level, estimating particle emissions and mixing states, thereby providing highly valuable data.

By analyzing 4 soil samples collected from the Taklimakan and Gobi deserts, the authors report that 57–88% of calcium-containing particles released soluble calcium. Calcite often exists as nanometer-scale coatings on larger particles, which likely accelerates dissolution and increases dust hygroscopicity, which implies faster alkalinity release from Asian dust than previously thought.

I find the paper highly interesting and generally well written. The combination of techniques for analyzing calcium-containing particles, both pre- and post-dialysis, at the individual particle level is impressive. The experiments are well executed, carefully examined and analyzed.

I do have a few suggestions to improve the presentation of the results. I would ask the authors to keep in mind in their presentation how their data can be more directly used for modeling purposes. Additionally, I hope to see more explanations of how laboratory-based results relate to natural processes would be valuable.

Response:

We thank the reviewer for the positive and constructive comments. Accordingly, we have enhanced the validation of our analytical methodology and refined the understanding of water-soluble calcium abundance in mineral dust particles. The principal revision is a re-interpretation of the dialysis experiments, which replaces the previous speculative discussion on acid neutralization with a more robust, evidence-based explanation of dissolution behavior. Our point-by-point responses are provided below in italics.

Specific comments (scientific questions/issues):

I realize that dust aerosols were generated from the samples using a resuspension chamber that simulates saltation bombardment. Are there comparisons of size distribution and chemical composition with field or wind-tunnel experiments, say equivalent to friction velocity of 0.54 m s^{-1}? If data exist, it would be interesting to provide such a comparison. Otherwise, some discussions of chamber limitation would help.

Response:

*Our published data show that the aerosol size distribution (Wu et al., 2023) and water-soluble ion composition (Wu et al., 2022) are comparable to field observations. The resuspension chamber's operational parameter (0.54 m s^{-1} equivalent friction velocity) is derived from Etyemezian et al. (2007). The dust generated under this operation condition demonstrates field-relevant characteristics (**Lines 106-111, Page 4**).*

I understand that dust samples were analyzed using X-ray spectroscopy for size, morphology, and elemental composition individually. The same sample was subjected to water dialysis to remove soluble components. The identical particle set was reanalyzed post-dialysis for comparison with pre-dialysis. This enables estimates of water-soluble calcium-containing particles and soluble calcium mass. I can see how these values can be used to calculate emission fluxes of soluble calcium particles, but cannot relate these values to what happens in nature.

Response:

*We clarify that the dialysis experiment (**Fig. S2**) aimed not to simulate atmospheric processing, but to quantify water-soluble calcium and probe its mixing state within dust particles. The key finding, loss of Ca signal with preserved particle morphology, indicates that soluble Ca exists primarily as surface coatings.*

*To address the role of thin water films more analogous to atmospheric humidification, we conducted a complementary water-vapor exposure experiment (**Fig. S8**). Under these conditions, soluble coatings recrystallized upon drying rather than being removed (**Figs. S9** and **S10**), highlighting that wetting-drying cycles may redistribute material.*

We fully agree that dialysis conditions (bulk water, neutral pH) differ from atmospheric aerosol water (thin films, often acidic). From this point of view, our experiment likely provides a conservative estimate of coating removal. The demonstrated existence of soluble Ca coatings remains directly relevant to particle-level properties such as cloud condensation activity. The presence of such thin coatings of Ca on fine particles may *enhance mass transfer rate (Batchelor-McAuley et al., 2022), a key factor in atmospheric processing kinetics. Thus, the experiment provides single-particle-resolved evidence that informs models of such facilitated atmospheric dissolution pathways.*

*The manuscript has been revised to clarify this intent and interpretation (**Lines 321-324, Page 14; Lines 373-376, Page 15**).*

Equations (3) and (4) should be written better, e.g., Eq. (4) can be simplied to

$$F_m = m_{ca}\, F_n$$

with m_{ca} being the average Ca mass per particle. Actually, I think it is better to estimate first dust-mass flux and then a Ca-mass flux, which is Ca mixing ratio times dust-mass flux. It would be much clearer and easy to use for future studies.

Response:

*According to the comment, a combined parameter (**Eq. 3**) was introduced to simplify all flux equations (see **Lines 157-177, Page 6**). In addition, equations for the total dust number and mass emission fluxes (**Eqs. 4** and **5**) have been added to detail the estimation methodology. For clarity, the total mineral dust aerosol emission fluxes from the Supplementary Table S5 of the first submission have been incorporated into **Table 1** of the revised manuscript, highlighting the abundance of water-soluble Ca components within the total dust particles (see **Lines 237-239, Page 8**).*

Figure 1 and 2: again, I would be very interested to see the mixing ratio, because this is the quantity one would use for global dust/Ca modelling.

Response:

*While the automated CCSEM/EDX analysis efficiently identifies Ca-containing particles and quantifies their elemental mass, determining the mixing state requires manually relocating each particle and verifying its EDX spectral mapping—a highly labor-intensive process. Therefore, to maximize the analytical impact within practical constraints, we focused on the mineral groups that exhibited the most significant dissolution. Our data indicate that Ca-O-rich (calcite-like) and Ca-S-containing (gypsum-like) particles experienced the sharpest decline in abundance after dialysis (**Lines 280-286, Page 11**), highlighting composition-dependent solubility. We manually examined the mixing states of all particles within these two key groups. Although this approach does not yield a complete inventory for all mineral types, it provides a robust mixing-state profile for the most soluble, and thus likely most climatically and biogeochemically active, fractions of calcium in* *mineral dust, which is the quantitative information most critical for modeling water-soluble calcium emissions.*

Technical corrections (technical corrections, typing errors, etc.):

Line 31:"Global dust emissions are estimated at 2000 ± 400 Tg yr–1 (Kok et al., 2020), with Asia contributing approximately 25–30 % of this total (Kok et al., 2021)." Note the debate on the estimates and uncertainty involved.

Response:

*In the revision, we have expanded our discussion in the introduction (**Lines 27-28, Page 2**) to acknowledge the range of published estimates (Kok et al., 2023; Zhao et al., 2023).*

Line 43: "below the direct entrainment threshold". Maybe you mean below the direct entrainment threshold of fine dust particles.

Response:

*Thank you for this precise suggestion. The text has been amended accordingly to "below the direct entrainment threshold for fine dust particles" in the revision (**Lines 41, Page 2**).*

Citation of papers from Chinese scientists. I find it is overall confusing in citing papers of Chinese scientists, e.g., Wang Z and Wang Y, but sometimes with no abbreviation of first name, e.g., Zhao. It may be time to think a way how to standardize the citations. Taking the first name does seem to make sense. This should be discussed with the publisher.

Response:

*To distinguish between the two authors with the same last name and publication year, we have now explicitly cited them as (Wang Y. et al., 2012) and (Wang X. et al., 2012). These changes have been made in the text (**Lines 55, Page 2; Line 93, Page 4; Line 313, Page 14**).*

References

Batchelor-McAuley, C., Yang, M., Rickaby, R. E., and Compton, R. G.: Calcium carbonate *dissolution from the laboratory to the ocean: Kinetics and mechanism, Chem. Eur. J., 28, e202202290, https://doi.org/10.1002/chem.202202290, 2022.*

Kok, J. F., Storelvmo, T., Karydis, V. A., Adebiyi, A. A., Mahowald, N. M., Evan, A. T., He, C., and Leung, D. M.: Mineral dust aerosol impacts on global climate and climate change. Nature Reviews Earth & Environment, 4(2), 71-86, https://doi.org/10.1038/s43017-022-00379-5, 2023.

Wu, F., Cheng, Y., Hu, T., Song, N., Zhang, F., Shi, Z., Hang Ho, S. S., Cao, J., and Zhang, D.: Saltation–sandblasting processes driving enrichment of water-soluble salts in mineral dust, Environ. Sci. Technol. Lett., 9, 921-928, https://doi.org/10.1021/acs.estlett.2c00652, 2022.

Wu, F., Song, N., Hu, T., Ho, S. S. H., Cao, J., and Zhang, D.: Surrogate atmospheric dust particles generated from dune soils in laboratory: Comparison with field measurement, Particuology, 72, 29-36, https://doi.org/10.1016/j.partic.2022.02.007, 2023.

Wang, X., Hua, T., Zhang, C., Lang, L., and Wang, H.: Aeolian salts in Gobi deserts of the western region of Inner Mongolia: Gone with the dust aerosols, Atmos. Res., 118, 1-9, https://doi.org/10.1016/j.atmosres.2012.06.003, 2012.

Wang, Y., Yu, W., Pan, Y., and Wu, D.: Acid neutralization of precipitation in Northern China. Journal of the Air & Waste Management Association, 62(2), 204-211, https://doi.org/10.1080/10473289.2011.640761, 2012.

Zhao, Y., Yue, X., Cao, Y., Zhu, J., Tian, C., Zhou, H., Chen, Y., Hu, Y., Fu, W., and Zhao, X.: Multi-model ensemble projection of the global dust cycle by the end of 21st century using the Coupled Model Intercomparison Project version 6 data. Atmospheric Chemistry and Physics, 23(13), 7823-7838, https://doi.org/10.5194/acp-23-7823-2023, 2023.

********Response to referee comments from Anonymous Referee #2, 11 Jan 2026********

Review of Hu et al.: Abundant water-soluble calcium coatings on fine Asian dust particles

The authors present a single-particle-scale study showing that calcium in freshly emitted Asian mineral dust is more soluble and reactive than commonly assumed. Using laboratory-generated dust from the Taklimakan and Gobi deserts and CCSEM analysis before and after water dialysis, they show that a significant fraction of Ca-containing particles carries water-soluble calcium, which the authors state occurs largely as nanometer-scale calcite and gypsum coatings on insoluble mineral cores. These coatings are claimed to dissolve rapidly without significantly altering particle size distributions, accounting for 20-40 % of the Ca mass in dust particles. The results could imply that Asian dust provides a faster and more effective source of alkalinity for atmospheric acid neutralization and ocean buffering than represented in many current models.

I think that the topic is interesting and might indeed have relevance for modeling the dust cycle in particular for East Asia, where the Ca fraction in the dust in particularly high.

It seems, however, that there are some additional explanations, clarifications and corrections or modifications of the conclusions required.

Response:

We extend our sincere thanks to the reviewer for their detailed feedback. The comments, suggestions and questions have helped us to significantly strengthen the quality of the manuscript. Below are our responses to each comment, presented in italics.

Major concerns:

- Low sample number – the authors base their conclusions on 4 samples from different deserts covering more than 500,000 km^2. As deserts can be highly variable on a sub-km scale, the precise location of sampling might be more relevant for the results than the desert where the sample was taken. Also, the sampling itself is largely unexplained.

Response:

We acknowledge that four samples cannot statistically represent an entire desert spanning hundreds of thousands of square kilometers. The aim of this process-oriented *study is not to produce a spatially resolved map of dust solubility across the region, which would require a far denser, grid-based sampling design. Instead, it is to investigate, at the single-particle level, the fundamental physicochemical properties (e.g., composition, morphology, and mixing state) that govern dust dissolution behavior.*

Accordingly, our sampling strategy was designed to capture typical surface soils within the desert system, prioritizing representativeness of key dust sources over exhaustive spatial coverage. We therefore focused on the two dominant and geochemically distinct landforms: sand dunes and gravel gobi. This selection was made because of their fundamental differences in sediment history, wind sorting, and surface aging—factors critically relevant to dust emission potential. It is noteworthy that both sand dunes and gobi soils have been identified as major dust sources in the broader Taklimakan-Gobi arid regions encompassing our study area (e.g., Laurent et al., 2005; Wang et al., 2012a, 2012b; Sweeney et al., 2016; Zou et al., 2018).

*The limited number of geographic sampling sites is compensated for by the high statistical robustness achieved at the particle-population level. Our analysis of over 10,000 individual particles per sample provides an empirically grounded, single-particle-level statistical basis for our conclusions, elucidating trends in the aqueous processing behavior of distinct particle types. The general physicochemical properties identified (e.g., the abundance and internal mixing state of water-soluble calcium) are likely transferable to other arid regions in East Asia with analogous mineralogical sources and climatic conditions. To address the reviewer's concern directly, we have added a justification for the sampling strategy in the Methodology section (**Lines 92–94, Page 4**), explicitly stating the rationale for selecting these two landforms as representative end-members. Further details of the sampling procedure are provided in **Lines 101–104, Page 4**, and are visually supported by field photographs in **Fig. S1**.*

- Unclear methodology description – there are some important details missing from the method description, and there are also some apparent mistakes in the classification tables.

Response:

*The manuscript has been updated to clarify the methodology, and the error in the classification table has been rectified accordingly (**Lines 92-94, 101-104, Page 4; Lines 183-189, 200-209, Page 7**, and **Table S2**).*

- Images shown don't support the mentioned observations – coatings are frequently mentioned but never shown. Also, a volume change of the particles is presented in the data but never shown in an image. If the analysis is really based on these images, there seem to be logical gaps regarding the volume and mass determination.

Response:

The inference that water-soluble calcium components exist as surface coatings was drawn from dialysis observations (experimental setup shown in ***Fig. S2****), which showed a loss of the calcium signal with no concurrent change in particle morphology (****Fig. 6****). Following the dialysis experiment, a separate experiment was performed in which dust particles were exposed to saturated water vapor for 24 h (see also experimental setup in* ***Fig. S8****) to directly probe the process involving thin water films on surfaces. In contrast to dialysis, this exposure did not remove soluble coatings. Instead, during subsequent drying under SEM vacuum, these components recrystallized on the surface. This process modified the particle morphology while preserving the calcium signal, as evidenced by SEM-EDS analysis (****Figs. S9*** *and* ***S10****). This sharp contrast confirms the presence of water-soluble calcium coatings and differentiates their fate: they are physically removed during dialysis but were retained and underwent morphological change during water-vapor exposure.*

*The single-particle data (including volume) presented in the supplementary tables of the paper represent the physicochemical characteristics of all particles within the same area before and after dialysis. Notably, the listed particles are not necessarily matched one-to-one according to the table sequence. After dialysis, approximately 2.8%–4.0% of particles (most likely chloride or other water-soluble minerals not mixed with insoluble components) totally disappeared. However, the particle number distributions by size (****Fig. 3****) and by shape (****Figs. S4–S6****) did not change significantly; therefore, the volume distribution based on the ellipsoid assumption also showed no pronounced variation. Furthermore, images of Ca-O-rich (****Fig. 6****) and Ca-S-containing (****Fig. S7****) particles before and after dialysis process reveal that the projective outlines (and thus the volumes) of these particles remained largely unchanged, which suggests the presences of water-soluble calcium coating on the order of 10 to 100 nanometers (****Fig. 6*** *and* ***Fig. S7****).*

- General implications – Conclusions based on the lab-generated dust are not necessarily valid for ambient dust. Also, it remains unclear whether the observed dissolution state is a final one. At least estimates for the dissolution kinetics or a time series of dissolution should be done. The pH of the solvent significantly impacts the kinetics but is higher than what one would expect for cloud water. As a result, it is not clear, how relevant the results are for the atmosphere.

Response:

*The dialysis experiment (**Fig. S2**) aimed to quantify water-soluble calcium and probe its mixing state, rather than to directly simulate atmospheric processing. Here, the loss of the Ca signal alongside unchanged morphology revealed that soluble Ca predominantly forms surface coatings. In contrast, the complementary water-vapor exposure experiment (**Fig. S8**) better approximates atmospheric humidification. In this case, soluble coatings recrystallized upon drying (**Figs. S9**, **S10**), implying that wetting-drying cycles may redistribute rather than remove such material.*

*To investigate the temporal evolution of dissolution, a double-dialysis experiment was conducted on a separate dust sample whose abundance of Ca-containing particles was within the range observed in the four main samples. The results indicated that the differences in the proportions of Ca-containing particles between the first and second dialysis fell below the standard deviations of this particle type prior to dialysis (**Table S6**). This demonstrates that a single 2-hour dialysis process is sufficient to effectively remove water-soluble Ca components from the dust particles. We have added this clarification on the dissolution time series to the revised manuscript (**Lines 205-207, Page** 7).*

*We acknowledge that our water dialysis conditions (bulk water, neutral pH) differ from those of atmospheric aerosol water (thin films, often acidic). To simulate an acidic environment, an additional dialysis experiment at pH = 5.1 ± 0.1 was therefore conducted on the same separate dust sample after the double-dialysis treatment. As shown in **Table S6**, more Ca-containing particles were removed under acidic conditions. This finding confirms that our neutral-water dialysis likely represents a conservative scenario, estimating minimal removal. Nonetheless, confirming the presence of soluble Ca coatings is directly relevant to atmospheric processes: such coatings on fine particles can enhance interfacial mass transfer rates (Batchelor-McAuley et al., 2022), a key kinetic factor in atmospheric processing. Therefore, our study provides single-particle-specific evidence for surface coatings that may facilitate atmospheric dissolution pathways. We have added this clarification on the acid dialysis to the revised manuscript (**Lines 207-209, Page** 7).*

Details:

L99: How was the location of sampling selected? Please give more details on the sampling, as it might have severe impact on the results (e.g. compare the compositional data for different sediment types here doi: 10.5194/acp-23-15815-2023).

Response:

Our sampling strategy was aimed to capture representative surface soils within two major dust source regions in northern and northwestern China, the Taklimakan Desert *and the Gobi deserts. Specifically, we selected two geochemically distinct landforms that are known to be dominant contributors to dust emissions: sandy dunes and gravel gobi. Detailed descriptions of the sampling locations have been included in the revised manuscript (**Lines 92–94 and 101–104, Page 4**). Photographs taken during field sampling are also provided in **Fig. S1**.*

L109: The 0.2 μm is the pore size, porosity would be e.g. a fraction of pores vs. solid volume.

Response:

*The description has been corrected to "pore size of 0.2 μm" (**Line 112, Page 4**).*

L117: How thick was the coating? From the images in Figure 6, it seems to be quite thick.

Response:

*While the diameters of the dust particles ranged from several hundred nanometers to tens of micrometers, the water-soluble Ca-rich coatings remained notably thin. By calibrating against the 200 nm and 500 nm scale bars in the high-resolution images (**Fig. 6** and **Fig. S7**), the thickness of these Ca-O-rich and Ca-S-containing coatings is estimated to be on the order of 10 to 100 nanometers.*

L120: two … spectrometers

Response:

*Yes, our CCSEM system features a field-emission gun SEM and dual EDX detectors. This configuration enables robust signal detection for fine particles down to 200 nm. We have clarified the description of this instrumentation in the revised manuscript (**Line 123-124, Page 5**).*

L122: parameter… compositions - check for singular / plural and make consistent.

Response:

*In the revision, the sentence has been revised to ensure grammatical consistency in singular/plural forms (**Line 125-127, Page 5**).*

L123: … provided reproducible sizing … how was the reproducibility checked?

Response:

*The reproducibility of the sizing procedure was evaluated using a PSL sphere standard (nominal diameter: 802 nm ± 6 nm). Three repeat CCSEM measurements, each analyzing over 6000 particles, gave a mean size of 796 nm ± 16 nm. We have clarified the description of this instrumentation in the revised manuscript (**Line 183-185, Page** 7).*

L123: What type of quantification was used for obtaining the elemental composition from the spectra? Was there any type of correction (ZAF or similar) applied? Was there a quantification threshold applied? The information is relevant in view of the small relative contributions used for classification. In L180 there is mentioned a ZAF-correction, but not which.

Response:

*Elemental composition was quantified based on the characteristic X-ray peak intensities, covering elements from carbon (C) to lead (Pb). Specifically, the EDX software converted these intensities into relative atomic percentages using a ZAF correction procedure (accounting for atomic number, absorption, and fluorescence effects). To ensure reliability in particle classification, a quantification threshold of 0.5 wt% was applied during the semi-quantitative analysis. These methodological details have been clarified in the revised manuscript (**Lines 186-189, Page** 7).*

L129: Figure S2 is illegible in the PDF (resolution). The referred paper of Zhang & Iwasaka used TEM grids for the analysis, here we have polycarbonate filters. Can you please provide more evidence that the particle location remained stable (e.g. some higher resolution detail images before / after).

Response:

*In revision, the following revisions have been made according to this comment: 1) The original Fig. S2 (now **Fig. S3**) has been replaced with a high-resolution version to ensure clarity. 2) To directly address the question of particle stability on polycarbonate* *filters, we have incorporated higher-resolution micrographs showing identical view fields before and after dialysis in the revised* ***Fig. S3****. These images provide clear evidence that most of the particle locations remained stable throughout the dialysis experiments, including dual water dialysis and one acid dialysis.*

Also, how do you make sure that the state you observe after dialysis is a final state, i.e. everything soluble is dissolved, and not just an arbitrary intermediate state of dissolution? You refer to the problem in L336-338.

Response:

*A double-dialysis experiment on a representative dust sample revealed that the difference in Ca-containing particle proportions between cycles fell within the pre-dialysis standard deviation (****Table S6****), showing that a single 2-hour dialysis effectively removes water-soluble Ca. This point regarding the dissolution time series has been clarified in the manuscript (****Lines 203-205, Page*** *7).*

L150: The classification criteria in table S2 require a bit more explanation. Are all elements not in the list for a certain category required to be zero or undetected? If not, there would be overlaps e.g. between the Ca-C-O category and for example the Ca-S-O category (imagine 98.4% O, 0.8% S, 0.8% Ca).

Or are the criteria evaluated from top to bottom, using the first match?

Response:

Regarding the presence of other elements: they are not required to be zero or undetected.

Regarding the classification logic: the criteria in ***Table S2*** *were applied sequentially from top to bottom using a first-match rule. This hierarchical approach ensures unambiguous and mutually exclusive classification, thereby preventing overlaps such as the example noted by the reviewer.*

Why is O included e.g. for Si-O and Al-Si-O, but not for Na-S-O?

Response:

The inclusion or omission of O in a category rule is based on its diagnostic role for identifying specific mineral phases. For silicate particles (e.g., quartz or kaolinite), identification critically relies on the co-dominance of Si and O, or Si, Al, and O. In contrast, for sulfate particles (e.g., gypsum, mirabilite, and glauberite), identification is based primarily on the presence of S coupled with Ca and/or Na; oxygen, while present, is not a differentiating factor among these sulfates and is therefore not specified in the category name.

Why is N included in Al-Si-O-Na? Why is C included in Al-Si-O-Ca?

Response:

The notations "N" in "Al-Si-O-Na" and "C" in "Al-Si-O-Ca" were indeed typographical errors. They have been revised to "Na" and "Ca", respectively, in the corrected ***Table S2****. We have thoroughly reviewed the table to ensure no similar errors remain.*

A part is given in Wt%, another in At%. For what reason?

Response:

*The combined use of wt% and at% in our classification (****Table S2****) is intentional and based on empirical mineral formulas. While the initial identification of major phases relies on the sum of weight percentages (wt%) of predominant elements, the final assignment to specific mineral categories (based on empirical formulas from mineralogical references) often requires adherence to characteristic molar ratios. Therefore, atomic percentages (at%) are incorporated into the classification rules to constrain these stoichiometric proportions, ensuring a more accurate and chemically consistent categorization.*

There are two categories with the same name (Al-Si-O-K), but slightly differing criteria.

Response:

Although EDX analysis yields identical elemental signatures (Al, Si, O, K) for both particle types, hence the shared "Al-Si-O-K" category name, they are distinguished by their characteristic Si/Al molar ratios. Particles with a ratio between 1 and 2 correspond to mica-group minerals, while those outside this range are classified as orthoclase. This distinction reflects underlying differences in their crystal structures, which EDX alone cannot resolve.

L155: How was the volume calculated, which assumptions were made?

Response:

The calculation was based on the core assumption of an ellipsoidal particle shape. For each particle, its major (D_{max}) and minor (D_{min}) axial diameters were measured, and the volume was subsequently calculated using these dimensions. The calculation was clarified in ***Lines 157-158, Page 6****.*

L160: How was the total mass of elemental Ca quantified? From the EDX spectra? Using oxide assumptions, or what?

Response:

The mass of each particle was derived from its volume and a composition-based density (using oxide reference tables). The Ca mass per particle was calculated from the particle mass and the Ca wt% (EDX data). The total elemental Ca mass was the sum of Ca mass across all identified Ca-containing particles. And the mass of water-soluble elemental Ca was calculated as the difference in total elemental Ca mass before and after dialysis. The calculation was clarified in ***Lines 157-164, Page 6****.*

L167: I assume v is the flow rate? Missing.

Response:

*Yes, "v" represents the air flow rate (5 L min^{-1}) of the aerosol sampler. We have clarified this in the revised text (****Lines 170-171, Page 6****).*

L181: If you classify particles with Ca > 0.5%, how do you use a 1.0% error margin?

Response:

We apologize for this oversight. The error margin should indeed be 0.1 wt%, and it has been corrected to this value in ***Line 189, Page*** *7 of the revised text.*

L187: They are termed 'mixed …' in the S2 table. Keep it consistent.

Response:

The terms in the text have been modified to match those used in ***Table S2****, and the changes are reflected in Lines* ***194-195, Page*** *7.*

L192: Table S3: What is the +/- variation in the table? Standard deviation?

Response:

Yes, the ± values in ***Table S3*** *are the standard deviations. We have added the abbreviation "S.D." to the table header accordingly to make this explicit.*

L217: Wouldn't one assume that the cloud water pH is generally lower than 6.4? E.g., references in doi: 10.1029/2019GL082067

Response:

*Yes, atmospheric water is often more acidic. To test this, we performed an additional dialysis at pH 5.1 ± 0.1, which removed more Ca-containing particles (****Table S6****). This confirms that our experiments with neutral water give a conservative removal estimate. The finding of soluble Ca coatings is still atmospherically relevant, as they can promote dissolution kinetics. We have added this point in the revision (****Lines 207-209, Page*** *7).*

L225: I don't understand what is meant by the decrease of the total number. What is the reference, from where the number decreased, and why is this specific to the single particle perspective (is there another in the paper?)?

Response:

Our analysis compared all particles within the identical fields of view before and after dialysis. The observed decrease in total particle count indicates the complete removal of entirely water-soluble particles, such as chlorides and pure sulfates. In contrast, for particles whose morphology persisted but lost their Ca signal, the finding points to the removal of internally mixed, water-soluble Ca coatings. Since our entire study is based on individual particle statistics without bulk measurements, the phrase "from a single-particle perspective" was redundant and has been deleted.

L227: Which elements were regarded for comparing the mobility? I could imagine that Cl under the measurement conditions is more mobile.

Response:

*The intended focus is on solubility. The text now correctly states that the disappearance of Ca signal indicates calcium components are soluble (**Lines 214-217, Page 8**).*

L227: I don't understand the logic behind the conclusion, that a stronger decrease in Ca-containing particle number compared to all dust particles points to a mixture. Wouldn't it be the other way around? If all particles were mixtures, the number would decrease similarly for both (or remain constant).

Response:

*The logic hinges on what happens to a particle after its soluble Ca dissolves. The key observation is that the total particle count did not decrease as much as the Ca-containing particle count. This is because many particles that lost their Ca signal (and thus exited the "Ca-containing" category) did not vanish entirely; they remained as insoluble residues and were still counted in the "total particle" number. Our data suggest the existence of particles where Ca is a soluble fraction within an insoluble matrix (as shown in **Lines 214-217, Page 8**).*

L231: How do we know about gravel surfaces in both deserts, if we're looking at the results of artificially generated dust? In a desert, there might be a Ca-dominated crust, which would probably have a considerable impact on the emissions process and is probably different for a topsoil, which was collected with a shovel.

Response:

We acknowledge that laboratory-generated dust samples cannot perfectly replicate all aspects of naturally emitted dust aerosols, and would like to clarify the rationale behind our approach:

Prior studies (e.g., Wu et al., 2022; Wu et al., 2023) have systematically compared the chemical and physical properties of dust generated from sampled soils with ambient aerosols, confirming their representativeness as potential dust source materials.

*Each surface soil sampling site was carefully selected to represent the dominant soil type and topography of the target desert area (see **Fig. S1**). Importantly, visible surface* *crusts were avoided during sampling, as they are not extensive landforms in our specific study regions. Our objective was to collect the loose, erodible material that constitutes the primary dust reservoir.*

Therefore, our lab-generated dust aerosols can, at least to a large extent, represent the physicochemical properties of major dust particles through saltation-sandblasting processes from the deserts.

L235: I understand from the method part, that the particle volume was estimated from the particle shape as seen in the electron microscope. However, for all particles shown in Figure 6 and S6, the electron images before and after dialysis are nearly identical. So, how can a mass loss be quantified?

Response:

*The mass loss is not derived from a measured change in total particle volume. Instead, it is quantified indirectly from the change in elemental composition. For each Ca-containing particle identified both pre- and post-dialysis, its mass is estimated from its volume (based on SEM) and a compositionally assigned density. The mass of water-soluble Ca within that particle is then calculated as: (particle mass) × (Ca wt% from EDX / 100). The total dissolved Ca mass is finally estimated by summing the difference in calculated Ca mass for all matched particles before and after dialysis (**Lines 157-164, Page 6**).*

Figure 1: The mass loss per particle doesn't seem to be significant. Please comment on that.

Response:

Yes, the mass loss per individual particle is not substantial. This observation strongly supports our interpretation that water-soluble calcium (Ca) primarily exists as an internally mixed coating on the surfaces of insoluble dust cores. The dissolution of this superficial layer leads to a mass change that is minor relative to the core's mass, while being chemically significant. Furthermore, the average mass flux ratios of water-soluble Ca from the Taklimakan and Gobi deserts align with previously reported calcium ion contents in both generated and ambient dust from the Taklimakan Desert (Wu et al., 2022). These results collectively underscore the prevalence of internally mixed, water-soluble Ca components in dust particles.

How can on the one hand for GB-Sand the mass of Ca per particle decrease, but at the other hand the volume increase?

Response:

This apparent contradiction is resolved by recognizing that dialysis selectively dissolves particles based on their solubility, thereby altering the statistical population. In ***Fig. 1(b)*** *and* ***1(c)****, the light blue and grey box plots represent two distinct groups of Ca-containing particles within the same original sample: those with water-soluble Ca components (light blue, pre-dialysis) and those with almost insoluble Ca minerals (grey). During dialysis, particles from the soluble group (light blue) lose their Ca and thus exit the "Ca-containing" category. Consequently, the post-dialysis group (grey) is predominantly composed of the initially insoluble particles. This shift from a mixed population to one enriched in insoluble grains explains the statistical change: the slight decrease in mean volume reflects the loss of many soluble particles, while the increase in median volume indicates the relative dominance of the persistently larger, insoluble Ca-containing particles in the remaining group.*

L237: Why does a different Ca particle mass indicate a different mineralogical composition? It should primarily come from a different particle size.

Response:

While particle size influences absolute mass, the difference in Ca mass highlighted here primarily indicates a difference in solubility and thus mineralogy. This is because the two particle groups (blue/pre-dialysis vs. grey/post-dialysis) have comparable sizes but contrasting responses to water: the Ca in the blue group is in partial-soluble forms (e.g., coatings) that dissolve, whereas the Ca in the grey group is locked in insoluble mineral structures. The distinct Ca mass therefore reflects this underlying difference in chemical form and mineralogical identity (see ***Lines 214-217, Page 8****).*

L239: For GB-Gobi the IQD gets larger. The min-max distance might well depend on the number of particles – the more you have to analyze, the higher the probability of extreme values. Please do some statistical checks on the significance.

Response:

We agree that the number of particles can influence the observed range. However, in this case, the increase in the interquartile distance (IQD) is not primarily a statistical artifact of sample size, but rather reflects a fundamental change in the particle population itself due to the dialysis process. The pre-dialysis ("blue box") and post-

dialysis ("grey box") groups are not independent samples from the same population; instead, the latter is a chemically altered subset of the former, formed by the selective removal of soluble calcium components. This selective process inherently creates a new population with its own distinct property distribution.

Consequently, applying standard statistical tests designed to compare independent groups (e.g., t-tests) would be conceptually inappropriate here, as the groups are intrinsically linked by the experimental treatment. Rather than comparing two independent populations, Fig. 1 depicts the property distribution shifts of a single particle population through a selective dissolution process. The change in IQD is thus a relevant descriptor of this transformation, directly resulting from the experimental mechanism.

L263: I don't see any evidence for a coating. These can be simply internally mixed aggregates, which are quite common in desert soils.

Response:

*In the revision, the description has been revised to indicate the possible presence of coatings (**Line 251, Page 9**). This nuance is further supported by additional evidence presented in the subsequent discussion.*

Figure 2: Please restrict the figure labels to significant digits corresponding to the measurement error.

Response:

*In the revision, the figure labels in **Fig. 2** have been revised to ensure that the number of significant digits corresponds to the respective measurement error of 0.1 wt%.*

L273: The form factor, as you define it, is calculated using the perimeter squared, so it is extremely sensitive to that value. That means that single pixel differences of the segmentation map have a strong impact on the result. Looking at your Figure 6, the images taken after dialysis are softer than before, i.e. they have less well-defined edges. As both Ca and non-Ca particles show a decrease in form factor, did you make sure that the change in image quality is not the reason for that?

Response:

Yes, the form factor is highly sensitive to the precise outline determined during segmentation. To ensure that the observed changes are not an artifact of image quality, all images (pre- and post-dialysis) were acquired and analyzed under identical CCSEM operating conditions, and the statistics are derived from a large population (>10,000 particles per sample, ***Fig. S6****). This controlled comparison minimizes systematic segmentation bias.*

*Within this framework, the consistent decrease in form factor for both Ca and total particles suggest a genuine physical change: coarser particle outlines after dialysis. We interpret this as evidence for the removal of soluble material, implying the prior presence of water-soluble coatings. Supporting this, manual measurements of the longest and orthogonal diameters (****Fig. 6*** *and* ***Fig. S7****) indicate an estimated coating thickness on the order of 10 to 100 nanometers.*

L274: The soluble Ca fractions you show in the images are not smooth.

Response:

*We have revised the text accordingly. The expression "smooth" has been changed to "uniform" (****Line 261, Page 10****).*

L282: Where is Ca in albite?

Response:

Although albite is the sodium end-member of the plagioclase series, natural samples often contain calcium due to solid solution. Its empirical formula can be represented as $Na_{0.95}Ca_{0.05}Al_{1.05}Si_{2.95}O_8$*, including a relatively low but present calcium content. The classification of Ca-containing minerals has been clarified in* ***Line 269, Page 10****.*

L293: Combining the thickness of the coating and the observation of no morphological change – could it be that the coating just preserves the shape of the particle and the particle itself inside it dissolved? I have a hard time imagining that particles are on one hand claimed to be partly dissolved (medium Ca-particles, Figure 2), but on the other hand should not show any morphological change.

Response:

*In fact, the opposite of the scenario is suggested. We propose that it is the thin, uniform water-soluble coating that dissolves, not the insoluble core particle inside it. The estimated nanometer-scale thickness of this coating is key to understanding why no pronounced morphological change is observed in SEM images. The removal of such a thin layer alters the particle's dimensions at a scale well below the detection limit for visible "shape change" in conventional manually-operated microscopy. The consistent, subtle reduction in particle diameter that we measured (**Fig. 6** and **Fig. S7**), without any drastic alteration in morphology, strongly supports the model of a thin, soluble surface layer being stripped away, leaving a persistent, insoluble core.*

L297: Again, missing evidence for a coating.

Response:

*In the revision, we have added a multi-part evidence chain (illustrated in **Fig. 6** and **Fig. S7**) for the presence of water-soluble Ca-rich coatings on insoluble mineral dust particles:*

1) Disappearance of the Ca signal after dialysis: This is clearly shown in both EDX spectra (for showing the elemental composition at the point computed to be most distant from the particle edge) and elemental mappings (for displaying the distribution of elements within its projected area). The strong Ca signal present before dialysis vanishes afterwards, indicating removal of a Ca-rich phase.

2) Morphological consistency with subtle size reduction: SEM micrographs show that particle morphology remains unchanged after dialysis, while a slight but measurable decrease in diameter occurs. This is precisely consistent with the loss of a thin surface layer rather than bulk dissolution of the core.

3) Emergence of core-element signals post-dialysis: Following dialysis, EDX spectra consistently reveal pronounced peaks of Si (and often Al), elements characteristic of the underlying insoluble silicate and aluminosilicate core. This further confirms that the removal of the surface Ca-layer exposes the persistent mineral substrate.

*These lines of evidence consistently point to the presence of a thin, water-soluble Ca-rich coating that is removed upon dialysis, leaving the insoluble mineral core intact and largely unchanged in shape. **(Lines 293-303, Page 12)***

L297: One would expect that the majority of compound here is crystalline (i.e. has a crystal lattice, is a mineral).

Response:

*The majority of particles that disappeared during dialysis were crystalline minerals with relatively pure mineralogical compositions, as noted in the text (**Lines 285-286, Page 11**). This observation is consistent with their high solubility. Their low number fractions in the overall sample (**Fig. 4**) indicate that such highly soluble, pure-phase crystals were not the dominant component of the bulk dust, but were the most susceptible to complete removal by dialysis.*

L299: If you have high resolution images, can you show the mentioned coating? It should also be visible on the EDX mapping.

Response:

*Multiple lines of evidence affirm thin, water-soluble Ca-rich coatings (see **Fig. 6** and **Fig. S**7): (1) chemical removal: loss of the EDX Ca signal. (2) morphological constancy: preserved SEM morphology with measurable size reduction. (3) core exposure: enhanced post-dialysis Si/Al signals indicating core exposure. Higher-resolution images have been added per the reviewer's suggestion (**Figs. 6** and **S**7).*

Figure 6: The authors claim a coating of Ca substances here. However, the images demonstrate that Ca is in the center of the particle in all shown cases. While it might be on the backside, I don't see any evidence of a coating. In the third row of images, also the Si seems to be gone, though it is difficult to compare the left and right mappings, as apparently the intensity-to-color scaling is different.

Response:

*The concern is an artifact caused by a size mismatch between the SEM micrograph and the EDX mapping in the original figure. In the revised **Fig. 6**, we have aligned them to the same scale. As corrected, the Ca signal is uniformly distributed across the entire projected area of the particle, supporting its presence as a surface layer rather than a concentrated core.*

Furthermore, the post-dialysis EDX mappings show a complete disappearance of the Ca signal, consistent with the removal of a water-soluble surface component.

*Regarding the Si signal, it is true that the element apparent changed. In the pre-dialysis EDX spectra (for all three particles in **Fig. 6**), the Si peak is absent because the dominant Ca signal and the nature of the surface coating likely attenuated the underlying core signal. After dialysis, with the removal of surface Ca coating, the Si* *peak (along with Al in* ***Fig. S7****) becomes visible in the spectra. This is not due to inconsistent intensity scaling, but rather to the unmasking of the insoluble silicate/aluminosilicate core, providing direct spectral evidence for the coating removal process.*

Spectra are illegible.

Response:

We have replaced the spectra in ***Fig. 6*** *and* ***Fig. S****7 with high-resolution versions for better clarity.*

L312-358: This is an introductory paragraph and literature review. The content should be mainly moved into the introduction (and maybe shortened), only results from the present work and directly connected papers should be discussed in a results section.

Response:

*In the revision, we have moved the general discussion on the atmospheric significance, dissolution kinetics, and modelling of calcite (and related minerals) to the Introduction section (****Lines 52-58, Page2-3****), where it provides a clearer rationale for our study.*

L327: A trajectory analysis commonly related, in atmospheric science, to an air parcel transport history. I think an aerosol transport model is meant here.

Response:

In the revision, we have deleted this extraneous discussion to maintain focus and accuracy. Indeed, "trajectory analysis" pertains to transport pathways, not to the detailed chemical processes we discussed.

L327: I don't see the logical contrast to the previous sentence here.

Response:

As the point did not directly pertain to our main line of argument, we have deleted it to improve the manuscript's focus in the revision.

L329-330: What is the connection to weathering here?

Response:

This discussion has been removed to ensure a more focused and compact narrative.

L331-338: That's interesting in general, but I don't see how it relates to this paper directly.

Response:

Yes, portions of the cited passage were of general interest but not directly pertinent to our specific findings. We have removed those tangential sentences in the revision.

We have retained and focused the subsequent discussion on aspects directly relevant to our paper, namely that calcite dissolution is a surface-controlled process with mass-transport limited kinetics (Laanait et al., 2015; Batchelor-McAuley et al., 2022). This concept directly supports our interpretation that the presence of soluble surface Ca coatings could modify the mass transfer rate and thus potentially accelerate dissolution kinetics, which is a key implication of our single-particle observations.

L353-355: I've seen no evidence for that, on the contrary. That would also need to be supported with calculations of dissolution kinetics or direct evidence of coatings, e.g. in mappings.

Response:

*We have refined our discussion to focus on the mechanistic framework supported by our data and the literature. We provided high-resolution mapping evidence for the coatings (**Fig.6** and **Fig. S7**) and specifically discussed that calcite dissolution is a surface-controlled process with mass-transport limited kinetics (citing, e.g., Laanait et al., 2015; Batchelor-McAuley et al., 2022). Within this established framework, our observation of rapid Ca removal is consistent with the presence of soluble Ca-rich surface phases that could enhance dissolution kinetics by modifying local mass transfer conditions. This presents a plausible interpretation of our results, which we now present more cautiously as a testable hypothesis rather than a definitive conclusion. Please refer to **Lines 293-303, Page 12**.*

References

Batchelor-McAuley, C., Yang, M., Rickaby, R. E., and Compton, R. G.: Calcium carbonate dissolution from the laboratory to the ocean: Kinetics and mechanism, Chem. Eur. J., 28, e202202290, https://doi.org/10.1002/chem.202202290, 2022.

Laurent, B., Marticorena, B., Bergametti, G., Chazette, P., Maignan, F., Schmechtig, C.: Simulation of the mineral dust emission frequencies from desert area of China and Mongolia using an aerodynamic roughness length map derived from the POLDER/ADEOS surface products. J. Geophys. Res. 110, D18S04. https://doi.org/10.1029/2004JD005013, 2005.

Sweeney, M.R., Lu, H., Cui, M., Mason, J.A., Feng, H., Xu, Z.: Sand dunes as potential sources of dust in northern China. Sci. China Earth Sci. 59 (4), 760–769. https://doi.org/10.1007/s11430-015-5246-8, 2016.

Wang, X., Hua, T., Zhang, C., Lang, L., Wang, H.: Aeolian salts in gobi deserts of the western region of Inner Mongolia: gone with the dust aerosols. Atmos. Res. 118, 1–9. https://doi.org/10.1016/j.atmosres.2012.06.003, 2012a.

Wang, X.M., Lang, L., Hua, T., Wang, H., Zhang, C., Wang, Z.: Characteristics of the Gobi desert and their significance for dust emission in the Ala Shan Plateau (Central Asia): an experimental study. J. Arid Environ. 69 (81), 809–821. https://doi.org/10.1016/j.jaridenv.2012.01.014, 2012b.

Wu, F., Cheng, Y., Hu, T., Song, N., Zhang, F., Shi, Z., Hang Ho, S. S., Cao, J., and Zhang, D.: Saltation–sandblasting processes driving enrichment of water-soluble salts in mineral dust, Environ. Sci. Technol. Lett., 9, 921-928, https://doi.org/10.1021/acs.estlett.2c00652, 2022.

Wu, F., Song, N., Hu, T., Ho, S. S. H., Cao, J., and Zhang, D.: Surrogate atmospheric dust particles generated from dune soils in laboratory: Comparison with field measurement, Particuology, 72, 29-36, https://doi.org/10.1016/j.partic.2022.02.007, 2023.

Zou, X.Y., Li, J.F., Cheng, H., Wang, J.P., Zhang, C.L., Kang, L.Q.: Spatial variation of topsoil features in soil wind erosion areas of Northern China. Catena 167, 429–439. https://doi.org/10.1016/j.catena.2018.05.022, 2018.

Revision sheet

Original manuscript		**Revised manuscript**	
Line 16	two typical Asian dust source regions	Line 24	in two deserts
Line 22	The abundance and mixing state of water-soluble calcium-containing particles in mineral dust emitted from Asian dust source regions provide realistic constraints for assessing their role in enhancing atmospheric acid neutralization and mitigating ocean acidification.	Line 20	The results provide a realistic constraint for assessing the abundance of water-soluble Ca coatings on fine particles emitted from two Asian deserts.
Line 29	Global dust emissions are estimated at 2000 ± 400 Tg yr–1 (Kok et al., 2020), with Asia contributing approximately 25–30 % of this total (Kok et al., 2021).	Line 27	Global dust emissions are estimated from 2000 ± 400 Tg yr^{-1} (Kok et al., 2021a) to 2566 ± 1996 Tg yr^{-1} (Zhao et al., 2023), with Asia contributing from 25-30 % (Kok et al., 2021b) to ~40 % (Kok et al., 2023) of the total.
Line 43	below the direct entrainment threshold	Line 41	below the direct entrainment threshold of fine dust particles
Line 55		Line 52	*Added:* Calcite, as a ubiquitous Ca-rich mineral in arid soils, constitutes 5–15% of dust from major source regions (Engelbrecht and Derbyshire, 2010; Knippertz and Stuut, 2014). It is a potent alkaline agent that neutralizes acids (e.g., H_2SO_4, HNO_3) during transport (Wang Y. et al., 2012), moderating aerosol pH and influencing reactivity (Usher et al., 2003; Craig et al., 2018). During these acid neutralization processes, calcite dissolution is kinetically controlled and not instantaneous, the extent of its chemical aging—and thus its buffering efficiency—is strongly influenced by atmospheric acidity and transport timescale (Morse et al., 2007).
Line 84	calcite- and gypsum-containing particles	Line 87	Ca-O-rich and Ca-S-containing dust particles
Line 89	Sand dunes and gravel soils are representative of different surface conditions of the desert terrain and potentially major sources of atmospheric dust from deserts (Mikami et al., 2005; Wang et al., 2012; Zou et al., 2018). In this study, 4 soil samples were collected from typical sand dune and gravel desert surfaces in two major dust source regions in China: the Taklimakan Desert and the Gobi deserts on the Alashan Plateau (Fig. S1).	Line 92	Sand dunes and gravel soils are recognized as major sources of atmospheric dust in the arid regions of northern China (Laurent et al., 2005; Mikami et al., 2005; Wang X. et al., 2012; Sweeney et al., 2016; Zou et al., 2018). To this end, four representative soil samples were collected from these two surface types in the Taklimakan Desert and the Gobi deserts (Fig. S1).
Line 99	Surface soil (0–5 cm) was collected by a plastic shovel and stored in a self-sealed polyethylene bag. All samples were air-dried and stored at room temperature without pretreatment.	Line 101	Surface soil (0-5 cm depth) was collected using a 20 cm × 5 cm plastic shovel. Following collection, samples were placed into self-sealing polyethylene bags (Fig. S1), air-dried in the laboratory, and then stored at room temperature without sieving. For

			each sample, the coordinates, type, and description of the surroundings were documented.
Line 109	0.2 μm in porosity	Line 112	pore size of 0.2 μm
Line 119	scanning electron microscope (MAIA3, Tescan, Brno, Czech Republic) equipped with two energy dispersive X-ray (EDX) spectrometer	Line 123	gun scanning electron microscope (MAIA3, Tescan, Brno, Czech Republic) equipped with dual energy dispersive X-ray (EDX) detectors
Line 122	particle image, size distribution, morphological parameter	Line 126	particle images, size distribution, morphological parameters
Line 128	dust particles	Line 131	dust particles (Fig. S2) without disturbing their original locations (Fig. S3)
Line 133	calcite and gypsum-like	Line 136	Ca-O-rich and Ca-S-containing
Line 141	$D_{\max}$ in formula (1)	Line 144	D_{avg} in formula (1)
Line 163-171	formula (3)-(4)	Line 165-177	*Rewritten*: formula (3)-(7)
Line 176	The accuracy and representativeness of particle size, morphology, and elemental composition were validated by the measurement of standard laboratory reference materials (700 nm polystyrene latex spheres, Duke Scientific Corp., Palo Alto, CA, USA) under identical CCSEM conditions.	Line 183	The accuracy of the particle size measurements was confirmed using standard polystyrene latex spheres (802 nm ± 6 nm, Duke Scientific Corp.) under identical CCSEM conditions. The mean size from three independent measurements (each with $n > 6000$ particles) was 796 nm ± 16 nm.
Line 179	Elements ranging from carbon (C) to lead (Pb) were identified on the basis of their X-ray peak intensities, which were converted into relative atomic percentages by EDX software with ZAF correction (Z: atomic number; A: mass absorption; F: fluorescence). An estimated error margin of 1.0 wt % was used	Line 186	Elemental composition was quantified based on the characteristic X-ray peak intensities, covering elements from carbon (C) to lead (Pb). The EDX software converted these intensities into relative atomic percentages using a ZAF correction procedure (accounting for atomic number, absorption, and fluorescence effects). A quantification threshold of 0.5 wt% and an error margin of 0.1 wt% were applied
Line 187	'silicate' or 'aluminosilicate'	Line 194	'Mixed silicate minerals' or 'Mixed aluminosilicate minerals'
Line 192		Line 200	*Added:* The total particle mass derived from CCSEM was compared with that measured by the gravimetric method (Table S4), showing the cumulative uncertainty arising from the ellipsoid volume assumption, the inference of density from composition, and the semi-quantitative nature of EDX analysis. Procedural blanks were measured for gobi and sand sample to quantitatively represent procedural contamination levels in this

			method (Table S5). To examine the dissolution time series, a separate dust sample from the Taklimakan desert (whose Ca-containing particle abundance fell within the range of the four main samples) underwent double-dialysis. As presented in Table S6, the differences in the number fractions of Ca-containing particles between dialysis cycles were within the pre-dialysis standard deviations for this particle group, indicating that the water-soluble Ca components are effectively removed during a single 2-hour dialysis process. Moreover, an additional acid dialysis experiment at pH = 5.1 ± 0.1 was performed on the aforementioned sample. The removal of more Ca-containing particles (Table S6) indicated that our relative neutral-water experiment represents a conservative-removal scenario.
Line 224	From a single-particle perspective,	Line 212	*Deleted*
Line 226	more than half of the Ca-containing particles disappeared after dialysis, identifying calcium as the most mobile element. The decline in the number of Ca-containing particles was greater than that of total dust particles, suggesting that the vanished Ca-containing particles were mixtures of water-soluble Ca compounds and other insoluble minerals. During water dialysis, the water-soluble Ca components dissolved, leaving behind the insoluble non-Ca minerals.	Line 214	after dialysis, the calcium signal disappeared in more than half of the initially Ca-containing particles. This greater decline in Ca-containing group compared to the total particle count suggests that the vanished particles were mixed-phase, comprising water-soluble Ca compounds associated with insoluble minerals. The dialysis process selectively dissolved the soluble Ca components from these particles, leaving behind insoluble residues.
Line 237	minerals	Line 224	residues
Line 250	(Table 1). These values account for 11.2 % and 30.0 % of the total dust particle number fluxes in each desert, respectively (Table S4). In mass terms, the average water-soluble Ca components were 0.5 μg m–2 s–1 for the Taklimakan Desert and 3.3 μg m–2 s–1 for the Gobi Desert (Table 1), representing 1.3 % and 3.3 % of the total dust particle mass in the Taklimakan and Gobi Desert, respectively (Table S4).	Line 237	These values account for 9.9 % and 29.2 % of the total dust particle number fluxes in each desert, respectively. In mass terms, these water-soluble elemental Ca accounted for 19.6–41.9 % of the mass of elemental Ca in both the Taklimakan and Gobi deserts. The average fluxes of water-soluble elemental Ca were 0.4 μg m^{-2} s^{-1} for the Taklimakan Desert and 3.2 μg m^{-2} s^{-1} for the Gobi deserts, representing 1.1 % and 3.3 % of the total dust particle mass fluxes in the Taklimakan and Gobi deserts, respectively.
Line 256	Table 1	Line 245	*Rewritten:* Table 1

Line 263	presence	Line 251	potential presence
Line 274	The development of more irregular particle edges confirms that the dissolved components had previously formed uniform coatings over the inherently rougher surfaces of the insoluble mineral cores.	Line 261	The resulting particles developed more irregular edges, supporting the idea that the dissolved components had formerly formed smooth coatings on the insoluble mineral surfaces.
Line 281		Line 269	*Added*: according to their empirical formula
Line 291	Therefore, all calcite and gypsum particles were relocated, and EDX mappings of each single particle were manually acquired to examine changes in their mixing states. Most calcite-type particles showed no obvious morphological changes after dialysis. These particles were classified as calcite internally mixed with other minerals (i.e., coating), accounting for 73 % of calcite-containing particles from sandy surfaces in the Gobi Desert to 91 % from gravel surfaces in the Taklimakan Desert 295 (Fig. 5). The proportion of dust particles with a gypsum coating was even higher, exceeding 81 % in all samples. Particles that disappeared after dialysis were categorized into the crystalline group, which may have possessed relatively pure mineralogical compositions. The morphology of particles coated with calcite (Fig. 6) and gypsum (Fig. S6) is illustrated along with their corresponding EDX spectra and elemental distribution mappings. Based on their projective outlines in high-resolution micrographs, the thickness of the calcite and gypsum coatings was estimated to be on the order of several 300 nanometers.	Line 280	To this end, we performed targeted manual EDX mapping on individual Ca-O-rich and Ca-S-containing particles to supplementally assess changes in their mixing states. Most Ca-O-rich (calcite-like) particles exhibited minimal morphological change following dialysis. These were inferred to be Ca-O-rich components internally mixed with other minerals (i.e., coating on) other minerals. Their proportion varied geographically, accounting for 73.2 % of Ca-O-rich particles from sandy Gobi desert surfaces to 91.4 % from gravel surfaces in the Taklimakan Desert (Fig. 5). The proportion of dust particles containing a Ca-S-containing (gypsum-like) coating was even higher, exceeding 81.3 % across all samples. In contrast, particles that vanished during dialysis were categorized as a crystalline group, likely consisting of relatively pure mineral phases.
Line 312	Calcite is a ubiquitous mineral in arid and semi-arid surface soils and constitutes a substantial portion of globally emitted alkaline dust. It typically constitutes 5–15 % of dust originating from major source regions such as North Africa and Asia (Engelbrecht and Derbyshire, 2010; Knippertz and Stuut, 2014). The atmospheric significance of calcite lies in its ability to neutralize acidic gases, thereby exerting a buffering effect on aerosol chemistry (Wang Z et al., 2002). Furthermore, upon deposition, it plays a crucial role in mitigating surface ocean acidification (Tipper et al., 2016). In many model simulations, calcite is often treated as a sparingly soluble mineral, with its dissolution considered to be kinetically controlled and highly dependent on ambient acidity (pH). Since calcite dissolution is not	Line 293	The morphology of dust particles coated with Ca-O-rich (Fig. 6) and Ca-S-containing (Fig. S7) material is presented together with their corresponding EDX point spectra (taken at the point computed to be most distant from the particle edge) and elemental distribution mappings (over the projected particle area) across the dialysis process. A multi-part chain of evidence confirms the presence of water-soluble Ca-rich coatings on insoluble mineral dust particles. First, both EDX spectra and elemental mappings show the disappearance of the Ca signal after dialysis, indicating the removal of a Ca-rich phase. Second, SEM micrographs reveal that particle morphology remains essentially unchanged after dialysis, while

	instantaneous but occurs over timescales relevant to atmospheric transport (hours to days), the extent of its chemical aging is strongly influenced by atmospheric acid concentrations and transport pathways (Morse et al., 2007). During transport, calcite acts as a potent alkaline agent, effectively neutralizing acids such as H_2SO_4, HNO_3, and SO_2 (Wang Y et al., 2012). This reaction can moderate aerosol pH, influencing their overall reactivity (Usher et al., 2003; Craig et al., 2018), and modify the particles' optical properties and cloud-forming potential (Craig and Ault, 2018; Zhi et al., 2025).		a subtle but measurable decrease in diameter occurs. This pattern is consistent with the loss of a thin surface layer rather than with bulk dissolution of the core. Third, post-dialysis EDX spectra display newly-appeared peaks of Si (and frequently Al), elements characteristic of the underlying insoluble silicate or aluminosilicate core. This further confirms that removal of the surface Ca-coatings exposes the persistent mineral substrate. Based on the projected outlines in high-resolution micrographs, the thickness of the Ca-O-rich and Ca-S-containing coatings is estimated to be on the order of 10–100 nm.
Line 324	In modeling studies, although simulated air parcels typically experience water saturation prior to homogeneous ice nucleation, the fate of mineral dust processed within liquid water clouds remains highly uncertain. For dust originating from the Taklimakan Desert, which often travels at altitudes well above the polluted Asian planetary boundary layer, chemical coatings are commonly neglected in trajectory analyses. In contrast, for Gobi-sourced dust, frequently transported at lower altitudes toward the Pacific coast, the omission of chemical aging processes may introduce significant uncertainty (Wiacek et al., 2010). Experimental investigations into the dissolution kinetics of anorthite have revealed that CO_2-mediated dissolution of Ca-feldspar is accompanied by accelerated carbonate-promoted weathering (Berg and Banwart, 2000). Further laboratory studies indicate that the reactivity of silicate minerals with CO_2 is governed by solution pH, pCO_2, and the parent silicate's crystal structure (Golubev et al., 2005). For instance, when calcium silicate ($CaSiO_3$) slowly dissolves in the presence of aqueous CO_2, a calcium-depleted leached layer develops, and released Ca^{2+} can subsequently precipitate as solid $CaCO_3$ (Plattenberger et al., 2018). The solubility of $CaCO_3$ increases markedly with rising CO_2 pressure, from approximately 1.5×10^{-4} mol L^{-1} in a CO_2-free atmosphere at 25 °C to about 0.008 mol L^{-1} under1 bar CO_2, with the additional dissolved carbonate existing primarily as HCO_3^- in solution. While calcite dissolution has been identified as a surface-controlled process (Laanait et al., 2015), its kinetics are also influenced by	Line 310	The atmospheric importance of such a thin, water-soluble Ca-rich coating on an insoluble mineral core lies in its capacity to neutralize acidic gases, thereby exerting a buffering effect on aerosol chemistry. Ca-rich minerals (e.g., calcite) are ubiquitous in arid and semi-arid surface soils and constitute a substantial fraction of globally emitted alkaline dust. During atmospheric transport, calcite acts as an effective alkaline agent, neutralizing acids such as H_2SO_4, HNO_3, and SO_2 (Wang Y. et al., 2012). Calcite dissolution has been identified as a surface-controlled process (Laanait et al., 2015), thus its kinetics are largely influenced by mass-transport limitations, underscoring the importance of particle size and mixing state (Batchelor-McAuley et al., 2022). Nevertheless, key constraints such as reaction timescales and size-dependent effects are often oversimplified or entirely omitted in global models, where dust particles are typically assumed to undergo either instantaneous complete dissolution or equilibrium partitioning (Pye et al., 2020). Our automated microanalysis shows that 56.9–88.2 % of Ca-containing dust particles released their water-soluble Ca components. We further infer that the Ca-O-rich component, most likely calcite, one of the most soluble minerals in the source dust, exists predominantly as 10–100 nm-thick coatings on micron- and submicron-sized insoluble particles. This specific mixing state may accelerate the dissolution kinetics of calcite by altering the mass-

	mass-transport limitations, underscoring the importance of particle size and mixing state (Batchelor-McAuley et al., 2022). Nevertheless, such critical constraints, including reaction timescales and size-dependent effects, are often oversimplified or entirely neglected in global models, where dust particles are typically assumed to undergo either instantaneous complete dissolution or equilibrium partitioning (Pye et al., 2020). Owing to its high deliquescence point, Ca(HCO$_3$)$_2$ behaves as a non-hygroscopic salt below 97.5 % RH, yet it serves as an effective cloud condensation nucleus due to its considerable solubility (Zhao et al., 2010). Ultimately, the ice nucleation activity of dust particles under specific temperature and humidity conditions depends on a range of factors, including mineral composition, particle size, chemical coatings, and the co-presence of other aerosol species (Wiacek et al., 2010).		transfer rate. These findings provide observational constraints on water-soluble Ca associated with fine dust particles from two Asian deserts and offer qualitative insights, from a single-particle perspective, into the physicochemical properties of freshly emitted mineral dust.
Line 345	Beyond the atmosphere, the long-range transport and deposition of water-soluble Ca-containing mineral dust fundamentally influence the marine carbon cycle (Barker et al., 2003). This process provides a critical external source of alkalinity to the open ocean. The dissolution of dust-derived calcite enhances the ocean's capacity to neutralize CO_2, shifting the carbonate system equilibrium and promoting greater oceanic uptake of atmospheric CO_2. Consequently, this mechanism exerts long-term negative feedback on climate change over millennial timescales (Archer et al., 2009; Middelburg et al., 2020). Therefore, calcite flux from mineral dust acts not only as an atmospheric chemical agent but also as a key biogeochemical connector between continental weathering, ocean chemistry, and the global carbon cycle. Its accurate representation in models is essential for reconstructing past and projecting future climate states. Our automated microanalysis revealed that 56.9–88.2 % of Ca-containing dust particles released their water-soluble Ca components. We further identified that calcite, one of the most soluble minerals in the source dust, predominantly exists as nanometer-sized coatings on micron- and submicron-sized insoluble particles. This specific mixing state potentially accelerates the dissolution kinetics of calcite and modifies the hygroscopicity of the dust particles. This finding implies a more rapid release of alkalinity from Asian Dust and a	Line 325	Moreover, our dialysis experiment was designed to examine the abundance and mixing state of water-soluble Ca components on dust particles, not to simulate atmospheric cloud processing. Dialysis in Milli-Q water (pre-equilibrated in open air at 25 ± 5 °C for 48 hours, pH 6.4 ± 0.7) effectively removed soluble material, and subsequent CCSEM analysis quantified the resulting changes in surface composition. In contrast, a separate humidity-exposure experiment (Fig. S8) demonstrated that under thin-film conditions, more representative of atmospheric moisture, these coatings dissolved and recrystallized upon drying rather than being fully removed (Fig. S9 and S10). This implies that our dialysis setup, employing bulk water at near-neutral pH, likely yields a conservative estimate of coating removal, given that atmospheric waters are often acidic and contain ions that could enhance dissolution. In model simulations, calcite is often treated as a sparingly soluble mineral whose dissolution is kinetically controlled and highly dependent on ambient acidity (pH). An additional acid dialysis performed on a separate dust sample after dual water dialysis (Table S6) confirmed that acidic conditions promote the dissolution of Ca-containing particles. Although not a direct replication of in-cloud conditions, this study

	consequently enhanced neutralization capacity for acidity across Earth's environmental compartments than previously recognized.		clearly establishes the presence of water-soluble calcium coatings on dust particles, a feature relevant to climate-related properties such as cloud-condensation activity.
Line 193	*Section:* 2.5 Advantages and limitations of methodological approaches	Line 338	*Section:* 3.4 Advantages and limitations of methodological approach
Line 363	Asian dust source regions. The prevalent occurrence (>73 %) of water-soluble calcite and gypsum as surface coatings in these dust particles suggests their rapid dissolution even in moderately acidic environments. This rapid dissolution behavior significantly enhances the acid-neutralizing capacity of freshly emitted dust generated through saltation-sandblasting processes. These findings provide critical constraints for assessing the role of Asian dust in amplifying atmospheric acid neutralization and alleviating ocean acidification. Furthermore, the results highlight the need for more realistic representations of atmospheric mineral dust in both experimental and modelling studies.	Line 371	two Asian deserts. Importantly, over 73 % of this calcium exists as surface coatings, making it readily available for rapid dissolution even under moderately acidic conditions. This rapid dissolution likely enhances the acid-neutralizing capacity of freshly emitted dust from saltation-sandblasting processes. Therefore, accurately projecting the role of dust in the Earth system requires that future experimental and modelling frameworks explicitly account for its transport, chemical evolution, and the mass balance of alkalinity delivery.
Line 393	Archer, D., Eby, M., Brovkin, V., Ridgwell, A., Cao, L., Mikolajewicz, U., Caldeira, K., Matsumoto, K., Munhoven, G., Montenegro, A., and Tokos, K.: Atmospheric lifetime of fossil fuel carbon dioxide, Annu. Rev. Earth Planet. Sci., 37, 117-134, https://doi.org/10.1146/annurev.earth.031208.100206, 2009.		*Deleted*
Line 403	Barker S., Higgins J.A. and Elderfield H.: The future of the carbon cycle: review, calcification response, ballast and feedback on atmospheric CO2, Phil. Trans. R. Soc. A., 361, 1977-1999, http://doi.org/10.1098/rsta.2003.1238, 2003.		*Deleted*
Line 407	Berg, A., and Banwart, S. A., Carbon dioxide mediated dissolution of Ca-feldspar: implications for silicate weathering, Chem. Geol., 163, 25-42, https://doi.org/10.1016/S0009-2541(99)00132-1, 2000.		*Deleted*
Line 420	Chen, W. H., and Lu, J. J., Microphysics of atmospheric carbon dioxide uptake by a cloud droplet containing a solid nucleus. J. Geophys. Res. Atmos., 108. https://doi.org/10.1029/2002jd003318, 2003.		*Deleted*
Line 448	Golubev, S. V., Pokrovsky, O. S., and Schott, J., Experimental determination of the effect of dissolved CO2 on the dissolution kinetics of Mg and Ca silicates at 25 °C, Chem. Geol., 217, 227-238, https://doi.org/10.1016/j.chemgeo.2004.12.011, 2005.		*Deleted*

Line 483		Line 479	*Added*: Kok, J. F., Storelvmo, T., Karydis, V. A., Adebiyi, A. A., Mahowald, N. M., Evan, A. T., He, C., and Leung, D. M.: Mineral dust aerosol impacts on global climate and climate change. Nature Reviews Earth & Environment, 4(2), 71-86, https://doi.org/10.1038/s43017-022-00379-5, 2023.
Line 493		Line 492	*Added*: Laurent, B., Marticorena, B., Bergametti, G., Chazette, P., Maignan, F., Schmechtig, C.: Simulation of the mineral dust emission frequencies from desert area of China and Mongolia using an aerodynamic roughness length map derived from the POLDER/ADEOS surface products. J. Geophys. Res. 110, D18S04. https://doi.org/10.1029/2004JD005013, 2005.
Line 504	Middelburg, J. J., Soetaert, K., and Hagens, M.: Ocean alkalinity, buffering and biogeochemical processes, Rev. Geophys., 58, e2019RG000681, https://doi.org/10.1029/2019RG000681, 2020.		*Deleted*
Line 520	Plattenberger, D. A., Ling, F. T., Tao, Z., Peters, C. A., and Clarens, A. F. Calcium silicate crystal structure impacts reactivity with CO2 and precipitate chemistry, Environ. Sci. Technol. Lett., 5, 558-563, https://doi.org/10.1021/acs.estlett.8b00386, 2018.		*Deleted*
Line 557		Line 554	*Added*: Sweeney, M.R., Lu, H., Cui, M., Mason, J.A., Feng, H., Xu, Z.: Sand dunes as potential sources of dust in northern China. Sci. China Earth Sci. 59 (4), 760–769. https://doi.org/10.1007/s11430-015-5246-8, 2016.
Line 571	Warneck, P.: The equilibrium distribution of atmospheric gases between the two phases of liquid water clouds, in: Chemistry of Multiphase Atmospheric Systems, NATO ASI Series, edited by: Jaeschke, W., Springer, Berlin, Heidelberg, Germany, 743-499, https://doi.org/10.1007/978-3-642-70627-1_17, 1986. Wiacek, A., Peter, T., and Lohmann, U., The potential influence of Asian and African mineral dust on ice, mixed-phase and liquid water clouds, Atmos. Chem. Phys., 10, 8649-8667, https://doi.org/10.5194/acp-10-8649-2010, 2010.		*Deleted*
Line 576		Line 570	*Added*: Wang, Y., Yu, W., Pan, Y., and Wu, D.: Acid neutralization of precipitation in Northern China. Journal of the Air & Waste Management Association, 62(2), 204-211,

			https://doi.org/10.1080/10473289.2011.640761, 2012. Wang, Z., Akimoto, H., and Uno, I.: Neutralization of soil aerosol and its impact on the distribution of acid rain over east Asia: Observations and model results. Journal of Geophysical Research: Atmospheres, 107(D19), ACH-6, https://doi.org/10.1029/2001JD001040, 2002.
Line 586		Line 585	*Added*: Zhao, Y., Yue, X., Cao, Y., Zhu, J., Tian, C., Zhou, H., Chen, Y., Hu, Y., Fu, W., and Zhao, X.: Multi-model ensemble projection of the global dust cycle by the end of 21st century using the Coupled Model Intercomparison Project version 6 data. Atmospheric Chemistry and Physics, 23(13), 7823-7838, https://doi.org/10.5194/acp-23-7823-2023, 2023.