********Response to community comments from johanes kepler, 10 Dec 2025********

We thank Johanes Kepler for the helpful comments. They helped us substantially improve the manuscript. Our point-by-point responses are provided below in italics. (The updated Figures mentioned below are also provided at the end of this letter for easy reference.)

1 Points 1–7

1. Dissolution selectivity may be over-interpreted. The manuscript infers that certain Ca-bearing phases (e.g., “calcite” and “gypsum”) are “most soluble” because many Ca-containing particles disappear after water dialysis (see discussion around the mineral fractions after dialysis). However, the dialysis conditions (bulk Milli-Q water, near-neutral pH, fixed duration) are not equivalent to atmospheric aerosol water (thin films, variable pH, variable ionic strength). Loss of Ca signal/particles can also arise from removal of coatings, detachment of fragile aggregates, or reclassification artifacts rather than true mineral dissolution. This weakens a direct mapping from “disappearance after dialysis” to “intrinsic solubility ranking”.

Response:

*To contextualize the atmospheric relevance and address the role of thin water films, more analogous to atmospheric humidification, we conducted a separate water-vapor exposure experiment. As shown in **Fig. S8**, dust particles were exposed to saturated water vapor for 24 h, allowing thin aqueous films to form on their surfaces. In this exposure experiment, soluble Ca coatings, if existed, were not removed. Instead, upon drying and under SEM vacuum, they recrystallized on the surface, altering morphology but not disappearing (**Figs. S9** and **S10**). This contrasts sharply with the dialysis experiment, where dissolved material is physically removed, and underscores that the wetting–drying cycles may redistribute soluble components rather than permanently eliminate them.*

We fully agree with the comment that dialysis conditions (bulk Milli-Q water, near-neutral pH, fixed duration) differ substantially from atmospheric aerosol water (thin films, variable pH, and ionic strength). Away from dust source regions, atmospheric water in various phases is often acidic and contains elevated ions, which could enhance the solubility of some Ca-bearing minerals such as calcite and gypsum (e.g., Frear & Johnston, 1929; Miller, 1952; Urey & Holland, 1965; Batchelor-McAuley et al., 2022).

Thus, our dialysis experiment likely provides a conservative estimate of coating removal potential.

*The loss of Ca signal particles after dialysis, coupled with preserved particle size and shape, indicates the removal of water-soluble surface coatings rather than artifacts from aggregate detachment or reclassification. While we do not equate "disappearance after dialysis" with an intrinsic solubility ranking, the experiment clearly (**Fig. S2**) demonstrates that water-soluble Ca exists as coatings on dust particles, a feature relevant to particle-level properties such as cloud condensation nucleus activity and the climate effects of mineral dust.*

*The additional experiments of exposing to water vapor (**Figs. S8, S9, and S10**) and the result were added in the revised manuscript, as detailed in **Lines 325-332 on Page 14**.*

2. Emission-flux estimates rely on strong chamber and sampling assumptions. The emission flux formulas

$$F_{\text{sol-Ca}}(N) = \frac{N_{\text{sol-Ca}}}{(\nu t)} \frac{A_{\text{filter}}}{A_{\text{view}}} \frac{Q}{A_{\text{chamber}}}, \qquad F_{\text{sol-Ca}}(m) = \frac{m_{\text{sol-Ca}}}{(\nu t)} \frac{A_{\text{filter}}}{A_{\text{view}}} \frac{Q}{A_{\text{chamber}}} \qquad (1)$$

implicitly assume uniform mixing, negligible wall/tubing losses, and representative sampling of the chamber outflow. Without explicit loss corrections and uncertainty propagation, these fluxes should be treated as order-of-magnitude estimates; absolute values may be biased even if relative comparisons are robust.

Response:

*We totally agree that the fluxes should be treated as order-of-magnitude estimates. While we implemented measures to minimize loss, including the use of conductive silicone rubber tubing and chemical/electrostatic passivation of all interior aluminum surfaces, a quantitative assessment of particle loss during sampling was not available. Furthermore, although the CCSEM-based mass calculations (closely relying on assumed particle density) were calibrated against a gravimetric method using nine collocated samples (**Table S4**), the overall uncertainty was not fully constrained. Consequently, the reported flux values are order-of-magnitude estimates derived from the measured particle counts and sampling parameters, even with corrections by procedural blanks (**Table S5**). These limitations have been explicitly noted in the "Strengths and Limitations" section in the revision (**Lines 339-346, Pages 14-15**).*

3. Phase identification from CCSEM–EDX is intrinsically ambiguous. The classification of “calcite”-type particles from elemental Ca–O signatures is not phase-specific: $CaCO_3$, CaO, $Ca(OH)_2$, and $Ca(NO_3)_2$ can yield similar EDX elemental ratios. Since CCSEM–EDX does not resolve chemical bonding, statements of the form “calcite coatings dominate” should be presented as “Ca–O-rich coatings consistent with carbonate/oxide/hydroxide/nitrate mixtures” unless corroborated by phase-sensitive methods (e.g., Raman, XRD/EBSD).

Response:

*We fully agree with this important point regarding the intrinsic ambiguity of phase identification based on CCSEM-EDX elemental compositions: phase identification based on EDX elemental ratios is inherently uncertain. While we addressed the limitations of identifying Ca-O-rich particles as calcite (**Lines 355-364, Page 15**), we also acknowledge that our work did not include phase-sensitive confirmation (e.g., Raman, XRD) and that the method cannot distinguish polymorphs such as calcite from aragonite.*

In the revision, we have revised the terminology throughout the manuscript. Specifically: (i) All instances of “calcite” have been replaced with “Ca-O-rich coatings consistent with carbonate/oxide/hydroxide/nitrate mixtures” or, where context permits, the more general term “Ca-O-rich dust particles”. (ii) All instances of “gypsum” have been changed to “Ca-S-containing”. (iii) Additionally, references to mineral types have been qualified as “-like” where appropriate to reflect the inferential nature of the identification.

*These edits have been made at the specified locations (e.g., **Line 87, Page 3; Line 136, Page 5; Line 280-285, Page 11; Line 293-303, Page 12; Line 319, Page 14; Line 365-366, Page 15**). These revisions accurately reflect the analytical limitations and strengthen the manuscript.*

4. Dialysis conditions are not a direct proxy for atmospheric processing. The experiment uses a 2-hour water dialysis in bulk liquid water at an average reported pH, whereas atmospheric processing typically occurs in microdroplets or thin aqueous films with evolving ionic strength and acidity. Thus, translating dialysis removal fractions into in-atmosphere dissolution kinetics (minutes–hours) is not straightforward without a kinetic model (diffusion limitations, interfacial reaction rates, RH dependence).

Response:

We fully acknowledge that our bulk dialysis conditions do not directly replicate the *complex, evolving microenvironment of atmospheric aqueous phases (e.g., thin films or microdroplets with dynamic pH and ionic strength). Please also see our response to the 1st major comment and the corresponding revisions. As stated in the manuscript (**Lines 325-328, Page 14**), the dialysis experiment primarily serves to reveal the chemical mixing state and removal potential of coatings, not to precisely simulate in-cloud kinetics.*

*We have noted that a kinetic model would be valuable for direct extrapolation to atmospheric timescales of in-atmosphere dissolution kinetics. However, our findings remain relevant. For surface-controlled dissolution processes (Laanait et al., 2015), the presence of such thin coatings on fine particles enhances 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 (discussed in **Lines 314-324, Page 14**).*

5. Climate and ocean-chemistry implications are extrapolated beyond the demonstrated chain. The manuscript links higher fractions of water-soluble Ca-bearing coatings to enhanced acid neutralization and mitigation of ocean acidification. To support this quantitatively, one typically needs (i) transport/deposition fractions to relevant ocean regions, (ii) chemical evolution during transport (e.g., re-precipitation), and (iii) a mass-balance for alkalinity delivery. Without that chain, these implications should be framed as qualitative hypotheses rather than established consequences.

Response:

*Our study provides observational constraints on the abundance of water-soluble Ca-bearing coatings on fine particles from two Asian desert sources. In the revision, we have revised the manuscript to limit the discussion to the scope of these results, removing speculative statements about enhanced acid neutralization and ocean acidification mitigation. The results are now framed as contributing to a qualitative, single-particle-level understanding of the physicochemical properties of freshly emitted mineral dust (**Line 20-22, Page 1; Line 321-324, Page 14; Line 373-376, Page 15**).*

6. Limited replication reduces representativeness and uncertainty quantification. Only four source-soil samples are analyzed, and the text notes that the time-intensive method precludes meaningful standard-deviation assessment. Accordingly, region-wide generalizations (e.g., “Asian dust source regions” broadly) should be tempered, and key reported fractions should be accompanied by uncertainty ranges (instrumental +

sampling + classification).

Response:

We agree that limited replication constrains regional generalization. In the revision, we have revised the text throughout, replacing broad phrases like "Asian dust source regions" with precise descriptors such as "two deserts" or "two Asian deserts" (e.g., ***Lines 15, 22 on Page 1; Line 175 on Page 6; Line 371 on Page 15****).*

*The key uncertainty in our particle classification stems from three main sources: (i) spatial heterogeneity during field soil collection, (ii) variability in generating surrogate dust aerosols in the lab, and (iii) random error in CCSEM/EDX elemental quantification. To provide a measure of analytical consistency, we performed repeated analyses on two additional dust samples (separate from the four main samples). We reported the detection rates for Si and Ca across these repeats as an indicator of method repeatability (****Lines 196-200, Page 7****). However, we acknowledge this does not constitute a full uncertainty budget spanning sampling and preparation (due to limited samples). Thus, we present the reported fractions (e.g., 56.9–88.2%) as observed ranges from our specific samples and have framed our conclusions to highlight the single-particle insights from these sites, rather than making statistical claims about broader regions.*

7. Minor technical and reporting inconsistencies that affect quantitative clarity. Examples include:

- Roundness and form-factor definitions depend on chosen diameter conventions; using D_{max} can bias shape metrics relative to an equivalent-area diameter.

Response:

*We have revised the calculation of particle roundness (****Line 143-145, Page 5****). In Formula (1) and its corresponding* ***Fig. S5****, D_{max} has been replaced with the average Feret diameter (D_{avg}) to avoid potential bias. Formula (2) for the form factor remains unchanged as it does not involve any diameter term.*

- Mixed or shifting units for fluxes between text and tables can confuse interpretation (ensure consistent units throughout).

Response:

We have revised the manuscript to ensure terminological and unit consistency *throughout the text. For clarity, the respective average values for particle number flux and mass flux have been incorporated into **Table 1**.*

- Histogram/binning normalization and axis labeling should be explicit (linear vs log bins; number vs number density).

Response:

*Consistent binning normalization and axis labeling schemes were applied uniformly to **Figs. 3, S4, S5**, and **S6**.*

- Uncertainty propagation from (i) ellipsoid volume assumption, (ii) density inference from composition, and (iii) EDX semi-quantification should be shown (even as a sensitivity analysis).

Response:

*In the revision, **Table S4** was added into the Supplementary Materials. The comparison between the total particle mass obtained via CCSEM and the gravimetric method reveals the propagated uncertainty associated with the key assumption of ellipsoid volume, density inference from composition, and EDX semi-quantification.*

Updated Figures

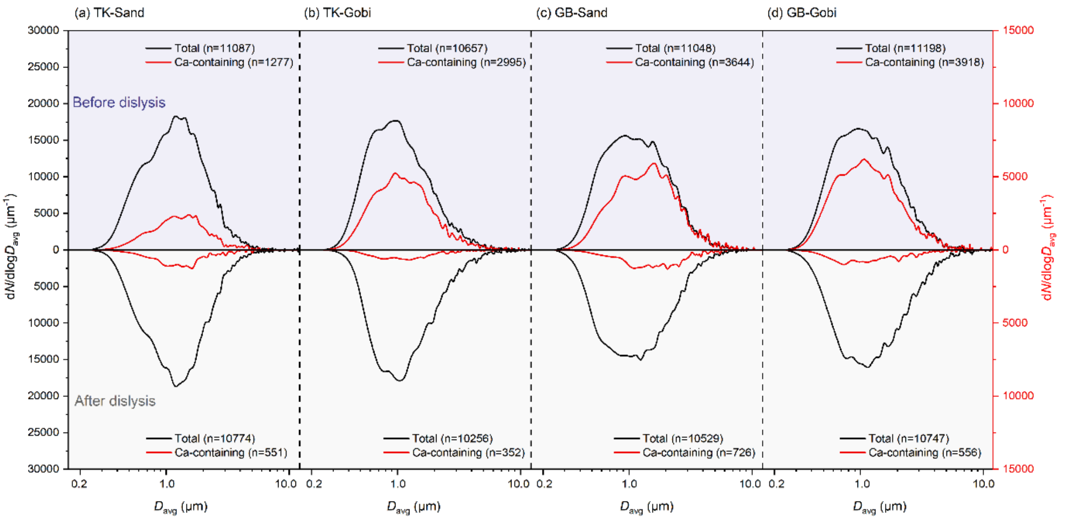


Figure 3. Particle number size distributions of Ca-containing and total dust particles generated from sandy (Sand) and gravel (Gobi) surface soils of the Taklimakan (TK) and Gobi (GB) Desert.

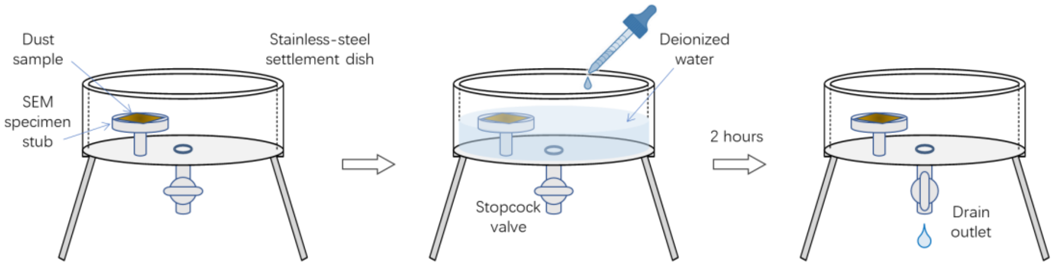


Figure S2. Schematic of water dialysis experiment.

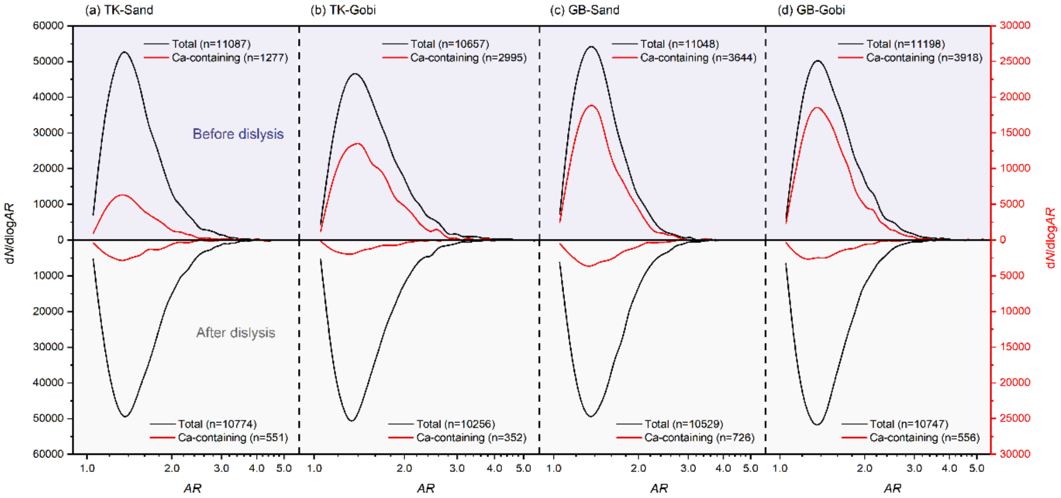


Figure S4. Particle number distributions of Ca-containing and total dust particles as a function of aspect ratio (AR), from sandy (Sand) and gravel (Gobi) surfaces of the Taklimakan (TK) and Gobi (GB) Desert.

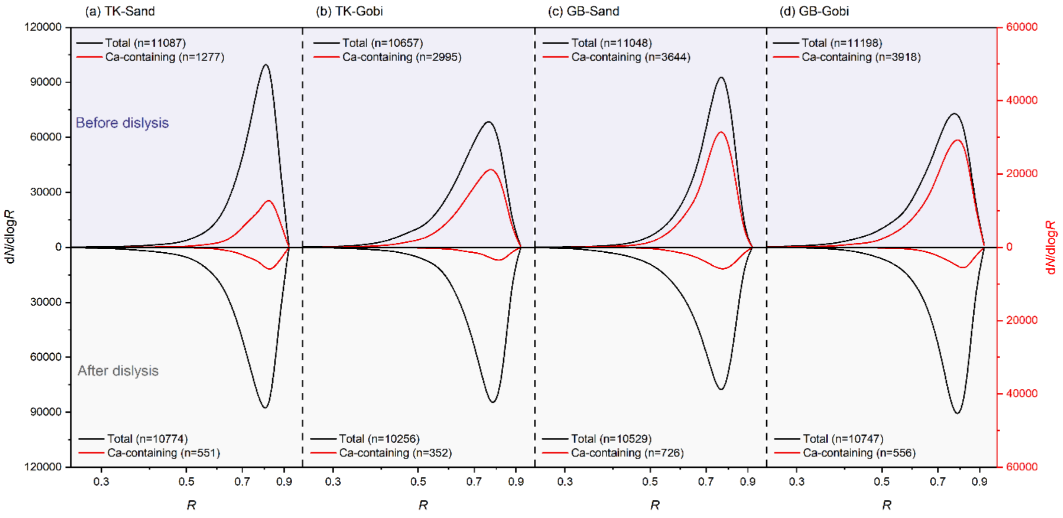


Figure S5. Particle number distributions of Ca-containing and total dust particles as a function of roundness (R), from sandy (Sand) and gravel (Gobi) surface soils of the Taklimakan (TK) and Gobi (GB) Desert.

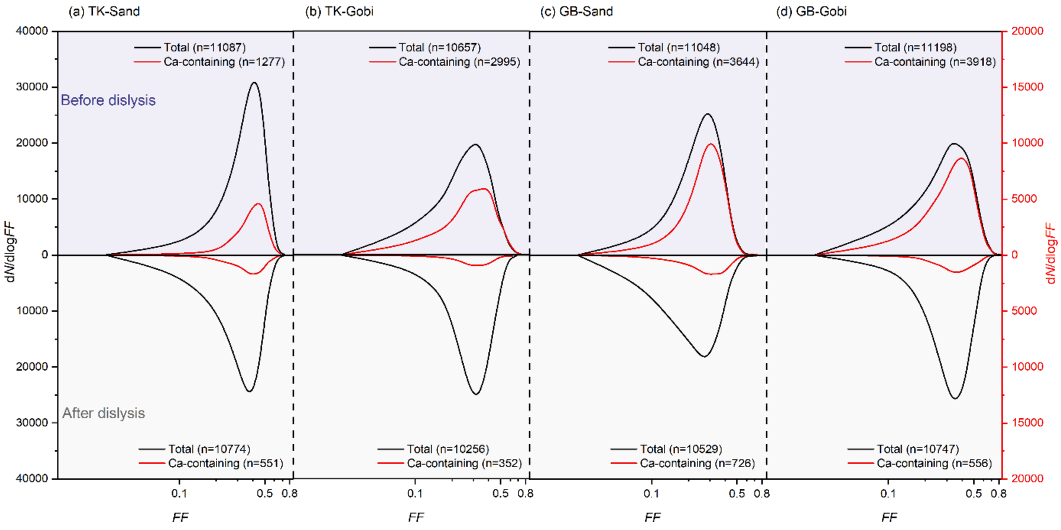


Figure S6. Particle number distributions of Ca-containing and total dust particles as a function of form factor (FF), from sandy (Sand) and gravel (Gobi) surface soils of the Taklimakan (TK) and Gobi (GB) Desert.

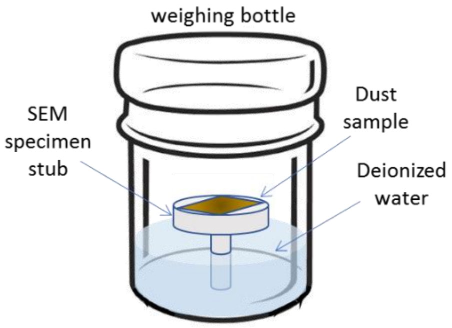
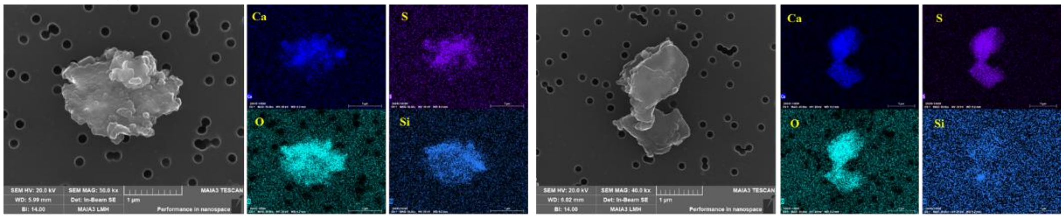


Figure S8. Schematic of saturated water-vapor exposure experiment

(A) Before exposure



(B) After exposure

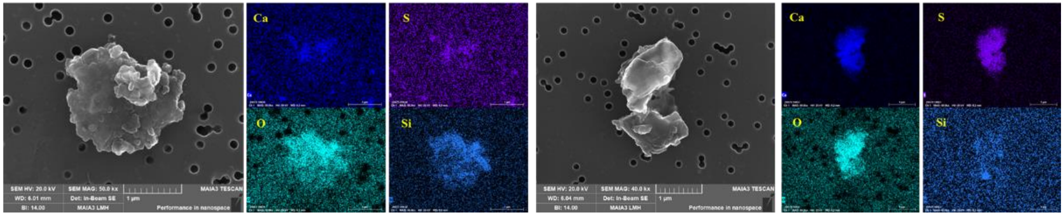
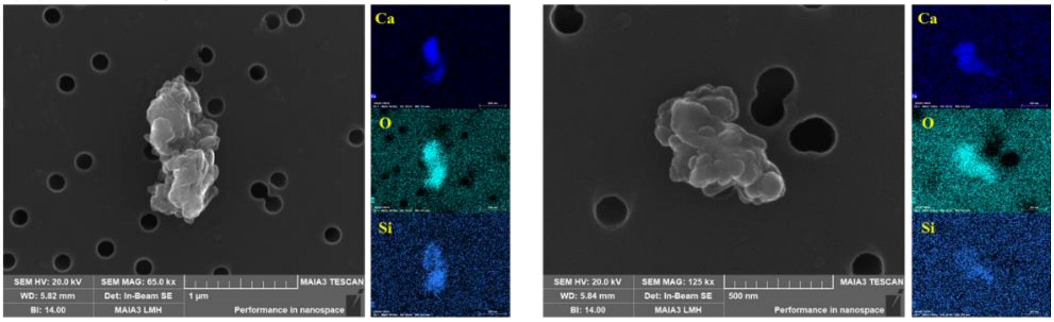


Figure S9. Morphological and elemental-distribution alteration of Ca-S-containing particles after saturated water vapor exposure.

(A) Before exposure



(B) After exposure

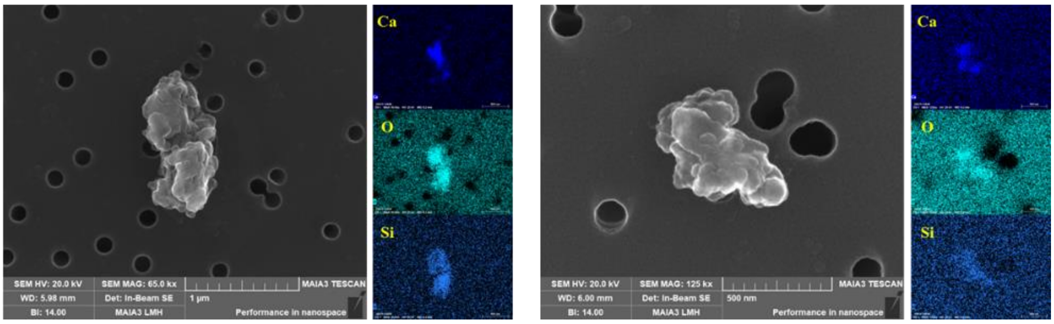


Figure S10. Elemental-distribution alteration of Ca-O-rich particles after saturated water vapor exposure without obvious morphological change.

Updated Tables

Table 1. Number and mass emission fluxes of total mineral dust and of water-soluble calcium-containing particles/components generated via saltation-sandblasting processes.

	the Taklimakan Desert			**the Gobi deserts**		
	Sand dune	Gravel soil	Mean	Sand dune	Gravel soil	Mean
Total dust particles						
Number flux (×10^6 particles m^{-2} s^{-1})	7.6	2.4	5.0	3.4	21.0	12.2
Mass flux (μg m^{-2} s^{-1})	56.4	16.6	36.5	28.6	165.6	97.1
Water-soluble Ca-containing particles/components						
Number flux (×10^5 particles m^{-2} s^{-1})	4.3	5.6	5.0	8.5	63.0	35.8
Mass flux (μg m^{-2} s^{-1})	0.2	0.6	0.4	0.4	6.0	3.2

Table S4. Inter-method comparison of dust mass quantified by computer-controlled scanning electron microscopy (CCSEM) and gravimetric analysis.

Sample	1	2	3	4	5	6	7	8	9	Mean ± S.D.
$mass_{\mathrm{Gravi}}$ (μg)	84	138	202	112	348	135	120	143	127	157 ± 78
$mass_{\mathrm{CCSEM}}$ (μg)	68	89	134	48	111	57	77	68	81	82 ± 27
Deviation (%)	–18.9	–35.4	–33.5	–57.0	–68.1	–57.7	–35.7	–52.6	–35.8	– (43.9 ± 15.7)

Note: $mass_{\mathrm{Gravi}}$ represents the dust mass loading on quartz filters determined by gravimetric analysis using a Sartorius MC5 microbalance (±1 μg sensitivity). $mass_{\mathrm{CCSEM}}$ represents the dust mass loading on polycarbonate filters obtained from CCSEM analysis. The percentage deviation is calculated as: Deviation (%) = $[(mass_{\mathrm{CCSEM}} - mass_{\mathrm{Gravi}}) / mass_{\mathrm{Gravi}}] \times 100$. S.D. represents the standard deviation.

Table S5. Procedural blank contamination levels of water-soluble Ca and total dust particles in sand dune and gravel soil samples.

Blank	water-soluble Ca number flux (particles m^{-2} s^{-1})	water-soluble Ca mass flux (μg m^{-2} s^{-1})	total number flux (particles m^{-2} s^{-1})	total mass flux (μg m^{-2} s^{-1})
Sand dune	7.3×10^{4}	0.06	9.5×10^{4}	0.6
Gravel soil	8.6×10^{4}	0.03	2.2×10^{5}	0.9
Mean	7.9×10^{4}	0.05	1.6×10^{5}	0.7

References

Frear, G. L., & Johnston, J.: The solubility of calcium carbonate (calcite) in certain aqueous solutions at 25. Journal of the American Chemical Society, 51(7), 2082-2093, https://doi.org/10.1021/ja01382a014, 1929.

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Laanait, N., Callagon, E. B. R., Zhang, Z., Sturchio, N. C., Lee, S. S., and Fenter, P.: X-ray–driven reaction front dynamics at calcite-water interfaces, Science, 349, 1330-1334, https://doi.org/10.1126/science.aab3272, 2015.

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.*