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Abundant water-soluble calcium coatings on fine Asian dust particles

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Abstract. The dissolution behavior of atmospheric calcium (Ca) mineral dust released from arid regions and their climate impacts via buffering effects are highly dependent on their size-resolved mineralogical composition. Due to the inherent complexity of mineral dust, tracing the chemical forms and mixing states of Ca minerals at single-particle level remains challenging. In this study, an automated microanalysis technique was employed to characterize the physicochemical properties of 43,990 individual mineral dust particles generated by saltation-sandblasting processes in two typical Asian dust source regions, along with their residual 42,306 particles after water dialysis. Both the total dust and the Ca-containing particles exhibited a modal peak in the submicron size range, before and after dialysis. After dialysis, 56.9 % to 88.2 % (by number) of the calcium-containing dust particles lost their soluble calcium components. These water-soluble constituents accounted for 19.6–41.9 % of the mass of calcium-containing particles in both the Taklimakan and Gobi deserts. In addition, more than 73.0 % of Ca-O-rich and Ca-S-containing particles occurred as surface coatings on other minerals and were effectively removed by water dialysis. 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.

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1 Introduction

Mineral dust represents a major component of atmospheric aerosol, accounting for up to 75 % of the global aerosol mass (Choobari et al., 2014). 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). Through direct effects, mineral dust perturbs Earth's radiative balance (Choobari et al., 2014; Adebiyi et al., 2023) primarily via scattering (Nousiainen, 2009), though it also exhibits considerable absorption depending on its mineral composition and mixing state (Falkovich et al., 2001; Formenti et al., 2011). Through indirect mechanisms, mineral dust acts as ice-nucleating particles (INPs) and cloud condensation nuclei (CCN) (Schepanski, 2018), thereby modifying cloud properties, albedo, and precipitation patterns (Engelbrecht and Derbyshire, 2010; Pye, 2015). The deposition of mineral dust onto terrestrial and marine ecosystems supplies essential micronutrients, stimulating primary productivity and affecting both ecological dynamics and marine biogeochemistry (Quigg, 2016; Mahowald et al., 2018). The transport distance, transformation, and environmental impacts of atmospheric mineral dust are strongly governed by its size distribution, chemical composition, and mixing state (Knippertz and Stuut, 2014). Although the fine fraction (diameter < 2.5 μm) constitutes only 5–20 % of emitted dust mass (Formenti et al., 2011; Choobari et al., 2014), it disproportionately contributes to the global particle population (Mahowald et al., 2014) and dominates long-range transport due to its prolonged atmospheric lifetime (Uno et al., 2009; Pye, 2015). A major emission mechanism is saltation-driven sandblasting (Shao et al., 1993), in which sand-sized particles (50–500 μm) bombard the surface, releasing fine dust even at wind speeds below the direct entrainment threshold (Alfaro, 2008; 2022). This process amplifies dust emission fluxes by 3–10 times (Parajuli et al., 2016) and is responsible for over 75 % of global fine dust emissions (Grini and Zender, 2004). Crucially, sandblasting largely modifies the dust's physical and chemical characteristics compared to the parent soil. It enriches the fine mode (Alfaro et al., 1997; Grini et al., 2002) and preferentially liberates soluble salts and clay coatings from soil aggregates, thereby elevating water-soluble ions (e.g., Ca^{2+}) in the aerosol (Wu et al., 2022). The resulting size distribution governs atmospheric transport duration and influences each mineral's global distribution (Panta et al., 2022), while chemical alterations enhance dust hygroscopicity, reactivity, and radiative efficiency, intensifying its role in climate and biogeochemical cycles.

Among the various chemical components of dust, atmospheric calcium (Ca) predominantly originates from soil dust particles and sea salts. Its buffering effects in the atmosphere and carbon dioxide fixation in the ocean can modify acidity across multi spheres of the earth (Tipper et al., 2016). Common Ca-containing minerals in natural dust such as calcite ($CaCO_3$) can rapidly neutralize acidic species in precipitation or upon deposition in terrestrial and marine environments (Cao et al., 2005). However, the buffering capacity of these particles is not uniform and is intrinsically governed by their dissolution behavior, which varies with particle size and heterogeneity. Evidence from multiple research approaches highlights the critical importance of size-resolved mineralogical composition: it has been shown to govern atmospheric acidity in trend analyses

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(Ren et al., 2011), to modify CCN-activity in laboratory experiments (Sullivan et al., 2009), and to regulate marine alkalinity in surface waters through field observations (Carter et al., 2014; Su et al., 2020). Moreover, the apparent hygroscopicity of calcium-rich dust is controlled by its chemical mixing state, a property determined by source mineralogy and chemical transformations during atmospheric transport (Sullivan et al., 2009). Following deposition, the dissolution behavior of atmospheric calcium carbonate remains sensitive to the physical and mineralogical characteristics of the dust, as observed both regionally along oceanic transects (Feely et al., 2002) and at a global scale (Sulpis et al., 2021). Therefore, the size-resolved mineralogical composition of atmospheric dust serves as a key link between continental dust sources and global climate feedbacks via the calcium cycle, a claim supported by both modern (Maher et al., 2010) and paleoclimate studies (Tipper et al., 2016).

The heterogeneous chemistry of individual dust particles, which is essential to their atmospheric processing and dissolution behavior, varies significantly with source region (Krueger et al., 2004). A particle's ability to uptake sufficient water to activate into a cloud droplet is governed primarily by its size and soluble hygroscopic content (McFiggans et al., 2006). Modern analytical techniques enable direct observation of atmospherically-processed calcium carbonate dust particles, revealing complex single-particle morphologies and reaction pathways (Laskin et al., 2005). Laboratory studies further indicate that the hygroscopic behavior of specific calcium salts reflects complex internal mixtures (Guo et al., 2019), which control the dissolution kinetics and acid-buffering efficiency of dust in the atmosphere and after deposition into aquatic environments (Usher et al., 2003). A mechanistic understanding of these micro-scale processes is critical for accurately modelling the global calcium cycle (Fantle and Tipper, 2014; Gussone et al., 2016) and dust's influence on the carbon cycle (Jickells et al., 2014; Steiner et al., 2021). However, tracing the chemical forms and mixing states of calcium-containing dust at the single-particle level remains challenging, as these properties evolve continuously during atmospheric transport.

Therefore, accurately predicting the climate impacts of realistic calcium-containing mineral dust aerosols requires detailed data on their size-resolved chemical composition and mixing state, as well as a fundamental understanding of compound–water interactions. In this study, laboratory-generated mineral dust particles produced by the saltation-sandblasting mechanism were collected and subjected to water dialysis treatment. Using computer-controlled scanning electron microscopy (CCSEM), the same set of individual dust particles was analyzed before and after water dialysis, enabling estimation of the number emission flux of calcium-containing particles and the mass of water-soluble calcium components. Changes in the particle size distribution, shape factors, and mixing states of all calcite- and gypsum-containing particles were quantified. The resulting emission flux and mixing state data provide key insights for assessing the potential climate and carbon cycle impacts of these mineral dust aerosols.

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2 Methodology

2.1 Sample Collection

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). The Taklimakan Desert is located in the Tarim Basin and covers an area of more than 337,000 km^2, about 85 % of which is shifting sand dunes in the central area of the basin, surrounded by a zone of gravels called the gobi belt (El-Baz, 1984; Sun and Liu, 2006). The Alashan (Alaxa) Plateau is located in the west part of Inner Mongolia, with the Badanjilin (Badain Jaran) Desert (49,000 km^2) in the mid-west of the plateau, the Tenggeli (Tengger) Desert (42,700 km^2) in the south, and the Wulanbuhe (Ulan Buh) Desert (10,000 km^2) in the south. Those Gobi deserts on the Alashan Plateau are considered as important mineral dust emission source regions in arid and semiarid China (Shao and Dong, 2006; Wang et al., 2008). Detailed information on the locations and surrounding geomorphology of the sampling sites is listed in Table S1. 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.

2.2 Laboratory Dust Generation

Mineral dust aerosol particles were generated from soil samples by means of a dust resuspension chamber system. The system generates dust particles from the desert surface soils based on the mechanism of dust saltation and sandblasting processes (Wu et al., 2022), which showed a consistent trend in particle size distribution and chemical composition with field dust aerosols (Wu et al., 2023). Dust particles were mobilized by a rotating annular blade in the chamber of the system, with an equivalent friction velocity of 0.54 m s^{-1} (Etyemezian et al., 2007), and the resuspended particles were led out into a cylindrical sampling cavity. Total suspended particulate (TSP) samples were collected through the sampling port of the cavity onto 47 mm polycarbonate filters (0.2 μm in porosity, Whatman International Ltd., Maidstone, UK) using a portable aerosol sampler (Model mini-Vol, Airmetrics Corp., Springfield, OR, USA). The operating flow rate of sampling was 5 L min^{-1}. The sampling duration for each sample varied between 1 and 5 minutes to minimize particle overlap on the filters, depending on real-time TSP mass concentrations measured using a DustTrak aerosol monitor (Model 8530, TSI Inc., Shoreview, MN, USA). After sampling, each filter was placed in a plastic cassette and stored at 5 °C until analysis.

2.3 Microanalysis and water dialysis

A rectangular section with the size of 10 mm × 10 mm was cut from the central area of the filter by a stainless-steel scissor and mounted onto an aluminum stub with conductive adhesive carbon tape for CCSEM analysis and water dialysis process. Before microanalysis, the samples were coated with carbon for particle conductivity. For each sample, all individual mineral

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dust particles dispersed in over 100 fields of view (100 μm × 100 μm for each field, aligned in a rectangular array) were physicochemically characterized one by one using a field emission scanning electron microscope (MAIA3, Tescan, Brno, Czech Republic) equipped with two energy dispersive X-ray (EDX) spectrometer (Xflash 6-60, Bruker, Karlsruhe, Germany). Automated microanalysis was performed by CCSEM analysis software (IntelliSEM, RJ Lee Group, Inc., Monroeville, USA) to obtain particle image, size distribution, morphological parameter (e.g., aspect ratio, roundness, and form factor), and elemental compositions. The quantitative analysis by the CCSEM provided reproducible sizing and identification of individual particles based on large numbers (> 10,000 in this study) for statistical counting (Mamane et al., 2001; Castillo et al., 2019).

Following microanalysis, the stubs were transferred to a stainless-steel settlement dish (Okenshoji Co. Ltd.). Dialysis with Milli-Q water was then conducted following the method of Zhang and Iwasaka (2004) to remove water-soluble components from the dust particles without disturbing their original locations. The CCSEM system then relocated the same set of over 100 pre-analyzed fields of view and performed automated microanalysis under consistent operating conditions (Fig. S2). This ensured that all particles analyzed post-dialysis were identical to those characterized before-dialysis, guaranteeing the comparability of measurements. The physical (size and morphology) and chemical (elemental compositions) properties of the dust particles were compared to confirm their alteration in shape and composition after dialysis. In addition to the automated microanalysis, manually operated EDX mapping was performed on all individual calcite and gypsum-like particles to compare their mixing states.

Particle diameters were measured using a rotated-feret box technique (ASTM F1877-16), with a box or a caliper being clockwisely rotated around the particles and particle length being recorded at each orientation. Average diameter (D_{avg}) is the arithmetic average of all the 90 measurements, and the maximum diameter (D_{max}) and the minimum diameter (D_{min}) are the largest and smallest of all the measurements, respectively. The aspect ratio (AR) of the particle is the ratio of D_{max} to its perpendicular width. The roundness (R) of a particle is a measure of how closely a particle resembles a circle and has a value between 0 and 1, where the R of a perfectly circular particle is 1. Roundness was calculated using the following formula:

$$R = \frac{4A}{\pi D_{max}^2}, \quad (1)$$

where A is the projected area of the particle. The form factor (FF), which is a dimensionless number sensitive to the irregularities in particle edges, i.e., the variations in roughness of a particle outline, was calculated using

$$FF = \frac{4\pi A}{p^2}, \quad (2)$$

where p is the perimeter of the particle outline.

For the quantitative elemental analysis, the weight percentages of elements from carbon (C) to lead (Pb) were determined based on their characteristic X-ray signals, which were fitted to Gaussian distributions (Okada and Kai, 2004). The relative weight percentage of each element (Z > 6, Carbon) in a particle is the ratio of its peak area to the total peak areas of all detected elements. Individual mineral dust particles were classified into several mineralogical categories according to how

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the particle's elemental composition followed the empirical formula of a specific mineral (Hu et al., 2022). The particle classification rules are listed in Table S2. The empirical formula and elemental composition information are available in the handbook of mineralogy (Cook, 2001) and several online databases at https://duffy.princeton.edu/mineralogy-and-crystallography-databases.

Assuming an ellipsoidal geometry, the volume of each particle was calculated from its measured major (D_{max}) and minor (D_{min}) axial diameters. The particle mass (m) was subsequently determined as the product of its volume and its estimated density (ρ), where ρ was inferred from the particle's EDX elemental composition (Ault et al., 2012). The number of water-soluble Ca-containing particles ($N_{sol\text{-}Ca}$) in a mineral dust sample was quantified as the difference between the numbers of Ca-containing particles before and after dialysis. The mass of water-soluble Ca ($mass_{sol\text{-}Ca}$) in a mineral dust sample was quantified as the difference between the total mass of elemental Ca before and after dialysis. Consequently, the emission flux of water-soluble Ca from mineral dust generated via saltation-sandblasting processes is given by:

$$F_{\mathrm{sol-Ca}(N)} = \frac{N_{\mathrm{sol-Ca}}}{(v \times t)} \times \frac{A_{\mathrm{filter}}}{A_{\mathrm{view}}} \times \frac{Q}{A_{\mathrm{chamber}}} \quad (3)$$

$$F_{\mathrm{sol-Ca}(mass)} = \frac{mass_{\mathrm{sol-Ca}}}{(v \times t)} \times \frac{A_{\mathrm{filter}}}{A_{\mathrm{view}}} \times \frac{Q}{A_{\mathrm{chamber}}} \quad (4)$$

Where, $F_{sol\text{-}Ca(N)}$ is the emission flux of water-soluble Ca-containing particles from Asian Dust source regions via saltation-sandblasting processes (unit: particles m^{-2} d^{-1}), $F_{sol\text{-}Ca(mass)}$ is the emission flux of water-soluble Ca (μg m^{-2} d^{-1}), is the air flow rate of the sampler (5 L min^{-1}), t is the sampling duration for the resuspension samples (min), A_{filter} is the total area of the sampling filter (a fixed area of 11.34 cm^2 in this work), A_{view} is the total area analyzed by CCSEM on the sampling filter (mm^2), Q is the exhaust airflow rate from the resuspension chamber (a fixed rate of 250 L min^{-1}), and $A_{chamber}$ is the bottom area of the resuspension chamber (a fixed area of 0.255 m^2).

2.4 Quality control and assurance

The laboratory dust generation processes and SEM-EDX analyses were conducted under consistent operating conditions for all samples, using an accelerating voltage of 20 kV and a working distance of ~8 mm in the Analysis mode. Only particles with diameters larger than 0.2 μm were measured for size distribution and particle classification due to the 0.2 μm pore size of the sampling filters. 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. Field blanks showed minimal contamination, with fewer than 20 particles detected per filter. 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

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number; A: mass absorption; F: fluorescence). An estimated error margin of 1.0 wt % was used in the semi-quantitative analysis of elemental contents for particle classification. The mineralogical categorization in this study was determined by comparing the elemental composition of individual particles to the key elements in the empirical formulas of reference minerals. However, since natural particles often exhibit potential impurities, amorphous phases, or solid solutions, those classified under category X are more precisely termed X-like particles to reflect their non-ideal match to the reference mineral. Moreover, aggregated particles with their elemental compositions deviated largely from the formulas were classified into the 'silicate' or 'aluminosilicate' groups.

The detection precision of CCSEM for Si- and Ca-containing particles was evaluated through repeat measurements of two laboratory-generated mineral dust samples from the Taklimakan Desert (sand dune surface soil and gravel soil). Under consistent resuspension and microanalysis conditions, measurements were performed in triplicate on both identical and separate fields of view. Analysis of 11,000 particles per measurement confirmed reproducible detection rates for both particle types (Table S3).

2.5 Advantages and limitations of methodological approaches

The automated microanalysis performed using a CCSEM system enabled the relocation of all view fields analyzed before dialysis and the measurement of all residual particles within these areas after dialysis. This process allowed quantification of the size distribution and elemental composition of the same set of dust particles on a single-particle basis, with each sample containing over 10,000 particles. By applying the same CCSEM operating conditions used prior to dialysis, the number of water-soluble Ca-containing particles and the mass of soluble calcium components were estimated based on several assumptions regarding particle shape and density. However, these assumptions, such as regarding particles as ellipsoids and assigning densities based on elemental composition, may introduce uncertainties due to the inherent physicochemical complexity of mineral dust particles in environmental samples.

It is also important to note a key limitation of the EDX-based CCSEM technique: its inability to determine chemical bonding. Consequently, the identification of calcite ($CaCO_3$) solely based on the detection of Ca–O-rich particles, where the combined weight percentage of Ca and O exceeds 99.0 wt %, is subject to uncertainty. This approach cannot exclude the potential presence of other Ca–O-rich phases such as calcium oxide (CaO), calcium hydroxide ($Ca(OH)_2$), and calcium nitrate ($Ca(NO_3)_2$, all of which exhibit EDX spectra similar to that of calcite. It is worth noting, however, that CaO tends to react with atmospheric moisture, forming $Ca(OH)_2$, which can further react with CO_2 to yield amorphous calcium carbonate (Barker, 1974; Kalinkin et al., 2005). In addition, $Ca(NO_3)_2$ is not commonly observed as a typical mineral in dust samples from source regions, as indicated by bulk measurements of calcium and nitrate among water-soluble ions (Wu et al., 2022) and downwind individual particle analysis (Laskin et al., 2005). Under these considerations, calcite is generally regarded as the most probable source mineral (Fitzgerald et al., 2015; Panta et al., 2022).

At the same time, while it is well established that atmospheric carbon dioxide dissolves in cloud droplets, reacts to form carbonic acid, and thereby lowers the pH of cloud water (Chen and Lu, 2003), the distribution of pH across individual cloud

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droplets under real atmospheric conditions remains unclear. Studies have reported that the pH of individual cloud droplets can vary widely (Warneck, 1986). In our experiments, all dust particles were subjected to a uniform 2-hour water dialysis. The dialysis utilized freshly-prepared Milli-Q deionized water that had been pre-equilibrated in open air at 25 ± 5 °C for 48 hours, yielding an average pH of 6.40 ± 0.71. This condition only represented a specific average profile of dust dissolution behavior in cloud water.

Finally, the current methodology remains time-intensive, requiring tens of hours per sample for the relocation of calcite and gypsum particles and the visual identification of their mixing states. Consequently, only four samples were analyzed, which precludes any meaningful assessment of standard deviation across individual environmental samples.

3 Results and Discussions

3.1 Emission fluxes of soluble Ca-containing particles

From a single-particle perspective, the total numbers of dust particle decreased slightly across all samples: by 2.8 % and 3.8 % for dust from sandy and gravel surfaces in the Taklimakan Desert, and by 4.7 % and 4.0 % for sandy and gravel dust in the Gobi Desert, respectively. Notably, 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.

Dust aerosols released from gravel surfaces in both deserts contained fewer particles with insoluble Ca components compared to those from sandy surfaces. While the proportion of particles containing water-soluble calcium from sandy surfaces ranged from 56.9 % in the Taklimakan Desert to 80.1 % in the Gobi Desert, gravel surfaces released even higher percentages of such particles, including 88.2 % and 85.8 % in the Taklimakan and Gobi Deserts, respectively (Fig. 1a). After the removal of water-soluble calcium, the remaining insoluble Ca-containing particles exhibited a lower average mass per particle. Nevertheless, the mass of individual Ca-containing particles still spanned 4–5 orders of magnitude (Fig. 1b), indicating distinct mineralogical compositions between water-soluble calcium compounds and insoluble calcium minerals. This interpretation is further supported by particle volume analysis, which revealed a slight reduction in average volume after dialysis, along with a markedly narrower distribution of single-particle volumes in both deserts (Fig. 1c).

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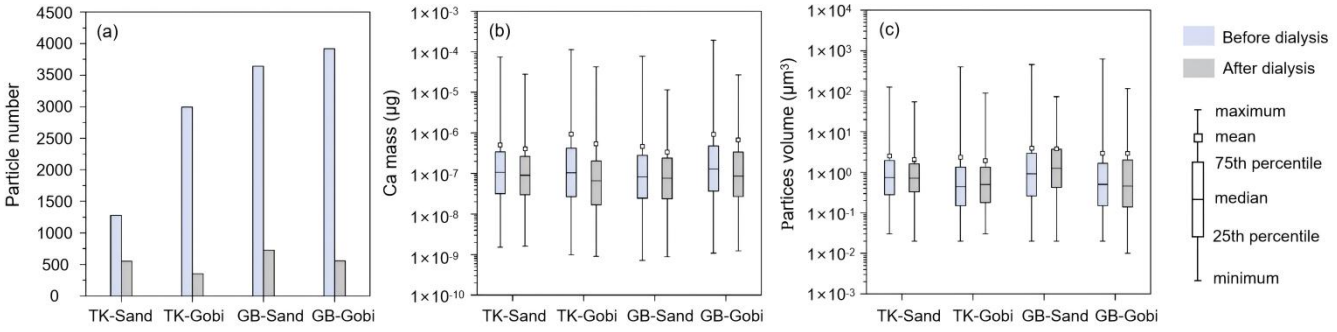

Figure 1. Comparison of total and insoluble Ca-containing single particles before and after water dialysis. (a) Number of Ca-containing particles; (b) Mass of elemental Ca per particle (μg); (c) Volume of Ca-containing particles (μm^3).

The emission fluxes of Ca-containing particles (Table 1) were estimated based on the measured number concentrations and elemental composition of mineral dust particles, combined with the dust generation operating conditions and aerosol sampling parameters. A friction velocity of 0.54 m s^{-1} was used to simulate dust mobilization from surface soil via the saltation-sandblasting mechanism. By considering the bottom area of the resuspension chamber and the exhaust airflow rate, the estimated emission fluxes of dust particles containing water-soluble Ca reached average values of 5.8×10^5 particles m^{-2} s^{-1} for the Taklimakan Desert and 3.6×10^6 particles m^{-2} s^{-1} for the Gobi Desert (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). This range is consistent with the previously reported calcium ion contents of 0.64–5.0 % in both generated dust and ambient dust in the Taklimakan Desert (Wu et al., 2022).

Table 1. Emission flux estimates of water-soluble Ca-containing dust particle numbers and their mass of soluble Ca components under the friction velocity of 0.54 m s^{-1}.

Surface soil	Taklimakan Desert		Gobi Desert	
	water-soluble Ca number flux (particles m^{-2} s^{-1})	water-soluble Ca mass flux (μg m^{-2} s^{-1})	water-soluble Ca number flux (particles m^{-2} s^{-1})	water-soluble Ca mass flux (μg m^{-2} s^{-1})
Sand dune	5.0×10^5	0.3	6.3×10^6	0.5
Gravel soil	6.5×10^5	0.6	9.2×10^5	6.0

Analysis of individual particles suggests an obvious mineralogical difference between water-soluble and insoluble Ca phases (Fig. 2). Particles with soluble Ca were concentrated in two types with the highest (40–60 wt %) and lowest (0.5–20 wt %) Ca content. The removal of these extreme-composition particles during dialysis, despite unchanged particle size mode,

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suggests the presence of both pure Ca minerals (high-Ca wt % group) and water-soluble Ca-compound coatings (low-Ca group) on the surface of other silicate or aluminosilicate minerals.

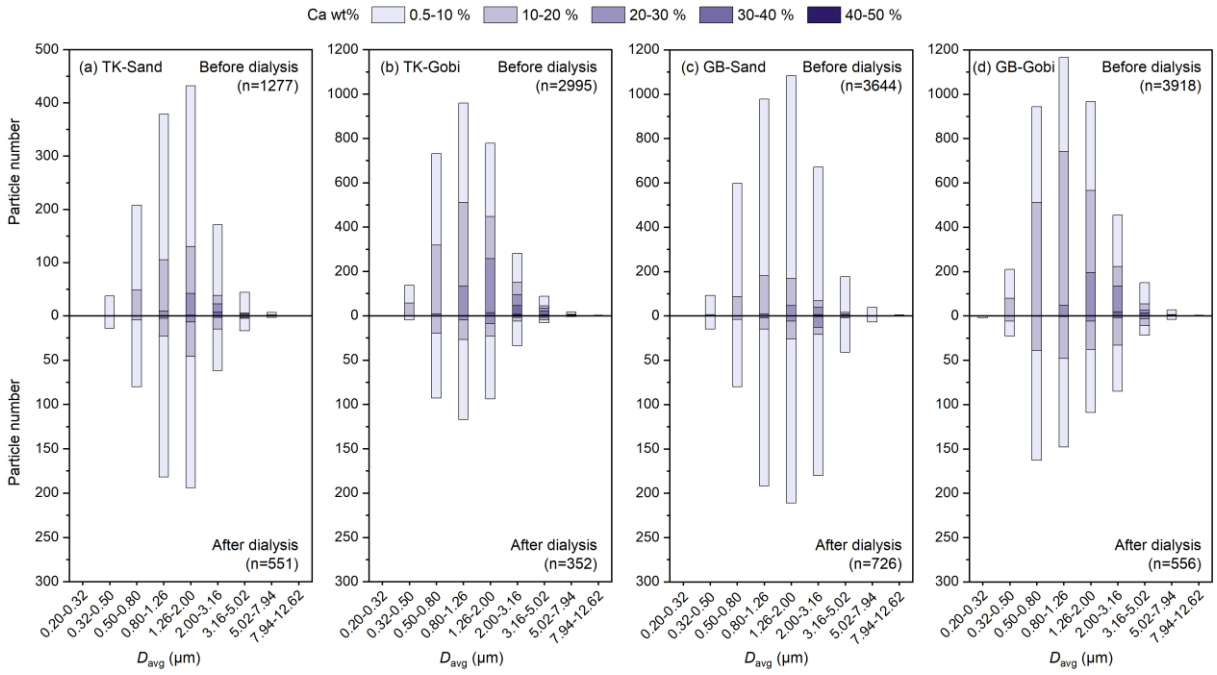

Figure 2. Relationship between particle size (μm) and calcium content (relative weight percentage in single particle, Ca wt %) for Ca-bearing particles before and after water dialysis.

3.2 Physicochemical properties of calcium particles

While post-dialysis size distributions (Fig. 3), elongation degree (Fig. S3), and roundness (Fig. S4) remained relatively consistent for both Ca-containing and total dust particles, their surface roughness exhibited obvious changes following the removal of water-soluble Ca compounds (Fig. S5). The resulting particles developed more irregular edges, supporting the idea that the dissolved components had formerly formed smooth coatings on the insoluble mineral surfaces.

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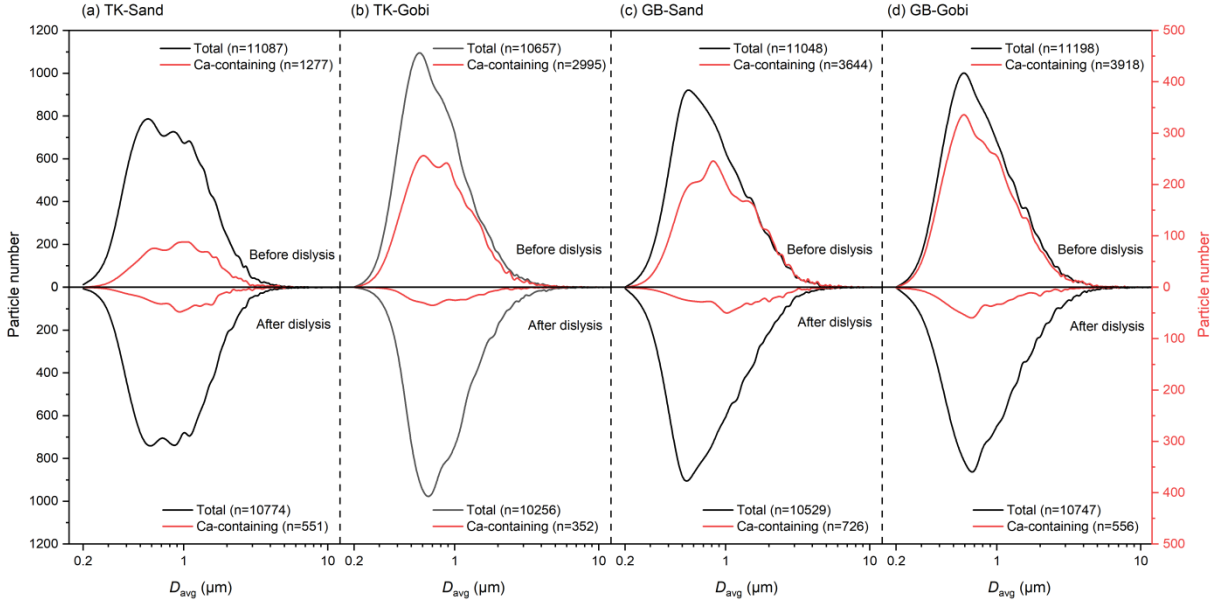

Figure 3. Particle number size distributions of Ca-containing and total dust particles generated from sandy (Sand) and gravel (Gobi) surfaces of the Taklimakan and Gobi Desert.

To further characterize the dissolution behavior of different minerals and their mixing states, individual Ca-containing dust particles was categorized into several mineralogical subgroups: clay minerals (e.g. smectite and vermiculite), feldspars (e.g. anorthite and albite), calcite, dolomite, gypsum, and others (comprising mixed silicates or aluminosilicates with elemental compositions not matching any specific mineral). The relative abundances of each mineral varied across dust source regions and surface soil types (Fig. 4). Post-dialysis, the lowest residual fractions of Ca-containing minerals were observed for calcite (ranging from 0.1 % to 0.7 % in total dust particles from the Taklimakan and Gobi Desert) and gypsum (0.0 % in all samples), suggesting them as the most soluble minerals.

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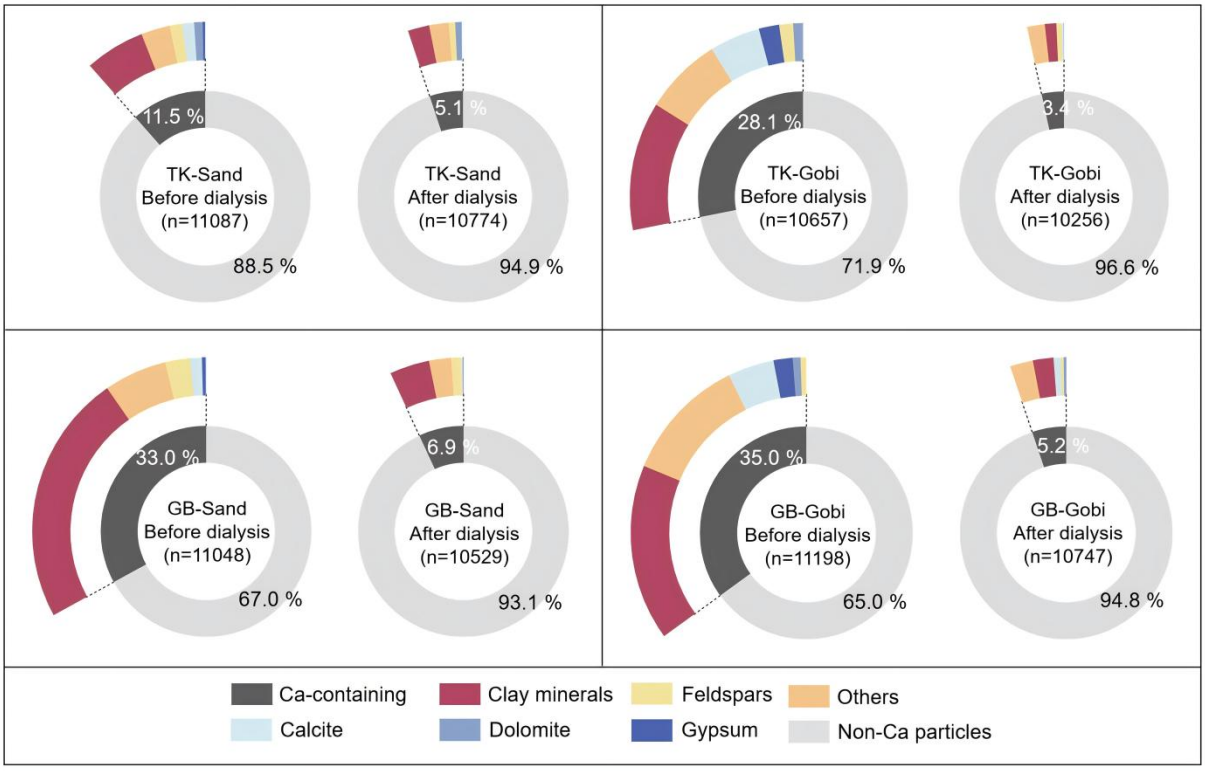

Figure 4. Relative number abundances of Ca-containing minerals before and after water dialysis. Dust samples were generated from sand dune (Sand) and gravel (Gobi) surface soils of the Taklimakan (TK) and the Gobi (GB) Desert.

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 (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 nanometers.

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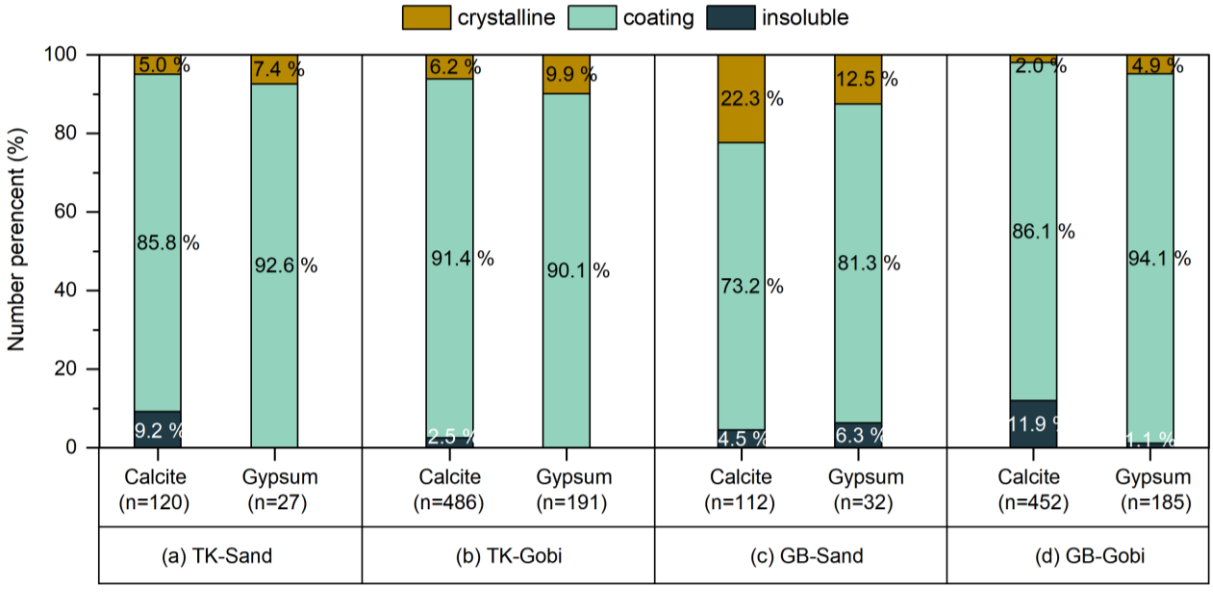

Figure 5. Relative particle number percentage of calcite and gypsum particles in different mixing states. The numbers in the paratheses are the counts of detected calcite and gypsum particles.

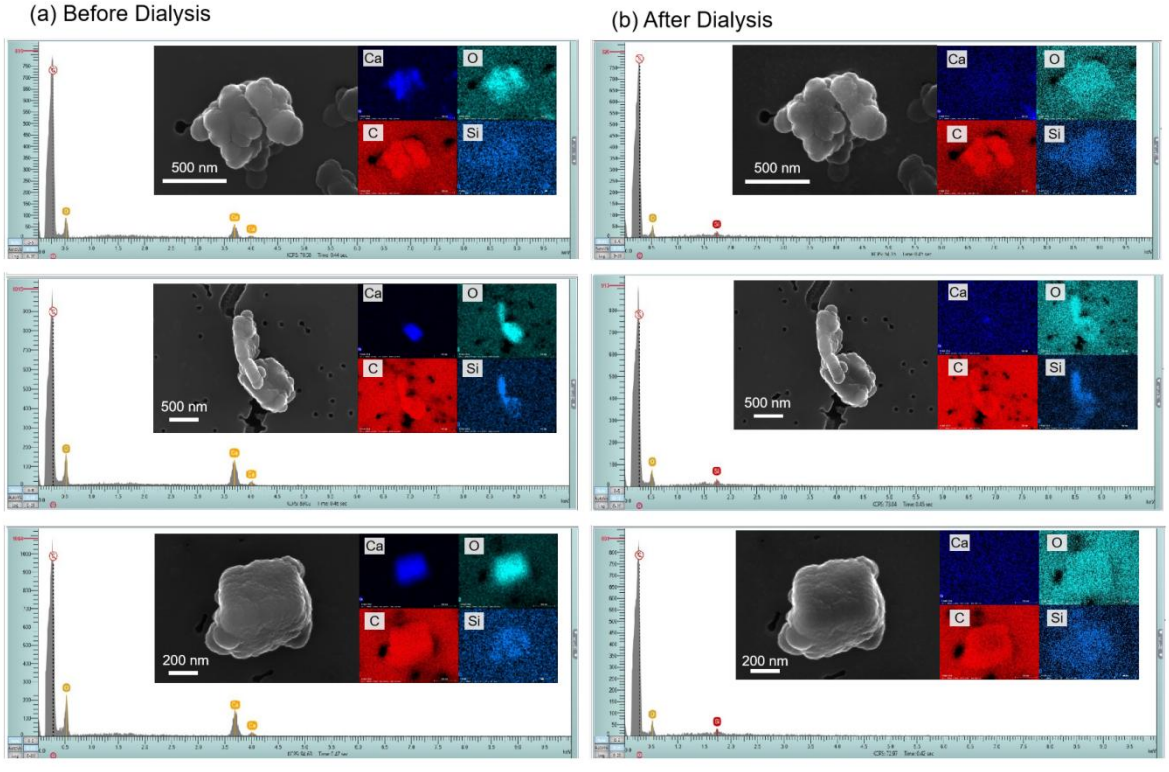

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Figure 6. Micrographs of typical Ca-O particles and their EDX elemental mappings / spectra illustrate the coating of calcite components on the surface of Si-containing particles.

3.3 Rapid buffering of soluble calcium components

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 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).

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 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).

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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).

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 consequently enhanced neutralization capacity for acidity across Earth's environmental compartments than previously recognized.

4 Summary and Implications

Mineral dust, emitted from wind-blown soils in deserts and semi-arid regions, ranks among the most abundant atmospheric aerosols by mass. It undergoes substantial chemical evolution and exerts a profound influence on the climate system. Our single-particle analysis enables a quantitative estimate of water-soluble calcium abundance (56.9–88.2 % by number) in mineral aerosols from 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.

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Data availability

All data supporting this study are available in the main 920 text and its supplementary materials. Additionally, the datasets (Physicochemical Properties of Water-Dialyzed Single Asian Dust Particles) have been permanently archived in the Mendeley Data repository (https://doi.org/ 10.17632/4js9dfy9hm.1).

Author contribution

JC and DZ designed the experiments and TH carried them out. TH, DZ, and YS developed the methodology and YS performed the individual particle analysis. TH, NJ, and YS prepared the manuscript with contributions from all co-authors.

Competing interests

The authors declare that they have no conflict of interest.

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