******Response to referee comments from Anonymous Referee #2, 10 Mar 2026******

We sincerely thank the reviewer for their thoughtful feedback. Our responses to the comments are provided below in italics. The updated figure mentioned in the response is also included at the end of this letter for easy reference.

Suggestions for revision

Many thanks to the authors for refining the publication, which seems good to me.

I still have one point that I might not have clarified well enough during the first review:

1. You coat the particles with carbon

2. You make an image, dissolve the soluble fraction, and then make an image again

3. You determine volume losses by comparing the images.

I'm highly concerned that this is biased for the following reason: if I look at the images in figure 6, I seem to be looking at the carbon coating - not on the actual particle (smooth, no surface structures). The actual particles still seems to be smaller (according to the mappings shown).

Now, if all the Ca is gone (transition from 3rd to 4th row), there must be an according significant void somewhere, and according to the mapping you show, it must be inside the visible 'particle'. But one can bare observe any change in the images (potentially a slight decrease in size, but not matching the loss).

To me it seems that the carbon coating stands as a separate empty shell, thus, biasing the later volume measurements.

For that reason I was trying to ask 'How thick was your carbon coating?', but I think I didn't formulate this clearly enough.

So, how thick is it?

Please show for comparison a particle, of which we know that is totally gone, e.g. a pure NaCl or similar.

Response:

The thickness of the carbon coating was estimated to be on the order of 10–15 nm,

based on standard operating procedures for carbon coating in electron microscopy. Some studies have reported coating thickness ranging from several nanometers (nm) to approximately 20 nm (Reed, 2005; Goldstein et al., 2017; Brodusch et al., 2018). To our knowledge, no evidence suggests that such coatings introduce significant bias in electron‐microscopic studies of atmospheric aerosol particles. For instance, Niimura et al (1994) did not find substantial bias despite using carbon-coated films for particle collection and additional metal coating for analysis. In the present study, both samples (before and after dialysis) were subjected to identical carbon coating conditions. The observed slight decrease in particle size, determined by automated microanalysis, exceeded the typical thickness of carbon coatings reported in the literature.

*Following the reviewer's suggestion, we performed a control experiment using ultrapure NaCl (guaranteed reagent, GR, Sinopharm Chemical Reagent Co., Ltd., China) as a reference material to test whether the carbon coating could form an empty shell that remains after dissolution of the soluble fraction. After dialysis, the NaCl particles completely disappeared (**Fig. S8**), demonstrating that under our experimental conditions no persistent carbon shell remained. The additional experiments and its result have been incorporated into the revised manuscript, as detailed in **Lines 302-305 on Page 12**, and supplementary **Fig. S8**.*

Regarding the apparent size difference between particles observed in secondary electron images and in EDX mappings, this arises from the different beam sizes used by the two detectors. Secondary electron imaging employs a smaller beam size to achieve high spatial resolution for morphological characterization, whereas EDX analysis uses a larger beam size to generate sufficient characteristic X‐ray signals. Furthermore, the central region of a particle is generally flatter than its edges, leading to relatively weaker X‐ray signals at the particle outline and consequently lower spatial resolution in the elemental mappings.

*More importantly, parts of the Ca‐containing particles were indeed removed during dialysis, as evidenced by the crystalline subgroups shown in **Fig. 5**. Our conclusions are based on multiple lines of evidence for the presence of water‐soluble Ca‐rich coatings on mineral dust particles: The disappearance of the Ca signal, the emergence of Si (and frequently Al) signals, and the largely unchanged overall morphology after dialysis. The NaCl control experiment further demonstrates that the carbon coating does not interfere with the dissolution of soluble parts in the particles during dialysis.*

Updated Figures

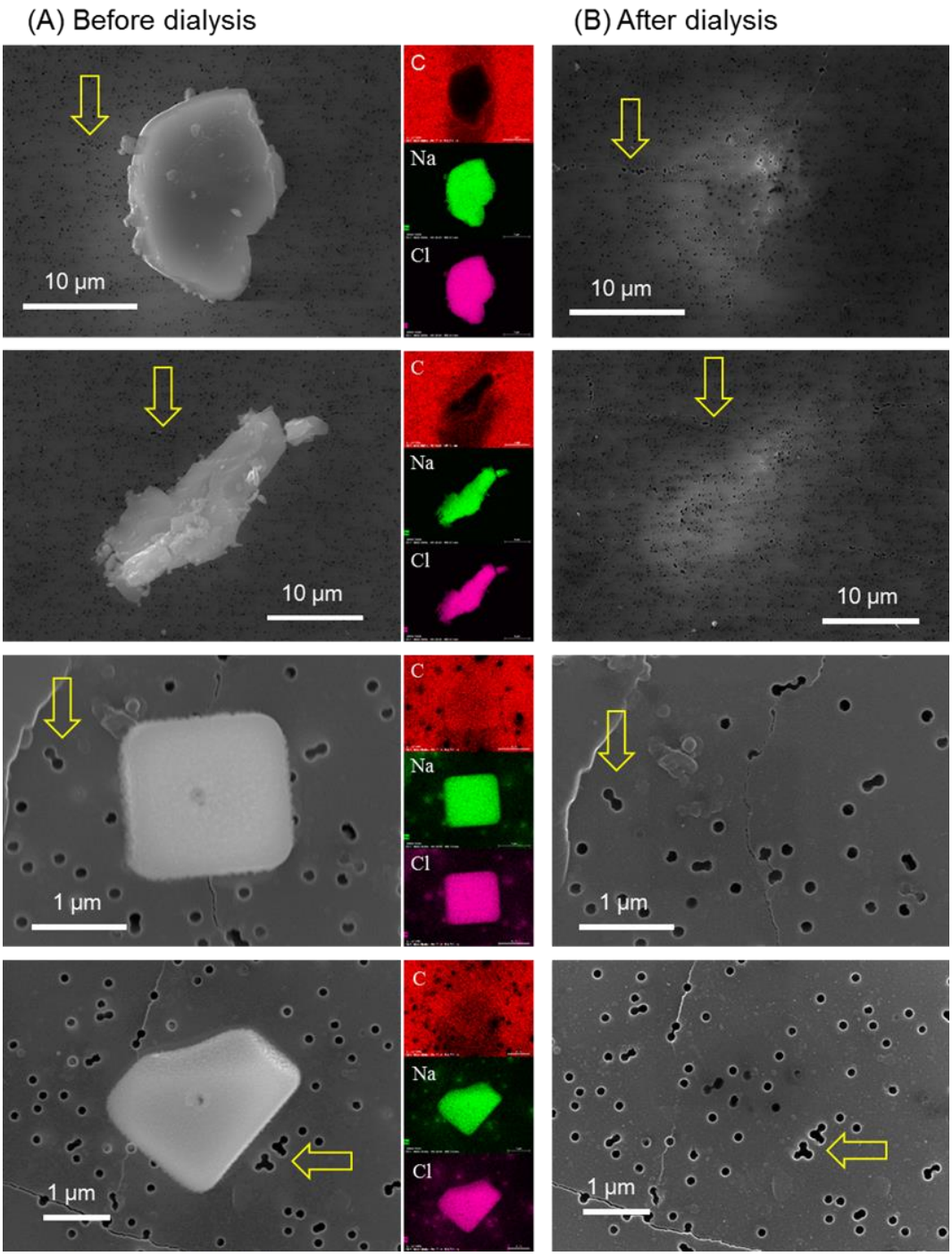

Figure S8. Micrographs and corresponding EDX elemental mappings of NaCl particles (A) before and (B) after dialysis, demonstrating the complete dissolution of those water-soluble particles. Yellow arrows indicate position markers that correspond to the same locations before and after dialysis, showing the disappearance of NaCl particles.

References

N. Niimura, K. Okada, Z. Fan, K. Kai, Y. Arao, G. Y. Shi, and T. Takahashi: A method for the identification of Asian dust-storm particles mixed internally with sea salt. Journal of the Meteorological Society of Japan, vol. 72, no. 5, pp. 777–784, https://doi.org/10.2151/JMSJ1965.72.5_777, 1994.

Brodusch, N., Demers, H., and Gauvin, R.: Field emission scanning electron microscopy: New perspectives for materials characterization, Springer, Singapore, 42-122 pp., https://doi.org/10.1007/978-981-10-4433-5, 2018.

Goldstein, J. I., Newbury, D. E., Michael, J. R., Ritchie, N. W., Scott, J. H. J., and Joy, D. C.: Scanning electron microscopy and X-ray microanalysis, Springer, New York, USA, 290 pp., https://doi.org/10.1007/978-1-4939-6676-9, 2017.

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