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Characterization of refractory aerosol particles collected in the tropical upper troposphere–lower stratosphere (UTLS) within the Asian tropopause aerosol layer (ATAL)

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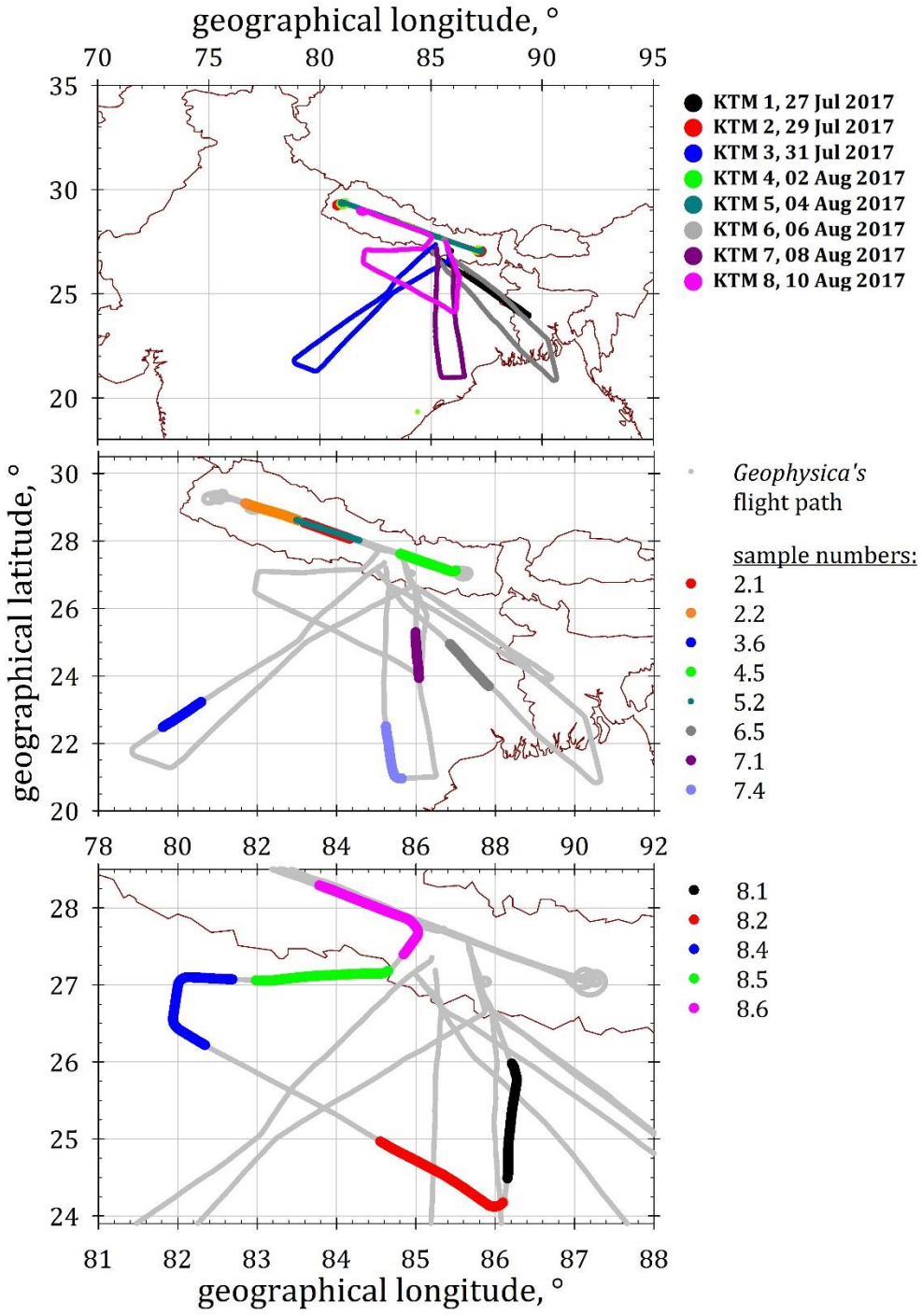

Figure S1: Flight patterns of the eight StratoClim2017 flights above the Indian subcontinent. While in the upper graph the complete flight patterns are given in color, in the two graphics below only the sections in which the particle collection took place are marked in color. The bottom graph shows the 5 particle collections during flight 8, the middle graph shows all other particle collections. The flight altitudes and positions in relation to the tropopause can be found in the manuscript in Table 1 and Figure 1.

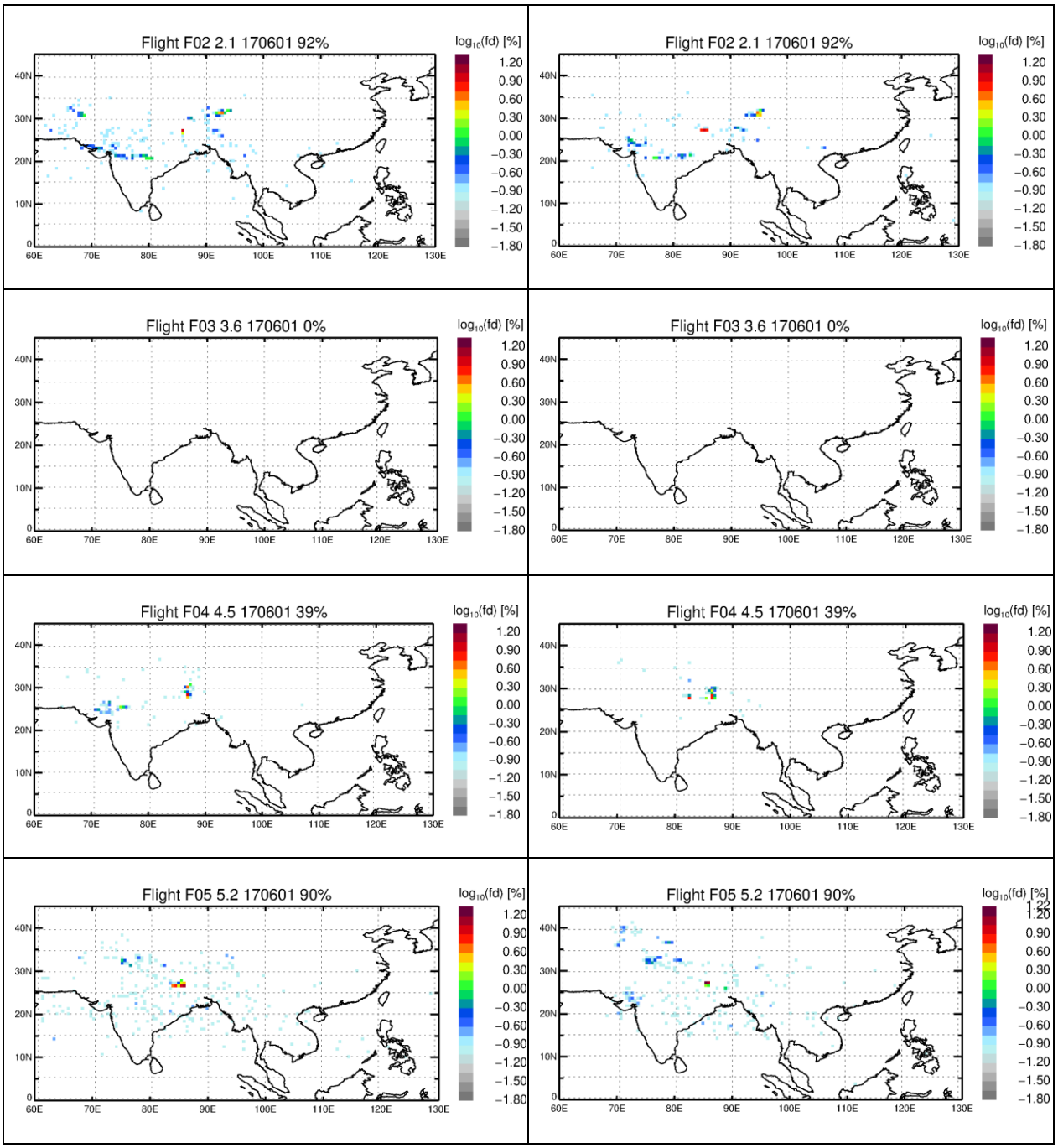

Figure S2a: left side) Frequency distribution (fd) of air mass origins at the model boundary layer for samples 2.1, 3.6, 4.5, and 5.2. Back-trajectories were calculated using ERA5 reanalysis back to the start time of the monsoon season (06/01/2017). Only back-trajectories are considered reaching the model boundary layer by then. Right side) Frequency distribution (fd) of the mean location of the strongest change of potential temperature along the back-trajectories (running mean over 6 hours) indicating the position of strongest uplift of air along the trajectory.

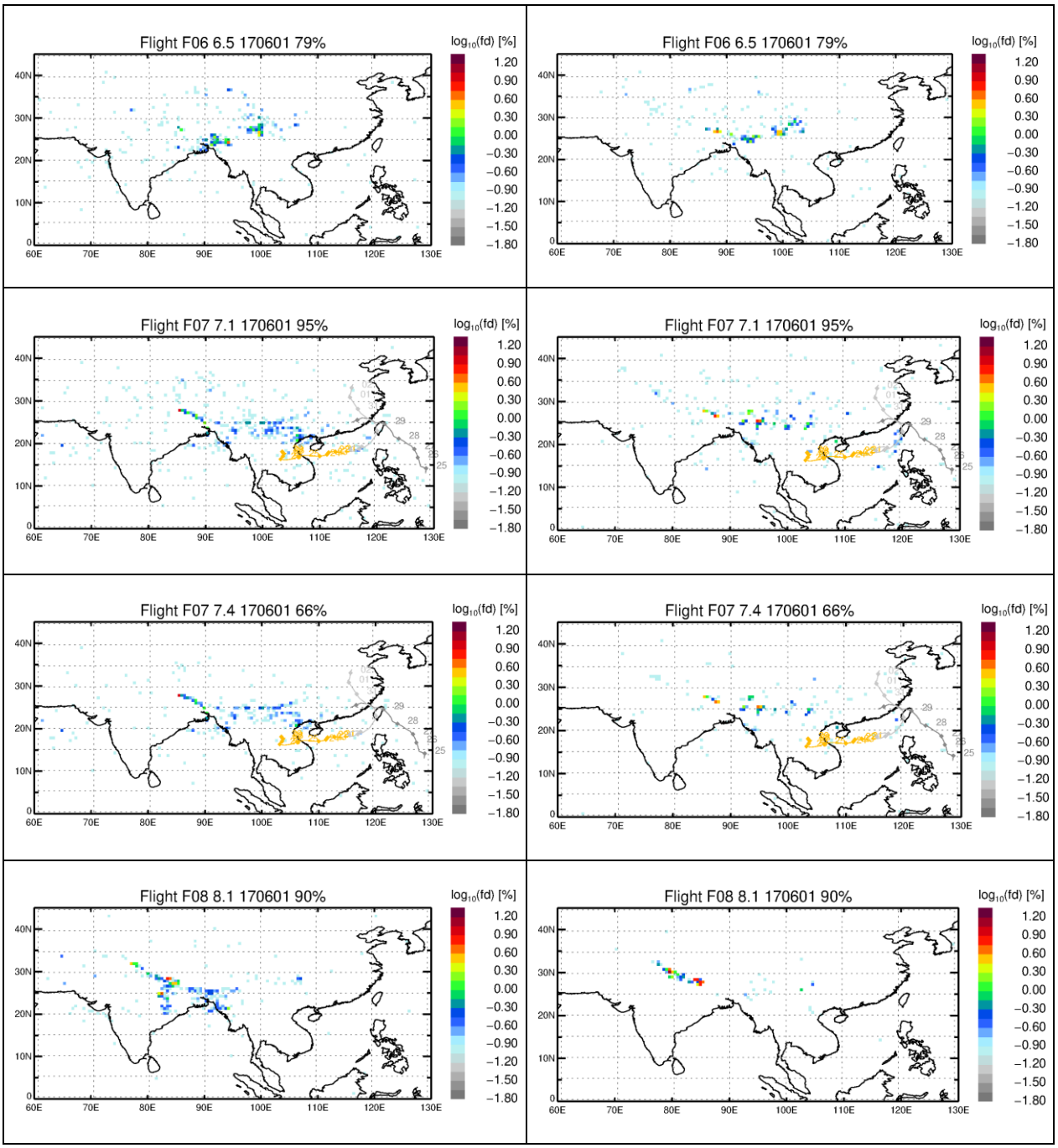

Figure S2b: left side) Frequency distribution (fd) of air mass origins at the model boundary layer for 6.5, 7.1, 7.4, and 8.1. Back-trajectories were calculated using ERA5 reanalysis back to the start time of the monsoon season (06/01/2017). Only back-trajectories are considered reaching the model boundary layer by then. Right side) Frequency distribution (fd) of the mean location of the strongest change of potential temperature along the back-trajectories (running mean over 6 hours) indicating the position of strongest uplift of air along the trajectory.

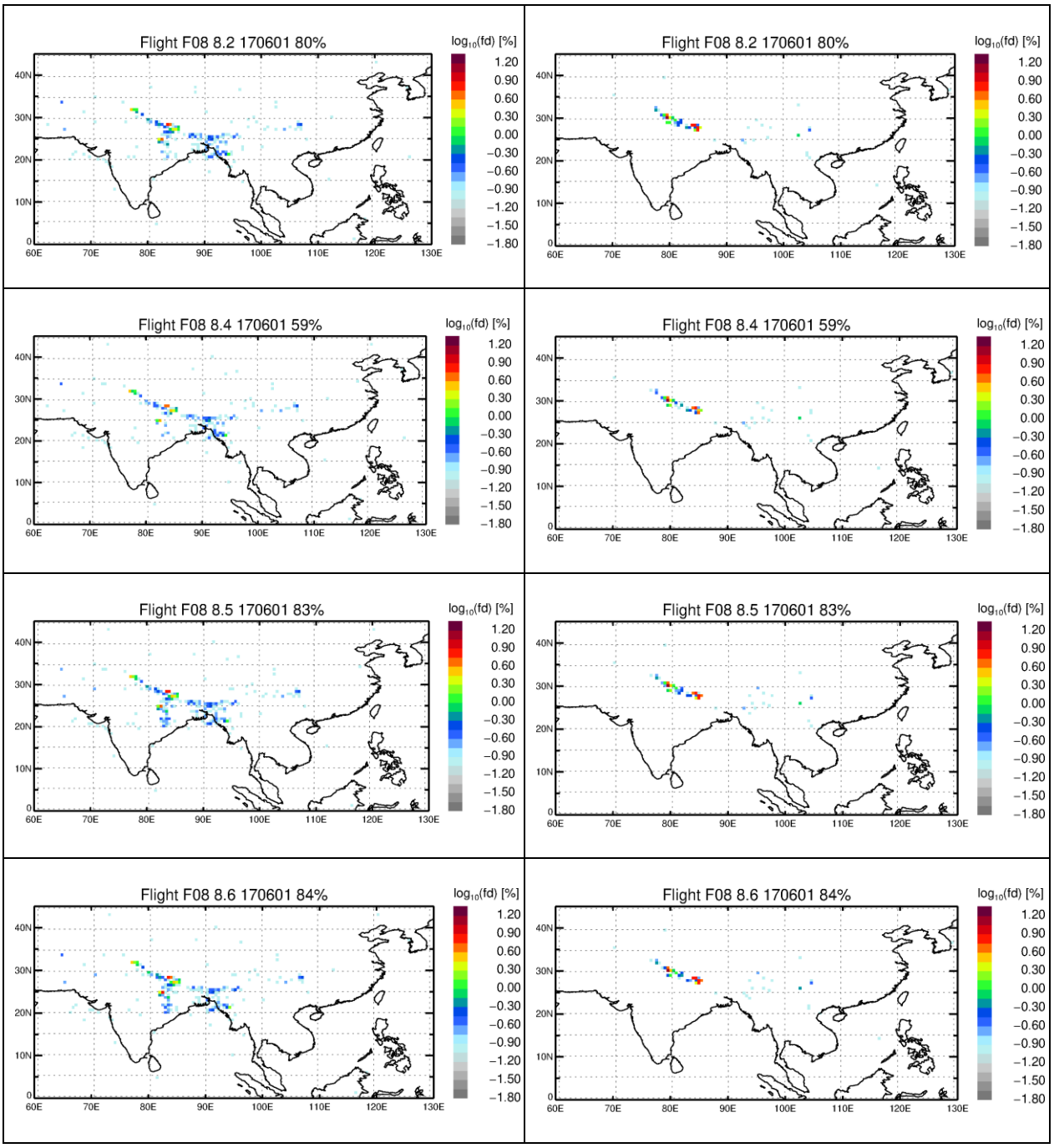

Figure S2c: left side) Frequency distribution (fd) of air mass origins at the model boundary layer for samples 8.2, 8.4, 8.5, and 8.6. Back-trajectories were calculated using ERA5 reanalysis back to the start time of the monsoon season (06/01/2017). Only back-trajectories are considered reaching the model boundary layer by then. Right side) Frequency distribution (fd) of the mean location of the strongest change of potential temperature along the back-trajectories (running mean over 6 hours) indicating the position of strongest uplift of air along the trajectory.

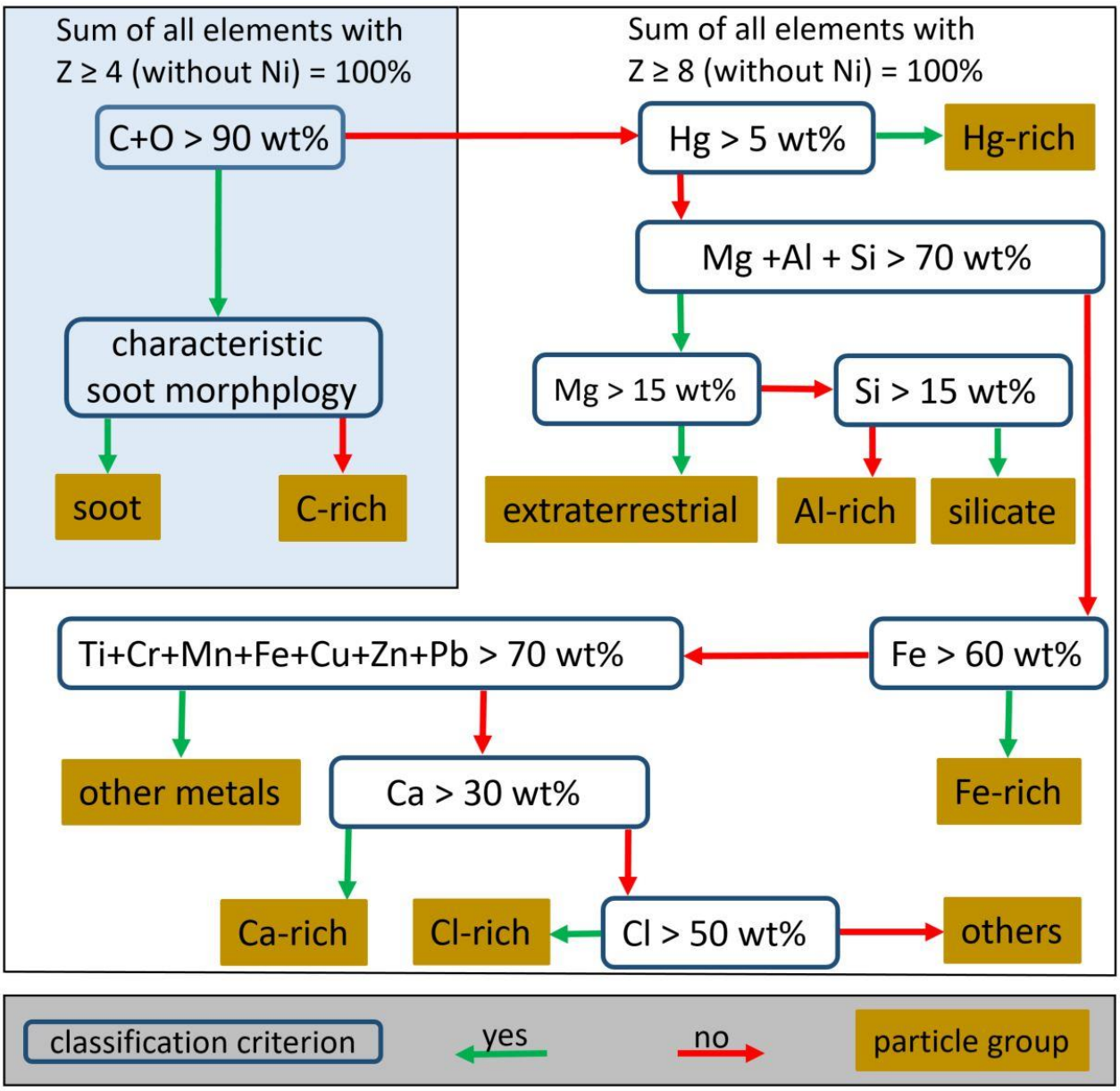

Figure S3: Classification criteria (blue) for the classification of the eleven particle groups (gold) based on the normalized elemental wt% of the EDX analysis. Green arrows indicate fulfilment of the criterion, red arrows indicate non-fulfilment.

For the classification of C-dominated particles the sum of all elements with Z ≥ 4 was used (without Ni, which is contained in the used TEM grid substrates). The morphology of all individual particles which fulfilled the C+O > 90 wt% criterion was additionally checked for the characteristic soot morphology (agglomerates of small spherical primary particles), which enabled the classification of the soot group. Since the morphological detection of the soot for very small particles (< 200 nm) does not always work reliably due to the limited resolution of the SEM, the determined soot content only represents a minimum percentage of this group.

For the classification of the non-C dominated particles the normalized sum of all elements with Z ≥ 8 was used (without Ni). This was used because when analyzing very small particles in the SEM, the TEM grid is also excited, making the elements C and Ni (contained in the TEM grid) visible in the particle spectrum, which would falsify the results of the particle analysis.

As the wt%-values shown in this classification refer to the normalized sum of elements with Z ≥ 8 (so not considering C, N, and O content) these values are not identical with the real relative elemental contents of the particles but are used for the best possible classification of the particle groups.

Hg-rich particles were always found in the form of very small nanoparticles that adhere to larger particles. When excited in the SEM, the larger particle is often dominating the spectrum, making the Hg only faintly visible in the spectrum. Therefore, the classification criterion of the Hg-rich group is fulfilled when > 5 wt% Hg in a particle analysis is received.

Figure S4: Typical SEM and TEM images, energy dispersive X-ray spectra and normalized wt% of main elements (100% = all elements (without Ni) with Z ≥ 4 for carbonaceous particles and Z ≥ 8 for all other particle groups (see Figure 3) of the ten classified refractory particle groups.

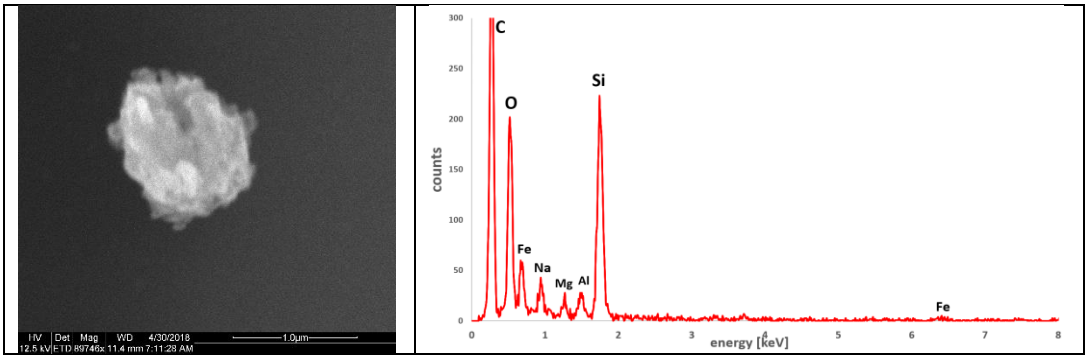

Figure S4a: Secondary electron image and EDX spectra of a silicate particle

(normalized wt%: Si= 70.5 wt%, Al= 6.5 wt%, Fe= 6.1 wt%, Mg= 4.6 wt%, Ca= 3.3 wt%, Na= 2.9 wt%)

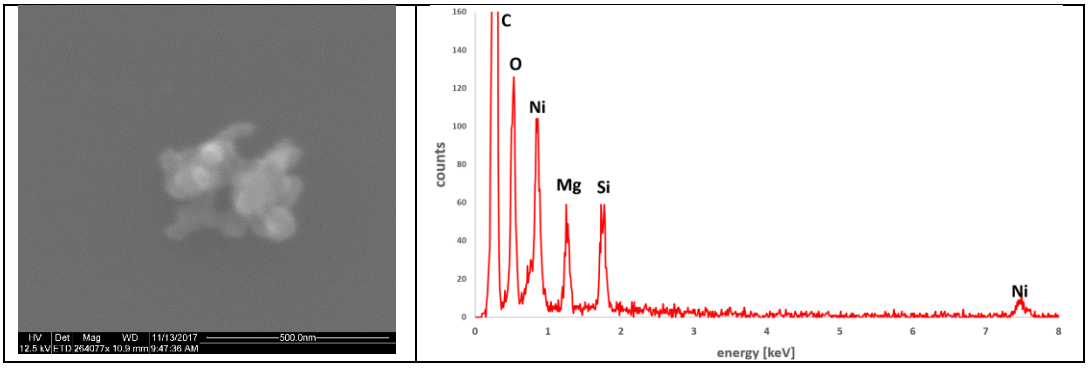

Figure S4b: Secondary electron image and EDX spectra of an extraterrestrial particle.

(normalized wt%: Si = 60.1 wt%, Mg = 35.6 wt%, Al = 1.0 wt%)

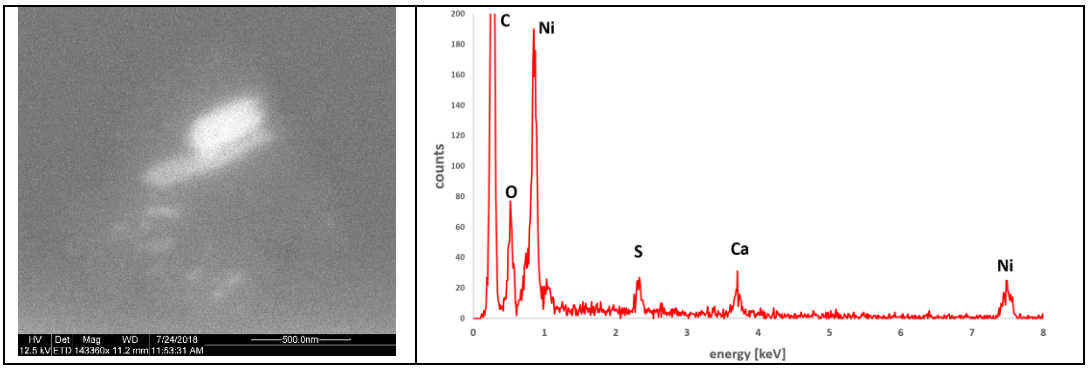

Figure S4c: Secondary electron image and EDX spectra of an Ca-rich particle.

(normalized wt%: Ca = 40.1 wt%, S = 27 wt%, Cl = 6.1 wt%, Na = 5.8 wt%, Mg = 5.6 wt%, Fe = 2.8 wt%)

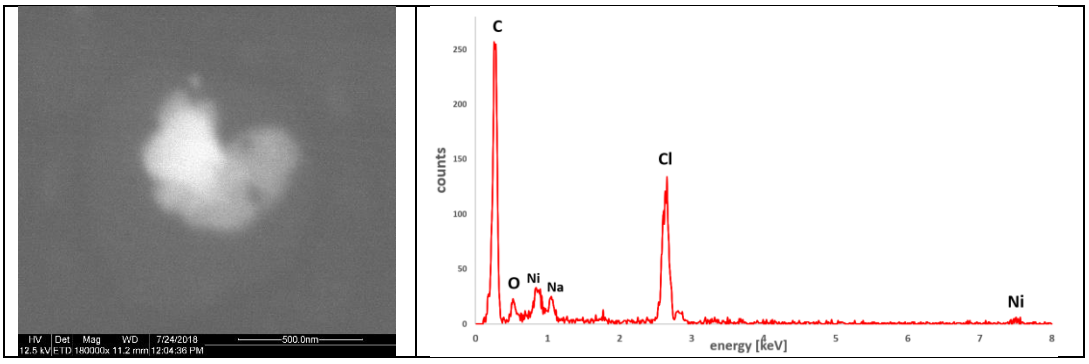

Figure S4d: Secondary electron image and EDX spectra of an Cl-rich particle.

(normalized wt%: Cl = 75.7 wt%, Na = 9.9 wt%, K = 1.9 wt%, Si = 1.7 wt%, Ca = 1.4 wt%)

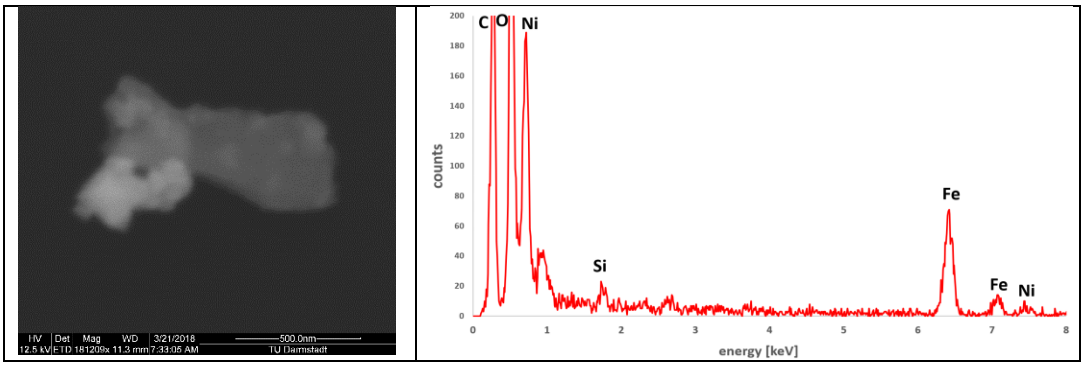

Figure S4e: Secondary electron image and EDX spectra of an Fe-rich particle.

(normalized wt%: Fe = 85.5 wt%, Na = 3.8 wt%, Si = 2.3 wt%, Cl = 1.5 wt%)

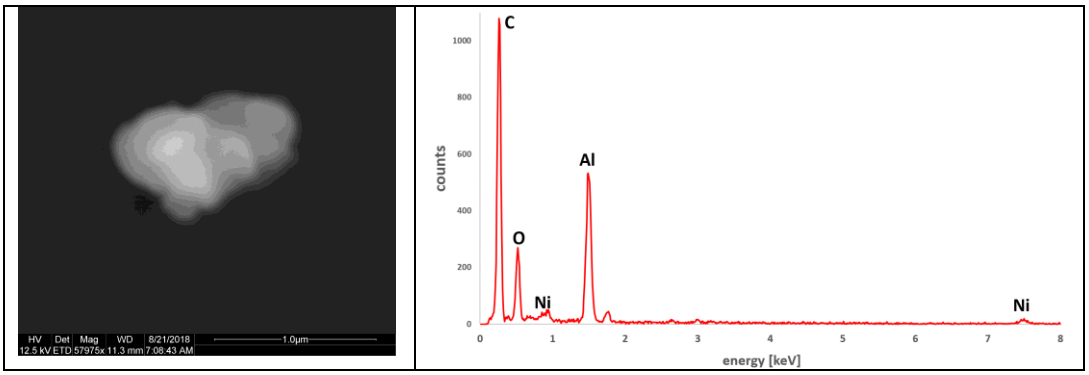

Figure S4f: Secondary electron image and EDX spectra of an Al-rich particle.

(normalized wt%: Al = 82.2 wt%, Si = 2.7 wt%, Cl = 1.9 wt%, S = 1.8 wt%)

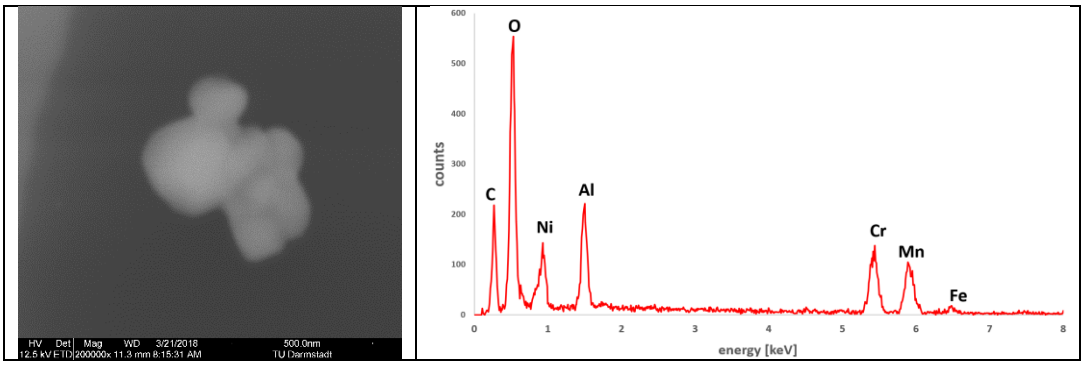

Figure S4g: Secondary electron image and EDX spectra of a particle from the “other metals” group.

(normalized wt%: Cr = 40.9 wt%, Mn = 39.7 wt%, Al = 12.9 wt%, Fe = 2.5 wt%)

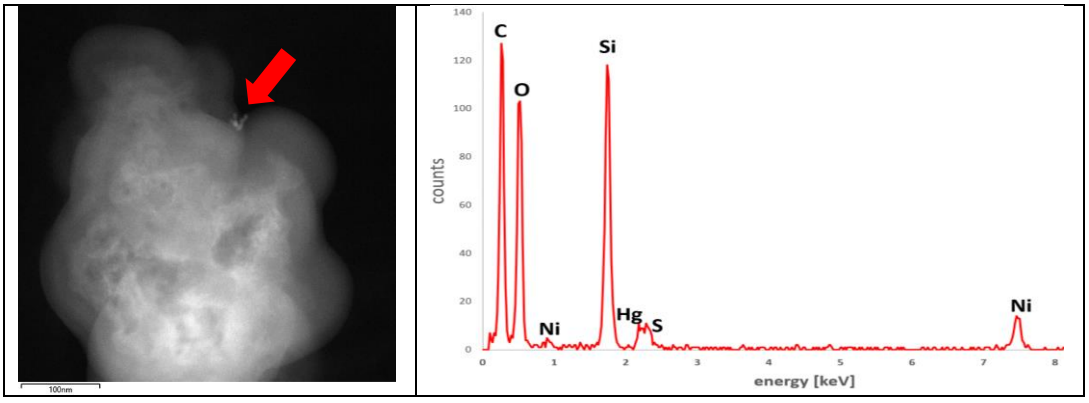

Figure S3h: TEM brightfield image and EDX spectra of a small (25 nm) Hg-rich particle (marked by red arrow) agglomerated on a larger (500 nm) silicate particle.

(normalized wt%: Si = 83.1 wt%, Hg = 9.3 wt%, S = 1.6 wt%)

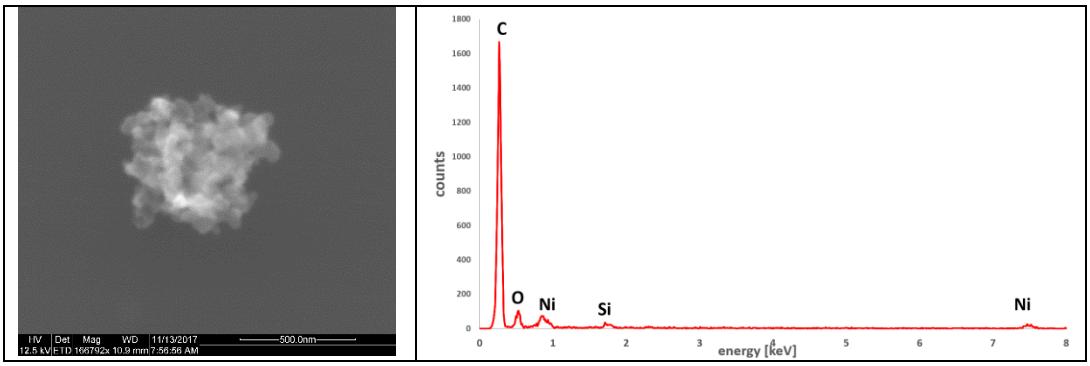

Figure S4i: Secondary electron image and EDX spectra of a soot particle.

(normalized wt%: C= 85.8 wt%, O = 10.5 wt%, Si = 1.7 wt%, S = 0.4 wt%)

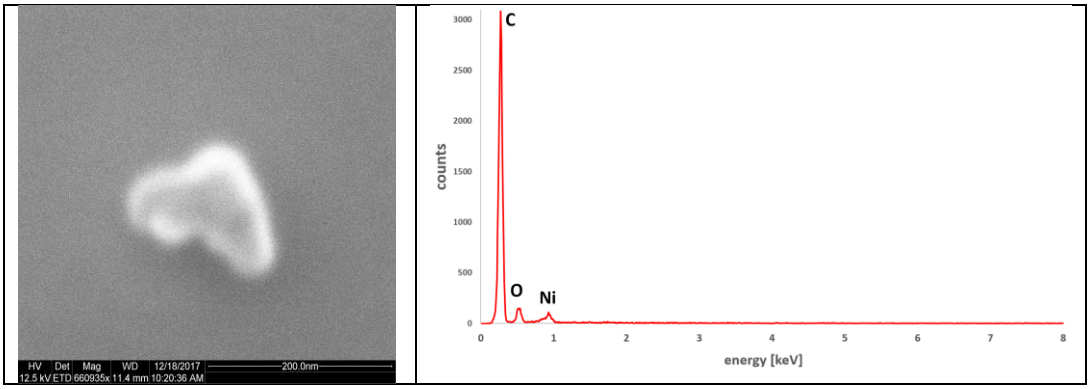

Figure S3j: Secondary electron image and EDX spectra of a C-rich (organic) particle.

(normalized wt%: C= 86.7 wt%, O = 10.2 wt%, Si = 1.1 wt%)