**Supplementary material guide**

The supplementary material of this paper is divided in 12 files, which are described hereafter.

File S1 – Ground-based images and cloud altitude parameterization.

File S2 – UAS flight reports. Lines highlighted in orange represent the time of ash sampling (for UAV - 10-11-2023 - Sakurajima, UAV - 11-11-2023 - Sakurajima, and UAV - 23-07-2024 – Etna datasets) or multigas analysis (for UAS - 19-11-2023 - Sakurajima dataset).

File S3 – Optical particle counter (OPC) raw data. Lines highlighted in orange represent the time of ash sampling.

File S4 – Airborne ash sample images and associated binarization.

File S5 – Particle counts from image analysis and associated grain size distributions (GSDs).

File S6 – Image and associated data from the airborne ash collector and counter with camera (A2C3) device.

File S7 – Multigas raw data. Lines highlighted in orange represent the data shown in the paper.

The four .kmz files are for the UAV flight visualization in a GIS software.

The .avi file is the image sequence acquired by the A2C3 camera during the ash sampling at Etna on the 23 July 2024.