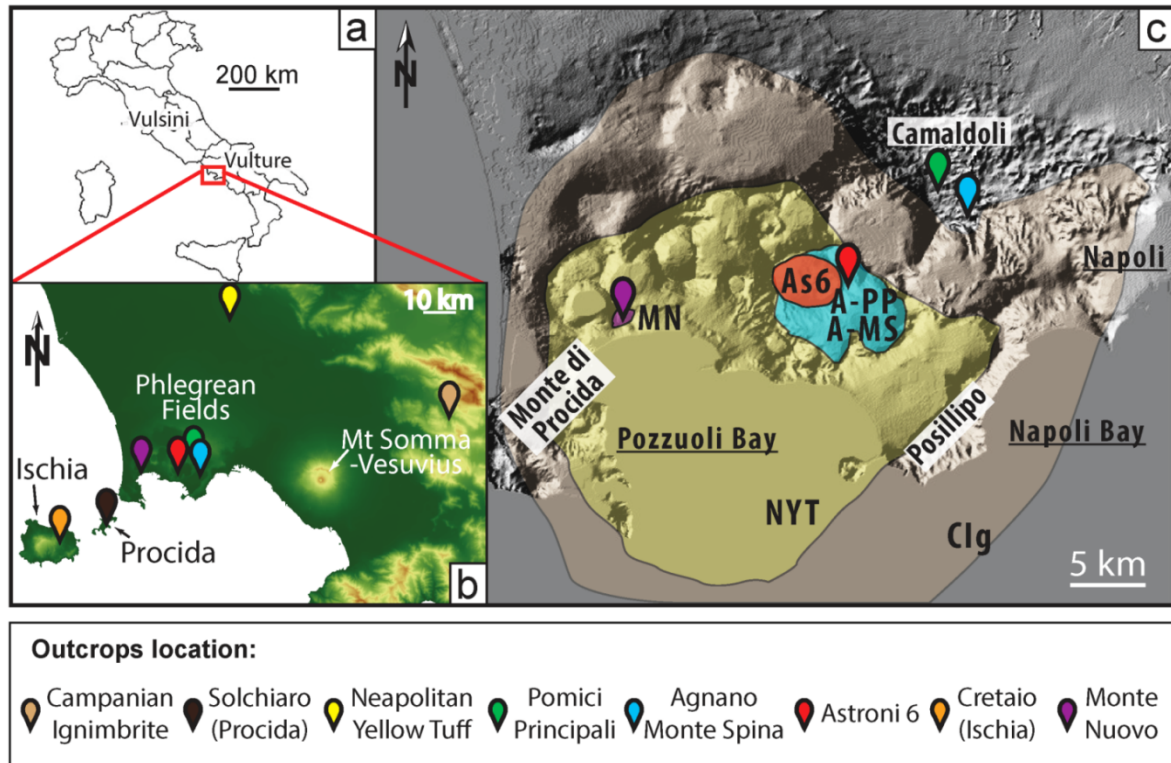


Supplementary Material

SM 1 – Localisation of the outcrops



SM1: (a) General map of Italy; (b) Location of Phlegrean Fields, Ischia and Procida respect to Monte Somma - Vesuvius. (c) Point location of the different outcrops. The CI fallout sequence was sampled at Acquafidia (Fig. 1; $40^{\circ} 55.732'N$, $14^{\circ} 42.043'E$), ~50 km East of Naples, and divided into seven eruptive layers (V1 through V7) following Signorelli et al. (1999). Samples of three fallout layers (LM3, LM5, and LM9; Orsi et al. 1995) of the NYT Lower Member sequence were collected near the city of Caserta, ~26 km North of Naples (Fig. 1; $41^{\circ} 6.031'N$, $14^{\circ} 16.421'E$). The products of the other Phlegrean eruptions were sampled within the CFc. Twelve fallout layers of the PP pyroclastic sequence (PP-1 through PP-12) (Fig. 1; $40^{\circ} 52.334'N$, $14^{\circ} 11.587'E$), and eleven A-MS fallout layers (A-MS-1 through A-MS-11) (Fig. 1; $40^{\circ} 51.532'N$, $14^{\circ} 12.243'E$) were sampled in the Camaldoli area. Four fallout layers of the As6 eruption sequence (As6-1 through As6-4) were collected in the Astroni crater (Fig. 1; $40^{\circ} 50.688'N$, $14^{\circ} 9.885'E$). Finally, the MN deposits were sampled (from MN1 to MN4) in the city of Pozzuoli, along a flank of the Monte Nuovo cone (Fig. 1;

40° 49.979'N, 14° 5.426'E). The latter deposits were sampled in a less systematic way than the other investigated pyroclastic sequences because the MN eruption did not have any purely sustained column phase. The CT sequence was sampled (CT-B through CT-F2; Orsi et al. 1992b) at Cretaio on the island of Ischia (Fig. 1; 40° 44.073'N, 13° 55.382'E). Scoriae from the pyroclastic sequence of the Solchiaro eruption were collected along a sea cliff near Marina di Chiaiolella at Procida (Fig. 1; 40° 44.862'N, 14° 0.128'E).

SM 2 – Material and Methods

Hereafter is described the sampled material and the methods used to perform analyses.

SM2.1. Sampling and sample preparation.

Plinian fallout deposits of four variable magnitude explosive eruptions (PP, A-MS, As6 at CF and CT at Ischia), and some pumice clasts emitted during the MN eruption at CF were sampled in natural sections. In the case of the voluminous ignimbritic eruption, three eruptive units corresponding to the first erupted magma of CI and three fallout layers of the NYT Lower Member sequence were investigated. The eruptive products of the Procida Solchiaro event were also sampled in detail. The sampled sections (Fig. 1) were chosen among those located along the dispersal axis of the fallout deposits. PDC deposits overlying Plinian fallout beds of CI fallout and corresponding to the latest emitted magma have been studied for comparison by taking into account the magma volume they represent.

At least 100 pumice clasts were collected from each eruptive layer for each studied eruption, in the size fraction 32-16 mm (-5φ to -4 φ). All clasts were then washed and sawed into three parts on each of which were performed density, glass composition, and textural analyses. To reach the main objective of the research, the analytical work was focussed on the characterization of the residual glass of the Plinian fallout activity.

SM2.2. Density measurements – sample selection criteria

Density measurements were based on the Archimedes' principle with three successive weightings. Each fragment was weighed in air (M_a) and then impregnated with molten paraffin to fill all open and connected vesicles without inducing any modification to its volume. The impregnated fragment was then weighed in air (M_a^i) and in water (M_w^i). Density (d) was calculated by the relation: $d = M_a / (M_a^i - M_w^i)$.

The distribution of the obtained values is generally Gaussian with at least 75% of the pumice clasts enclosed within the mode. Such a distribution permits one to select the most representative pumice clasts (at least 6) for the subsequent chemical and textural investigations. This protocol of selection was used for CT, CI, PP, A-MS and AS-6 samples. For the MN eruption, density values were only measured on few clasts collected from the MN pyroclastic sequence. No density measurements were performed on the NYT collected pumice clasts because of their small number and size. Scoria clasts randomly collected from the Solchiaro pyroclastic sequence were used only for glass composition analyses.

SM2.3. Glass composition analyses

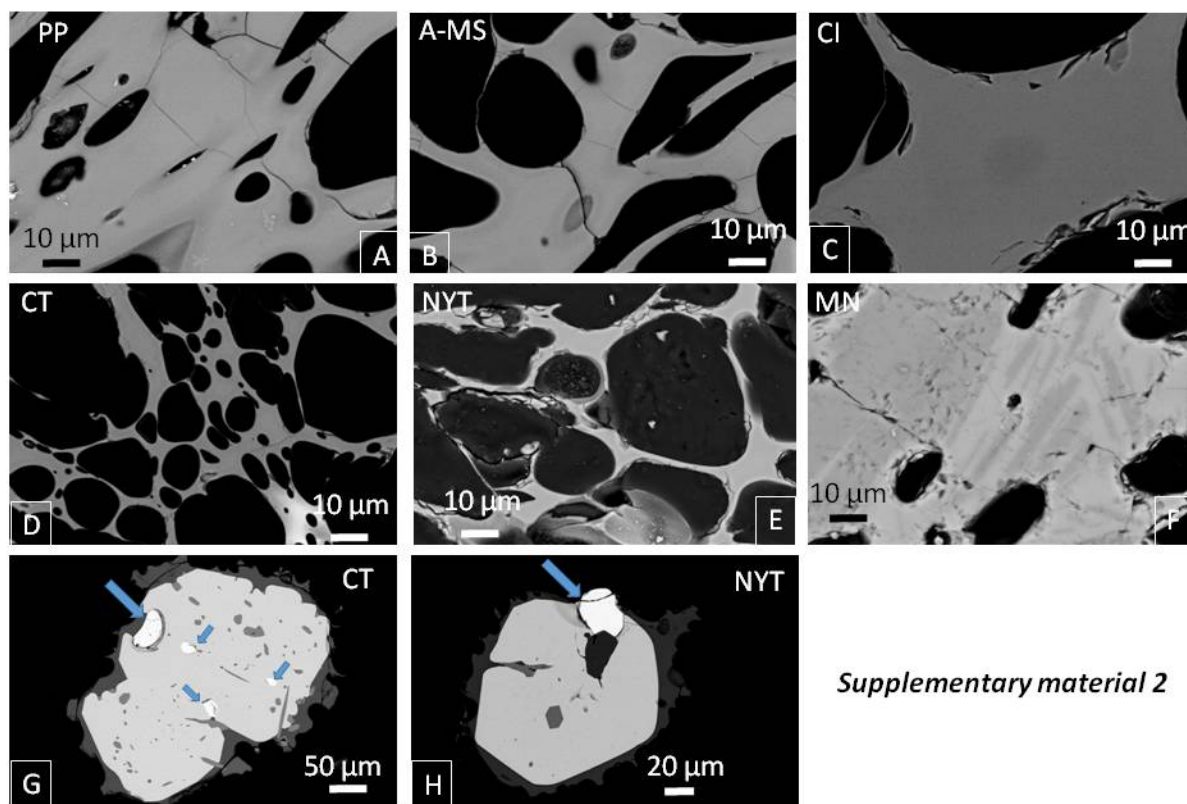
In order to perform glass composition analyses one part of each selected pumice clast was embedded in resin, abraded and polished. The analyses were carried out by electron microprobe (CAMECA-SX 100, CAMPARIS, France) measurements with an acceleration voltage of 15 kV, a beam current of 4 nA for major elements and a dwell time of 5s for Si and Na to limit Na loss and 10s for the other elements (Ti, Al, Fe, Mg, Mn, Ca, K, P). For halogen elements (F and Cl) and S, analyses are performed with an acceleration voltage of 15 kV, a beam current of 45 nA and a dwell time of 130s for the F, to 180s for the Cl and S depending on the minimum value of halogen concentration measured. Three natural glass samples (obsidians from Lipari, Aeolian Islands, Italy; Little Glass Mountain, California, United States; and Corbetti volcano, Ethiopia), used as internal standards, were analysed

for inter-calibration of each EPMA session (Balcone-Boissard et al., 2008). Detection limit for S is 80 ppm, close to analyses contrary to F and Cl (200 ppm).

SM2.4. Textural investigations.

Back-Scattered Electron (BSE) images were acquired on the selected samples in order to qualitatively describe the residual glass (microcrystallinity, alteration, etc.). Images of the characteristic textures of pumice and scoria clasts are provided in supplementary material (SM3).

SM 3 – Textural characteristics



SM3: Textural characterisation through SEM images of representative matrix pumice clasts of the different studied eruptions. *Glassy matrix glass:* A- Pomice Principale; B- Agnano Monte Spina; C- Campanian Ignimbrite; D- Cretaio Tephra; E- Neapolitan Yellow Tuff; *Microcrystallized matrix glass:* F- Monte Nuovo. Images of the sulfur globules (blue arrow) identified in magnetites of the Cretaio tephra (G) and Neapolitan Yellow Tuff (H) eruption.

SM 4: Glass composition (a) and halogen content (b) of the recognized pyroclastic flow (PF) units. PF-U1 was sampled within the most evolved unit (Sant'Anna), PF-U2 within the intermediate unit (Massa) and PF-U3 within the less evolved unit (Ponti Rossi).

