Supplementary Materials for

First field evidence of the electrical multipolar nature of volcanic aggregates

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Supplementary Materials

S1. Identification of the aggregate type using the terminal velocity measurements

The methodology proposed in this paper was not conceived for a direct quantification of the aggregate type. In fact, no adhesive tape was placed at the base of the two copper plates for further analysis at the Scanning Electron Microscope (SEM) as the presence of the external electric field may have altered and contaminated the initial population of particles collected on the tape at the moment of the impact. Some clue can be inferred by those aggregates that directly impacted on the vertical copper plates due to the breaking action of the electric field, but these represent a small portion of the entire dataset. Nevertheless, a reliable and robust quantification of the aggregate type can be obtained overlapping the measurements of the terminal velocity versus aggregate size with previous studies, specifically designed to investigate the morphology of ash aggregates (33, 37). In their study the authors applied the HSC and SEM tapes to study the inner structure of ash aggregates falling at Sakurajima Volcano (Japan) over several years in similar eruptive scenarios to those analyzed in the present work. The overlapping of the two datasets is reported in Fig. S1.

![Figure S1. Aggregate type classification. Terminal velocity as a function of the aggregate diameter. Overlap of the data reported in the present work with the results published by Diaz Vecino et al. and Gabellini et al. (33, 37).](image-url)
Fig. S1 shows a good agreement of our data with those recorded by and Diaz Vecino et al. and Gabellini et al. In particular, most of the observed aggregates coincide with the trends of PC1 and PC3 types. The lack of PC2 types at our sampling location can be due both to physical reasons behind aggregation, such as more favorable conditions for PC1 and PC3 formation, or simply to the sedimentation that occurred earlier in time and thus closer to the vent respect to our location for PC2. Anyway, the lack of some types of aggregates during some explosions has been already reported by Gabellini et al. (see Fig. 5A in (35)).
S2. Aggregate type, density and sign of the charge

Aggregate type as a function of the aggregate density, charge polarity and aggregate diameter (Fig. S2).

**Figure S2. Aggregate density and diameter.** Aggregate type as a function of aggregate density and aggregate size.
S3. Error bars associated with the evaluation of the bulk electric charge, surface charge density and charge-to-mass ratio

In figure Fig.S3 we report the values of the bulk electric charge, charge surface density and charge to mass ratios carried by single aggregates and aggregate fragments with their uncertainties.

Figure S3. Uncertainties on the charge quantifications. (a) Bulk charges carried by aggregates and aggregate fragments; (b) Surface charge density; (c) Charge to mass ratios.
S4. Design of the field apparatus

The system developed for the measurement of the electric charge on the aggregates during a tephra fallout is made of four logical units (Fig. S4): i) the Wimshurst Electrostatic Machine (WEM) made by Sparkit and modified by us to operate in a volcanic eruption; ii) copper plates inside the wind shield; iii) High Speed Camera (HSC) controlled by a portable computer (Panasonic Toughbook); iv) a portable meteorological station.

Figure S4. Field setup. Description of the field setup used to measure the electric charges present on the surface of falling volcanic ash at Sakurajima Volcano (Japan).

In the following some details of each logical unit is provided:

i) The WEM is a modified version of the Sparkit Wimshurst machine (http://www.sparkitelectrostatics.com/). From the original architecture we modified the control of the motors, the power supply and the final capacitors. The two DC motors are now controlled by an Arduino Mega 2560 v3 that also provides the required power supply. The Arduino IDE interface allows programming the ATmega2560 chip mounted on board by means of the Processing language. Four digital pins are in charge of controlling the DC motors; three pins (one digital, two analogue) control the joystick. The joystick is added in order to rapidly select and run different programs according to the needs of the campaign: a testing routine, a routine where the WEM runs for a fixed amount of time (set by the user) and a continuous running program. In order to test the setup once placed in the desired
location, we run the testing routine. Once the ash fallout started, we selected the continuous running program. Running the WEM creates charges of different polarity that accumulates on the copper plates.

ii) The condenser is made of two copper plates of 13.3 cm x 4 cm per latus. The thickness is less than 0.2 mm. The distance between the plates is 1.8 cm. The condenser is mounted on an insulated platform and a couple of wires connect the plated to the WEM. The plates are encapsulated in a wind shield box 18 cm high in order to prevent the contamination from external air currents. The box is made of two transparent glasses on the plane of the camera and two wooden blocks on the other sides.

iii) The High Speed Camera used in this work is a Phantom M110 camera with a 60 mm Macro f2.8, controlled by a Panasonic Toughbook laptop. The camera was mounted on a Manfrotto tripod an protected from the ash fallout by means of a dedicated box.

iv) Meteorological data were acquired throughout the ash fallout using a Kestrel 5500 meteorological station. The continuous acquisition of data was particularly important to exclude the contamination of wind on the horizontal motion of falling ash during the measurements.
S5. Electric field outside the copper plates measured using a field mill and estimation of the potential difference between the capacitor

The electric field outside the copper plates has been measured by means of a Statometer II HAUG that was located perpendicular to center of the plates and then moved along the axis. Each plate has been measured independently just after the volcanic fallout. The uncertainties associated with each data point are evaluated as explained in section S7. Fitting the data for the positive plate with Eq.5 we find: $\Delta V_+ = 10.6 \pm 0.7 \text{ kV}$, where this is the uncertainty related only with the fitting procedure. Considering also the systematic component of the error due to the analytical approximation of the real field ($\approx 20\%$ of relative error at maximum for the setup under analysis, according to Fig.7 of Parker (2002)), we finally get $\Delta V_+ = 10.6 \pm 2.2 \text{ kV}$.

![Figure S5. Measurement of the external electric field of the HVCP (positive plate). Electric field measured outside the capacitor on the positive plate (data points) and best fit according to the analytical expression of Parker (47).](image)

Repeating the same procedure for the negative plate we find from the fitting procedure $\Delta V_- = 7.8 \pm 0.6 \text{ kV}$. Adding also the systematic component of the analytical approximation of Eq.5 we get $\Delta V_- = 7.8 \pm 1.7 \text{ kV}$.
Figure S6. Measurement of the external electric field of the HVCP (negative plate). Electric field measured outside the capacitor on the negative plate (data points) and best fit according to the analytical expression of Parker (47).

Following Taylor (47) the best estimator for $\Delta V$ can be found as the weighted average of $\Delta V_+$ and $\Delta V_-$ and their uncertainties: $\Delta V = 8.9 \pm 1.3$ kV.
S6. Determination of the electric field inside the copper plates inverting the motion of a conductive sphere

The electric field $\vec{E}_{\text{int}}$ inside the capacitor can be derived by the motion of a conductive sphere located within the plates by means of a wire. The system forms a vertical electric pendulum (see Fig. S4). We used a metal sphere with mass $m_s = 0.7$ g and radius $R_s = 0.5$ cm attached to cotton wire of negligible mass. Two distinct acquisitions were done by means of a Phantom M110 HSC with a framerate of 1500 fps (equivalent to a $\Delta t = 667\mu$s): the first one just before the volcanic eruption (hereafter named dataset A), the second one during the ash fallout (dataset B). The maximum angles of the center of mass of the sphere respect to the vertical line in the oscillatory motion of the pendulum are respectively 5.4° and 8.1° for dataset A and dataset B. However, we discarded the frames in which the sphere is close to the plates because in these regions the actual force exerted on the conductor is less than $\vec{F}_e = Q_0 \vec{E}_{\text{int}}$ due to the presence of image charges ($\vec{F}_e \approx 0.832 Q_0 \vec{E}_{\text{int}}$ according to Khayari and Perez (29)). Focusing on the central part of the trajectory, and considering the small angles associated with the entire oscillation span, allows us to approximate the problem as a one-dimensional motion along the horizontal direction (hereafter labelled as $x$). We define the positive orientation of $x$ from the positive plate to the negative one, i.e. $\vec{E}_{\text{int}} = E_{\text{int}} \hat{x}$.

![Figure S7. The electrical pendulum. The electrical pendulum of dataset A is made of a conductive sphere within the copper plates that can be charged and thus repelled by the electric plates.](image)

The motion of the sphere in a $x$-time diagram is well fitted by a parabolic equation such as $x_{\text{fit}} = c_2 t^2 + c_1 t + c_0$ ($R^2_{\text{fit}} = 0.9999$), which implies that the electric force $\vec{F}_e$ is much
greater than the drag force $F_D$ in the situation under analysis. A parabolic trajectory is in fact the consequence of a constant force acting on the body, as expected by the motion of a charge $Q_0$ in a uniform electric field (Fig. S8). Nevertheless, we included the drag force in the equation of motion used for the inversion process. The dataset A is composed of 21 single trajectories and for each of them the electric field has been derived independently. The same procedure has been applied to dataset B, made of 50 single oscillations.

Figure S8. Trajectory of the electrical pendulum ad a function of time. Two distinct trajectories of the center of mass of the sphere along the horizontal axis, recorded using an HSC with 1500 fps. The red dots indicate the measurements, the black line is the parabolic fit $x_{fit} = c_2 t^2 + c_1 t + c_0$. 

\[ x_{fit} = c_2 t^2 + c_1 t + c_0. \]
**Figure S9. Trajectory and velocity of the electrical pendulum.** Oscillatory motion of the center of mass of the conductive sphere (dataset A): the trajectory of sphere along the horizontal axis (above); the velocity along the x axis (down). Both the horizontal coordinate and the velocity are expressed as a function of time (in seconds).

Following the approach of Khayari and Pérez and Drews et al., the charge acquired by the conductive sphere in contact with the plate can be determined using the method of image charges that gives (29, 31):

\[
Q_0 = \frac{2}{3} \pi^3 \varepsilon_a R_s^2 |\vec{E}_{int}| \tag{Eq.S1}
\]

Where \( \varepsilon_a \) is the absolute electrical permittivity of air (\( \varepsilon_a = \varepsilon_r \cdot \varepsilon_0 = 8.859 \cdot 10^{-12} F \ m^{-1} \)).

Then the electric force exerted on the metal sphere is:

\[
\vec{F}_e = Q_0 \vec{E}_{int} \tag{Eq.S2}
\]

The drag force acting on the sphere, considering zero the velocity of the air (as measured with the meteorological station) is:

\[
\vec{F}_D = -\frac{1}{2} \rho_a C_D A_s v_s^2 \hat{v}_s \tag{Eq.S3}
\]

Where \( \hat{v}_s \) denotes the velocity versor, \( \rho_a = 1.2 \ kg \ m^{-3}, A_s = \pi R_s^2, v_s \) is the velocity of the sphere and \( C_D = \frac{24}{Re_p} \cdot \left( 1 + 0.15 \ Re_p^{0.687} \right) + \frac{0.42}{1 + \left( \frac{42500}{Re_p^{1.16}} \right)} \) is the drag coefficient of Clift and Gauvin for spheres (49). The magnitude of the electric field \( |\vec{E}_{int}| \) can be constrained solving the equation of motion:

\[
m_s \frac{d\vec{v}_s}{dt} \hat{v}_s = -\frac{1}{2} \rho_a C_D A_s v_s^2 \hat{v}_s + \frac{2}{3} \pi^3 \varepsilon_a R_s^2 |\vec{E}_{int}|^2 \hat{v}_s \tag{Eq.S4}
\]

We numerically solved Eq.S4 using Matlab ODE45. Several values of \( E_{int} = |\vec{E}_{int}| \) are tested in the inversion process by means of an iterative process from the minimum \( E_{int,min} = 100 \ \frac{kv}{m} \) to the maximum \( E_{int,max} = 1000 \ \frac{kv}{m} \). The best fit is found minimizing the sum of the absolute values of the differences between the computed and the observed positions at each time along the trajectory, from the initial time of integration \( (t_0) \) to the final time \( (t_{end}) \). The initial time \( t_0 \) corresponds to the first point of the trajectory \( x_0 \) where we observe a stable motion of the sphere after its impact with the plate. The initial condition on the velocity, \( v_s(t_0) \), is calculated from the HSC images. The procedure is repeated independently for each of the 21 parabolic trajectories of dataset A and 50 parabolic trajectories of dataset B (Fig. S10). Applying the error propagation on the mean we find the final values for the electric field in the two dataset: \( E_{int,A} = 591 \pm 5 \ \frac{kv}{m} \) and \( E_{int,B} = 630 \pm 21 \ \frac{kv}{m} \).
Figure S10. Electric field inside the HVCP. Electric field inside the copper plates according to the inversion of the equation of motion of dataset A and dataset B.
S7. Estimation of the uncertainty associated with the measured electric field by means of a combination of laboratory and field values

The aim of this section is to quantify the uncertainty related to the field measurements of the electric field $E_{\text{field}}^k$ obtained with the field mill ($\sigma_{\text{field, tot}}^k$) at a distance $k$ from the center of each copper plate. In order to do so, we combine field and laboratory measurements to improve the final estimation. The statistical component of the uncertainty ($\sigma_{\text{field, stat}}^k$) is derived from laboratory experiments that allowed constraining the relative error due to the intrinsic variability of the apparatus ($r_{\text{lab, stat}}^k$). On the other hand, the systematic relative error ($r_{\text{field, sys}}^k$) associated with the instrument is given by the manufacturer of the field mill and it can be directly applied to the electric field measured during the volcanic fallout.

$$
\sigma_{\text{field, tot}}^k = \sqrt{(\sigma_{\text{field, stat}}^k)^2 + (\sigma_{\text{field, sys}}^k)^2} \approx \sqrt{(r_{\text{lab, stat}}^k \cdot E_{\text{field}}^k)^2 + (r_{\text{field, sys}}^k \cdot E_{\text{field}}^k)^2}
$$

This approach allows having a precise evaluation of the statistical component of the global error, that was not possible to obtain under the conditions occurred during the volcanic fallout.

In the laboratory we performed 54 measurements per plate, placing the probe of the field mill along the axis perpendicular to the center of the plate (6.9 cm from the bottom part). The RH = 30%, the room temperature of $T = 22$ °C and the height were chosen in order simulate the conditions occurred during the fallout. A single measurement of the electric field in the laboratory is described as $E_{\text{lab, i}}^k$, where $i = \{1,2,...,N_i\}$ denotes the $N_i$ repetitions done at a distance $k$ from the plate. $\bar{E}_{\text{lab}}^k$ is the average value of the electric field at location $k$. Each single measurement is described in terms of a statistical component of the uncertainty $\sigma_{\text{lab, i}}^k$ given by the standard deviation:

$$
\sigma_{\text{lab, stat}}^k = \sqrt{\frac{1}{(N_i - 1)} \sum_{i=1}^{N_i} (E_{\text{lab, i}}^k - \bar{E}_{\text{lab}}^k)}
$$

(Eq.S6)

Following Taylor (1997) we assume that all the single measurements are characterized by the same statistical uncertainty $\sigma_{\text{lab, i}}^k = \sigma_{\text{lab, stat}}^k$. It is worth noticing that the error on the mean value is $\sigma_{\text{st}}^k = \frac{\sigma^k}{\sqrt{N_i}}$ (but this is not the case here, since we want to use the variability of the
dataset at a distance $k$ to constrain $\sigma_{\text{stat}}^k$, i.e. $\sigma_{\text{stat}}^k = \sigma_i^k$). Knowing $\sigma_{\text{lab,stat}}^k$ and $E_{\text{lab,i}}^k$, $r_{\text{lab,stat}}^k$ can be evaluated as the average of the relative errors: $r_{\text{lab,stat}}^k = \frac{\sigma_{\text{lab,stat}}^k}{\bar{E}_{\text{lab}}} \left( \sum_{i=1}^{N_i} \frac{1}{E_{\text{lab,i}}} \right)$.

In the quantification of the global uncertainty associated with the field measurements we assume that the systematic error of the field mill, $\sigma_{\text{sys}}^k$, is equal to a 10% of the displayed value ($\alpha_{\text{sys}} = 0.1$) according to the manufacturer. In terms of the relative error we find: $r_{\text{field,tot}}^k = \sigma_{\text{field,tot}}^k / E_{\text{field}}^k$.

| $x$ | 0.03 | 0.04 | 0.05 | 0.06 | 0.07 | 0.08 | 0.09 | 0.10 | 0.15 |
|-----|------|------|------|------|------|------|------|------|------|
| $r_{\text{field,tot}}^k$ | 0.12 | 0.10 | 0.12 | 0.11 | 0.12 | 0.13 | 0.17 | 0.20 | 0.27 |

Table S1. Relative error (positive plate). Relative error on the measured electric field for the positive plate

| $x$ | 0.03 | 0.04 | 0.05 | 0.06 | 0.07 | 0.08 | 0.09 | 0.10 | 0.15 |
|-----|------|------|------|------|------|------|------|------|------|
| $r_{\text{field,tot}}^k$ | 0.15 | 0.11 | 0.10 | 0.10 | 0.11 | 0.10 | 0.12 | 0.13 | 0.19 |

Table S2. Relative error (negative plate). Relative error on the measured electric field for the negative plate