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Key Points:
- Natural observations on the influence of temperature and water content on volcanic lightning have been modelled in laboratory experiments.
- The magnitude of electrification and discharging in plumes decreases significantly with added water.
- At higher temperatures a decrease in magnitude of individual discharges is observed, while the total neutralized charge stays similar.

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Electrification of Experimental Volcanic Jets with Varying Water Content and Temperature

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Abstract Volcanic lightning—a near ubiquitous feature of explosive volcanic eruptions—possesses great potential for the analysis of volcanic plume dynamics. To date, the lack of quantitative knowledge on the relationships between plume characteristics hinders efficient data analysis and application of the resulting parameterizations. We use a shock-tube apparatus for rapid decompression experiments to produce particle-laden jets. We have systematically and independently varied the water content (0–27 wt%) and the temperature (25–320 °C) of the particle-gas mixture. The addition of a few weight percent of water is sufficient to reduce the observed electrification by an order of magnitude. With increasing temperature, a larger number of smaller discharges are observed, with the overall amount of electrification staying similar. Changes in jet dynamics are proposed as the cause of the temperature-dependence, while multiple factors (including the higher conductivity of wet ash) can be seen responsible for the decreased electrification in wet experiments.

Plain Language Summary Volcanic explosive eruptions are accompanied by lightning strikes generated from the volcanic dust cloud. Here we have experimentally studied the effects of atmospheric water and plume temperature on the frequency and intensity of the lightning strikes. The results will feed a model for the use of volcanic lightning strikes to estimate plume contents and intensity.

1. Introduction

Volcanic lightning is the result of ash electrification in volcanic plumes. It has been observed at volcanoes all around the world, representing a large variety of magma compositions and eruptive styles (Mather & Harrison, 2006; McNutt & Williams, 2010; Nicoll et al., 2019). It is also associated with several geophysical signals that can be used for volcano monitoring from a safe distance and especially in unfavorable weather conditions (Behnke & McNutt, 2014).

Several observation techniques have been used at active volcanoes to characterize the electrical activity of eruptive plumes, including measurements of electric field variation (R. Anderson et al., 1965; Kikuchi & Endoh, 1982; Miura et al., 2001), direct measurements of fallout-particles (Gilbert et al., 1991; Miura et al., 2001), and volcanic lightning mapping (Behnke et al., 2013; Thomas et al., 2007). In attempts to correlate variations in the electrical activity with plume dynamics, some of these studies have employed multiparameter data sets, including infrasound data (J. F. Anderson et al., 2018; Cimarelli et al., 2016; Haney et al., 2018; Smith et al., 2017), high-speed video recordings (Cimarelli et al., 2016), magnetotelluric measurements (Aizawa et al., 2010; Aizawa et al., 2016), and seismic data (Smith et al., 2018).

Among the noninductive charging mechanisms thought to be operating during volcanic ash emissions, tribo-electrification (Aplin et al., 2016; Cimarelli et al., 2014; Harrison et al., 2010; Houghton et al., 2013; Méndez Harper & Dufek, 2016) and fracto-electrification (Aplin et al., 2016; James et al., 2000; James et al., 2008; Méndez Harper et al., 2015) are considered the most relevant in explosive eruptions where magma fragmentation and consequent production of turbulent particle-laden jets are naturally produced.

Mechanisms more similar to the electrification generated in meteorological thunderclouds (ice-graupel interaction, e.g., Stolzenburg et al., 1998) have also been proposed, as (1) water vapor is abundant in volcanic plumes and (2) ice formation and riming of ash particles are observed within the plume which can reach the upper troposphere (Arason et al., 2011; McNutt & Williams, 2010; Thomas et al., 2007; Williams & McNutt, 2005). The occurrence of electrical discharges in explosive eruptions triggered or controlled by the presence of external water has drawn attention to the effect of water vapor in the generation of volcanic plume electrification. Documented examples of volcanic lightning produced in water-rich conditions include the eruptions of Capelinhos, 1957–1958 (Machado et al., 1962), Surtsey, 1963 (R. Anderson et al., 1965), Eyjafjallajökull, 2010 (Arason et al., 2011; Harrison et al., 2010; Petersen et al., 2012), Bogoslof, 2017–2018 (Haney et al., 2018; Van Eaton et al., 2018), and Anak Krakatau, 2018 (Prata et al., 2019).

Due to the complexity of natural systems, field observations alone cannot resolve the effects of the different factors contributing to the electrification of volcanic plumes. Nevertheless, despite the potential first-order importance of (1) higher temperatures and (2) the ubiquitous presence of water vapor in volcanic plumes, the vast majority of experiments to date have neglected the effects of water and temperature on electrification and discharge mechanisms.

In our study we quantify the effects of water content and temperature of experimentally generated jets on their resulting electrical activity. We consider the effects of both parameters on the charge and discharge modality of volcanic ash and discuss natural occurrences in light of our experimental results.

2. Methods

2.1. Experimental Setup

Following on from previous experimental studies of volcanic lightning (Cimarelli et al., 2014; Gaudin & Cimarelli, 2019), our experiments have been carried out using a 26-mm diameter autoclave apparatus, in which natural ash samples are pressurized to 9 MPa by the addition of argon gas. A furnace is placed around the autoclave to heat the samples to temperatures relevant for volcanic plumes (room temperature [RT, 23–25 °C] to 320 °C). Temperatures in this study are those at which the gas-particle mixture was thermally equilibrated before ejection. For the analysis of the influence of water content, different amounts of water (<27 wt%) were added before pressurization and heating of the ash and water mixture. Once target temperatures and pressures are attained, the mixture is rapidly decompressed to generate a particle-laden jet. The jet is free to expand into a vertical, ~3-m-high collector tank, situated above the autoclave. Argon was chosen as gas carrier phase due to its similar dielectric properties to air and for preventing oxidization of the sample at high temperatures.

In previous experimental studies of volcanic lightning, the discharges produced by the jets were measured using either antennas (Cimarelli et al., 2014) or a Faraday cage (FC) placed inside the collector tank (Gaudin & Cimarelli, 2019). In this study, the whole collector tank has been insulated to serve as a FC (Figure 1). By this, the expanding particle-laden jet is fully contained within the FC, enabling precise analysis of the associated charge throughout the experiment. Moreover, the new setup enables easier operation of the apparatus and analysis of the data, thus improving repeatability of the experiments and consistency of the measurements.

The setup is equipped with two pressure sensors that record the static pressure inside the autoclave (where the gas-particle mixture is placed) and the dynamic pressure at the nozzle (where the ejected mixture enters the FC). A thermocouple at the bottom of the gas-particle mixture in the autoclave records the temperature of the mixture before it undergoes decompression. In some experiments a high-speed camera was additionally used to record the rapid evolution of the resulting jets entering the FC at frame rates of up to 30,000 fps.

2.2. Samples

We used pre-sieved loose tephra from the lower unit of the 13-ka Laacher See eruption (provided by ROTEC GmbH in Mülheim-Kärlich). The ash is phonolitic in composition and contains 5–8% phenocrysts, mainly sanidine, plagioclase, and clinopyroxene (Wörner & Schmincke, 1984). Only the nonwashed grain size fraction between 90 and 300 μm was used for the experiments. The concentration of very fine ash (<63 μm) for samples of this grain size fraction is ~3.2 wt% (Gaudin & Cimarelli, 2019). Douillet et al. (2014) derived four different grain shape parameters for the same type of samples, also provided by ROTEC GmbH. Sphericity (0.81 ± 0.07), symmetry (0.87 ± 0.02), aspect ratio (0.68 ± 0.02), and convexity (0.98 ± 0.02) were all relatively constant over a wide range of grain sizes. As the samples are granular and dominantly fine-grained, particle fragmentation has only a minor influence on our experiments, so that triboelectrification can be expected to dominate the electrification process.

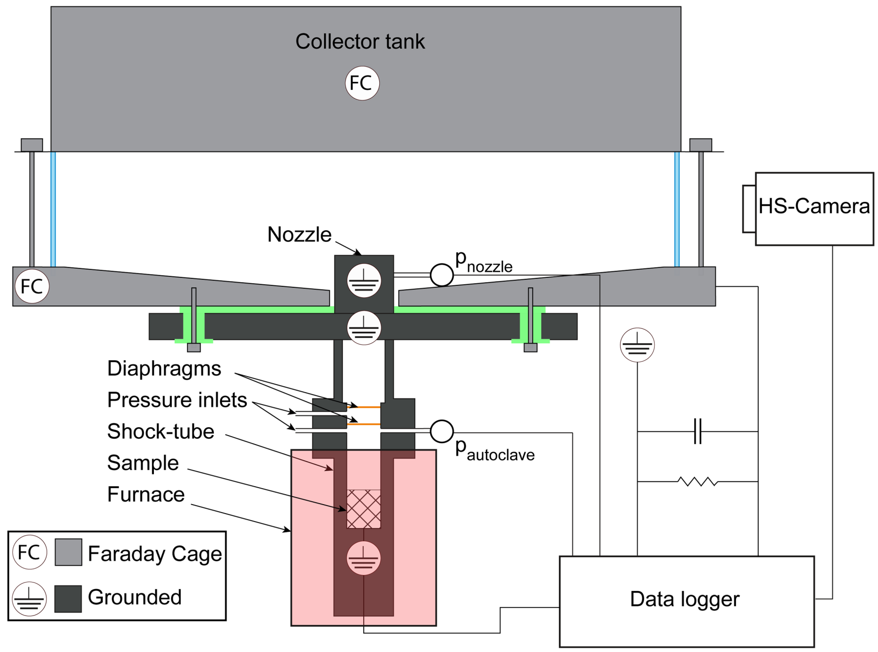

Figure 1. The experimental setup. The autoclave containing the sample can be surrounded by a furnace. Argon gas is conveyed into the autoclave through the gas inlets until the target pressure is reached. Above, the nozzle is the uppermost grounded part of the setup. The collector tank acts as a Faraday cage. A high-speed camera is used to record the experiments. All sensors are connected to a data logger.

2.3. Experimental Strategy

Given the intrinsic difficulties in directly measuring the amount of water in volcanic plumes in the field, there are very few quantitative data (Williams & McNutt, 2005). Previous studies indicate that water contents of pre-eruptive magmas typically are 1–8 wt% (Carey et al., 1995; Gardner et al., 1995; Grove et al., 2002; Roggensack et al., 1997; Sisson & Layne, 1993; Wallace & Gerlach, 1994), with some studies reporting up to 15 wt% (Grove et al., 2005, 2003). In our experiments investigating the influence of water, 0–27 wt% of water have been added before bringing the samples to the target temperature and pressure. In these experiments the temperature was kept constant at 320 °C, so that the amount of added water was the only variability. The conditions at 9 MPa and 320 °C for this set were chosen to ensure that the water is predominantly in the gas phase before the rapid decompression.

Temperature was varied for dry experiments between RT and 320 °C in order to cover the range of temperatures expected in natural volcanic plumes (Suzuki et al., 2005; Suzuki & Koyaguchi, 2012).

Two main parameters were used for the analysis of the electrical activity. First, the net charge of the gas-particle mixture entering the FC is computed as the integration of the current leaving the FC. Discharges are detected within that signal as sharp drops of the net charge. The cumulative discharge is the sum of all charge neutralized by the electrical discharges produced by the particle-laden jets and is computed following Gaudin and Cimarelli (2019). It is worth noting that only those discharges that change the net charge can be measured by the FC, that is, when they go from inside to outside the FC. In the setup used in this study, the only possibility for that is for discharges directed from the jet to the nozzle or the autoclave.

High-speed video measurements of the opening angles and gas velocities were used to study the evolution of the jets. Jet opening angles were measured 3 cm above the nozzle and were given here as a deviation from vertical. Gas velocities were measured tracking the front of the shock wave propagating into the FC. However, the need to use fine-grained particles to produce electrification and discharges (Gaudin & Cimarelli, 2019) hindered further analysis and comparison with previous studies, particularly those on particle velocity profiles (Cigala et al., 2017) and the influence of the Mach disk and over-pressurized region around the nozzle (Méndez Harper et al., 2018), as individual particles are not visible in the jet.

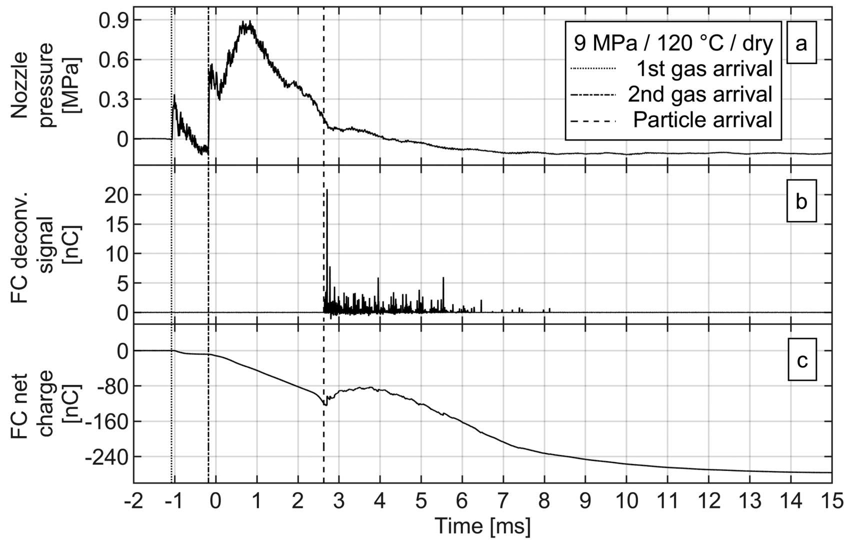

Figure 2. A typical experiment recording (9 MPa initial pressure, 120 °C, dry). Nozzle pressure (a), the deconvoluted signal of the FC (b), and the net charge (c) are all plotted against experimental time. First and second gas arrival, as well as particle arrival, are marked with vertical lines.

3. Results

3.1. Characteristics of All Experiments

The results shown here are taken out of the data set presented in Stern et al. (2019). Common features can be recognized in all experiments as also reported in previous studies using a similar setup (Cimarelli et al., 2014; Gaudin & Cimarelli, 2019). The pressure signal at the nozzle clearly shows (1) the escape of argon gas contained between the two diaphragms as first, minor peak; (2) the arrival of the gas escaping the sample chamber producing a much larger and broader peak; and (3) the arrival of the particles (Figure 2a). Discharges coincide with the arrival of the particles at the nozzle and are thus attributed to the charge carried by solid particles within the expanding jets. Furthermore, discharges appear between 2 and 9 ms after burst of the diaphragms (Figure 2b). We observe the introduction of dominantly negative charge into the FC that is neutralized by discharges of opposite polarity (Figure 2c).

3.2. Influence of Water

The largest change visible in physical jet parameters at higher water contents is the opening angle of the expanding jet (Figure 3a), which increases from 29° for dry experiments to 42° for experiments with 1.8 wt% of water. With higher water contents, the increment reduces until reaching a maximum of 55° for 27.0 wt% of water. The video analysis reveals that gas velocities (measured as the front of the condensing gas under expansion) are not substantially affected by the variations in water concentration.

Dry experiments at 320 °C produce around 215 nC of discharge (Figure 3b). Already for the smallest amount of added water (1.8 wt%), this value decreases to 87 nC. At higher water contents, the amount of discharge progressively anneals to zero.

3.3. Influence of Temperature

At higher temperatures, we observe an increase of the gas velocity. This is linked to the speed of sound largely depending on the gas temperature (White, 2011): The Mach number remains stable against the temperature variations in our study (Figure 4a). The jet opening angles are observed to vary much less than for the experiments with added water, only showing a decrease from 35° at RT to 30° at 120 °C. At higher temperatures, the opening angle remains relatively constant (Figure 4a). A slight, linear increase in nozzle pressures is further observed from ~0.85 MPa at RT up to ~1.05 MPa at 320 °C.

The relationship between the temperature and the detected total amount of discharge is more complex: There is no obvious relationship of the cumulative discharge with temperature (Figure 4b). A clear correlation can only be seen in the magnitude of individual discharges within the investigated experiments

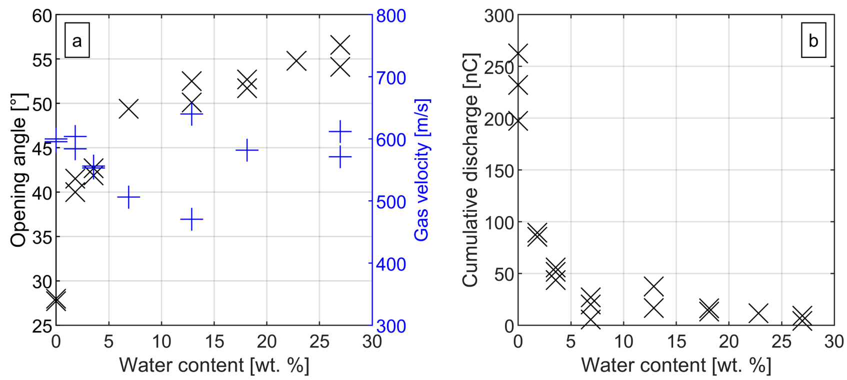

Figure 3. Results for the analysis of the influence of water. All experiments were done at 320 °C with variable amounts of water. Panel (a) shows physical jet parameters; opening angle (×) and gas velocities (+) and panel (b) shows cumulative discharge.

(Figure 4c) with a decrease from ~10 nC (RT) down to 1 nC (220 °C). At higher temperatures a larger number of discharges are neutralizing similar amounts of total charge.

We also observe a variation in the charge buildup in our experiments with varying temperatures (Figure 4d). First, charge builds up slowly in the early phases of the experiments at RT and generally more steadily than in experiments at higher temperatures with constant rates between 1 and 7 ms. In contrast, we observe the highest charging rates at higher temperatures early on in the experiments, with decreasing rates once discharges are firstly observed. Second, in the experiments at high temperatures, we observe a decrease in the net charge that persists for ~2 ms after the onset of discharging (visible on the graphs as sharp drops in the curves). The charging for RT experiments seems much more stable, and no significant decrease of the net charge through discharging can be observed at any time. Later discharges at lower temperatures may still be of similar magnitude as early discharges, while for higher temperatures (especially >120 °C), those late discharges are generally smaller than the early ones. Also, even though more charge is generated in the experiments at RT, less discharge occurs (Figure 4b), caused by an additional increase of the net charge after the last discharge at higher temperatures, which was not observed for RT experiments. Finally, the time in which discharges occur is much shorter in the RT experiments, lasting from ~3 to 6 ms (see Gaudin & Cimarelli, 2019), whereas at higher temperatures, we observe discharges lasting from 2.5 to 9 ms (Figure 4e).

4. Discussion

4.1. General Characteristics

Our results indicate that in our experiments the charge is carried solely by the particles and that discharging is observed as soon as the particles escape the nozzle—a good agreement with previous studies (Cimarelli et al., 2014; Gaudin & Cimarelli, 2019). In contrast to previous experiments, the quality of the charge data recording of the new setup enables insights into the charging processes of the particles, illustrated by the fact that the net charge reaches an asymptotic maximum at the end of each experiment (Figure 4d). We observe that ejected particles carry predominantly negative charges. This indicates that positive charges must be transferred by the particles to the grounded metal autoclave upon decompression, thus being neutralized before the jet enters the FC.

4.2. Influence of Water

The effect of external water in contact with magma is known to further enhance the explosivity of a volcanic eruption. This has been proposed to occur through a mechanism best known as fuel coolant interaction (Wohletz, 1986; Wohletz et al., 2012). For water/magma mass ratios of 0.3 to 0.4, such interaction is most efficient, whereas for higher ratios, the expansion would produce saturated steam with important implications on the buoyancy of the produced eruptive plume (Koyaguchi & Woods, 1996).

Previous studies investigating the influence of relative humidity (RH) on the amount of charge generated by triboelectrification have produced contrasting results, either reporting a general decrease (Greason, 2000; Harper, 1957; Schella et al., 2017) or an increase at low RH followed by a decrease above ~30% RH

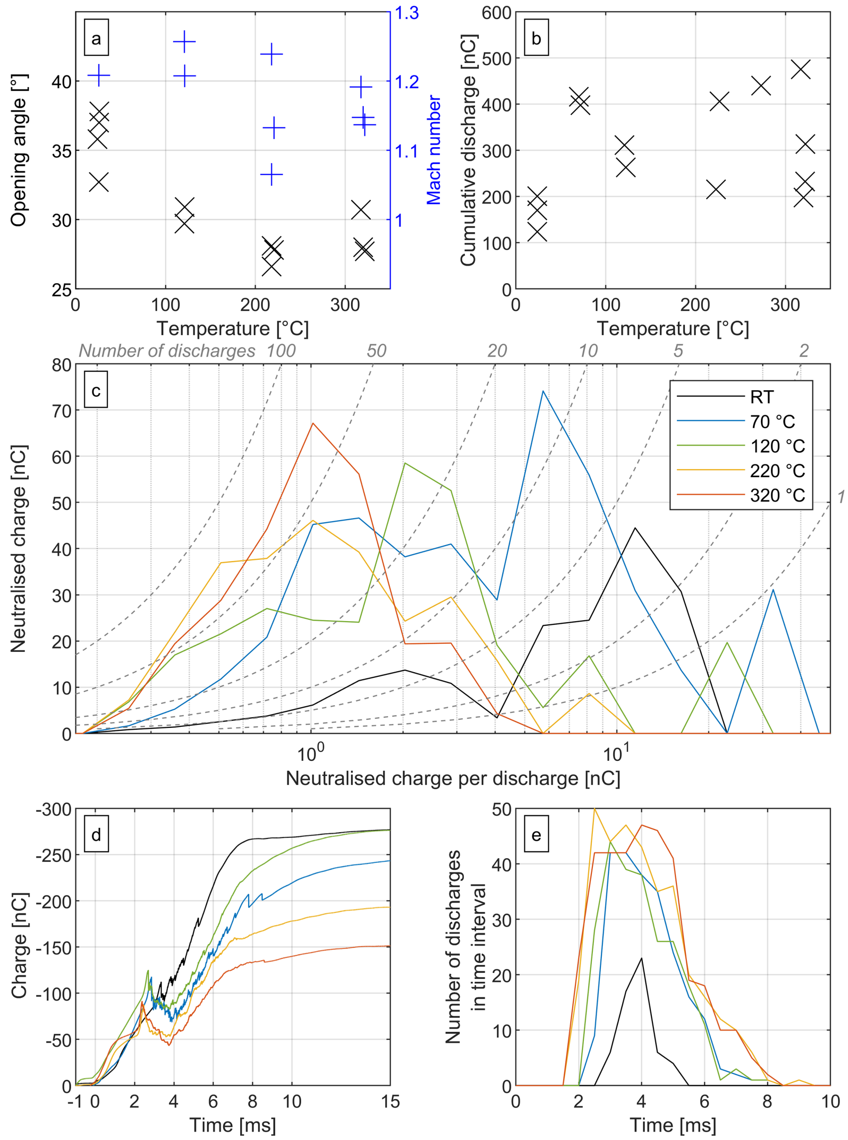

Figure 4. Results for the analysis of the influence of temperature in the experiments. All experiments were done at varying temperatures without added water. Panel (a) shows physical jet parameters: opening angle (×) and Mach number (+), panel (b) displays the cumulative discharge, panel (c) shows magnitude distribution of individual discharges, panel (d) displays the charge buildup (instantaneous drops are discharges), and panel (e) shows a histogram of the number of discharges in 0.5-ms intervals.

(Hiratsuka & Hosotani, 2012; Xie et al., 2016). In our experiments, we also observe a decrease in the total charge neutralized for higher concentrations of water. One reason for this is that the increased jet opening angle drives the charged particles significantly further apart. Following Paschen's law (Paschen, 1889), the breakdown voltage would consequently increase, thus inhibiting the discharge. The increase in the opening angle of the jet is related to the much higher expansivity of a wet jet.

Similarly to our experiments, James et al. (2000) used a Faraday cup (volume 4×10^{-4} m^3) to determine the effect of charge of volcanic ash under controlled RH conditions and observed an enhanced charge decay over the whole duration of the experiment (250 s), producing difficulties for the analysis of the electrical signal. However, these issues can be neglected in our experiments, because we measure the number and magnitude of discharges, rather than variations in the net charge and our measurements are done over a much shorter timeframe (20 ms), during which the particle-laden flow does not expand to fill the volume of our FC (0.4 m^3), minimizing the effect of charge leakage.

The decrease in produced cumulative discharge is also analogous to an observed increase of conductivity at increased water concentrations in ash (Wardman et al., 2012), which could lead to an increased charge transfer during collisions, hindering effective charge clustering. Moreover, the high RH that is to be expected in the jet would additionally lower the charge generated on the particles due to the higher conductivity of humid air (e.g., Greason, 2000; Schella et al., 2017).

4.3. Influence of Temperature

Very few studies have investigated the influence of temperature on triboelectric charging processes, especially in the temperature range relevant for volcanic plumes. Greason (2000) observed a drop of charge generation in experiments by 10% to 50% from 10 °C to 30 °C, with the larger drops associated with experiments at lower RH. Studies at higher temperatures are scarce and not directly applicable to the volcanic case. In these studies, it was not possible to determine whether material parameters or real triboelectric effects are the main reason for the observed drop of charging efficiency at temperatures above the freezing point (Lu et al., 2017; Su et al., 2015; Wen et al., 2014).

In our dry experiments at variable temperatures, the slight decrease in jet opening angle from RT to higher temperatures could indicate a higher particle (and charge) density within the jet, allowing for more particle collisions. However, we do not observe an increase in the built-up charge; thus this seems unlikely. Also, the increase of gas velocity and nozzle pressure (both around 25%) seems insufficient to explain the decrease of neutralized charge by an order of magnitude.

The total neutralized charge shows a wide spread at higher temperatures, underlining that there is a natural variability in the system. Our experiments show that the magnitude of individual discharges decreases and that their number increases with increasing temperature.

Previously, Cimarelli et al. (2014) linked the magnitude of individual discharges to the electrical potential between charge clusters that develop during experiments at RT. Higher temperatures can however introduce further complexities into the system, changing particle and gas properties and the overall jet dynamics. There is paucity of data available for the electrical properties of volcanic ash. However, studies on soil suggest that the conductivity does not change significantly above the freezing point (Institute of Electrical and Electronics Engineers, 2013). For temperatures above 300 °C it has been shown that there is a positive correlation of the temperature with conductivity (Alvarez et al., 1978), although even in that case it was not determined to be a dominating parameter. As the maximum temperature used in our study is 320 °C, we infer that material effects related to the ash are minor.

Considering the effect of gas electrification at high temperature, Galli et al. (2019) showed that the breakdown voltage for argon gas increases by around 25% for an increase in temperature from RT to 400 °C at a pressure of 1 MPa, which is very similar to the nozzle pressures measured here (0.8 to 1.1 MPa). For our experiments this would mean that the jets at elevated temperatures need more charge buildup to discharge and neutralize larger charges simultaneously. In contrast, we observe smaller discharges when the temperature is increased, so that we can deduce that the increased breakdown voltage caused by gas effects is also not a dominating parameter.

The observed decrease of net charge at the onset of discharging activity at higher temperatures for up to 3 ms and the lack of that observation in RT experiments emphasizes a stronger role of the jet dynamics.

While the introduction of additional net charge remains very effective at RT until 7 ms, we observe very few discharges after ~5 ms. In contrast, the much slower charging rates at higher temperatures persist for longer times and allow for discharges to be detected as late as 9 ms. Even though this seems to be at least partly linked to the charging efficiency, and thus the amount of particle-particle collisions in the autoclave, influences of the charge separation and discharging mechanisms cannot be ruled out.

While the overall amount of charge buildup is generally similar for all temperatures, the total amount of neutralized charge is larger at higher temperatures. A possible explanation is given by a prolonged, decreased distance between particles and thus charges, which would reduce the breakdown voltage following Paschen's law (Paschen, 1889). This could also explain the decreased magnitude of late discharge in experiments at elevated temperatures. Both would indicate that charge clusters are continuously driven back together, by, for example, an increased amount of convection of multiple, smaller charge clusters as previously suggested by Cimarelli et al. (2014). On the other hand, at RT we observe that late discharges may be larger, caused by an increased distance between charge clusters. Here we therefore see larger, but fewer charge clusters that tend to produce few, but large discharges instead of multiple, smaller ones.

Overall, we suggest that the experiments at higher temperatures have an increased turbulence that causes charging to be more rapid and discharging to be much more efficient, producing much higher number of smaller discharges.

4.4. Implications for Natural Volcanic Lightning

Our experiments largely reproduce features that have been observed in recent cases of volcanic plume electrification during explosive eruptions displaying extensive magma-water interaction.

The decrease in discharge generated at higher water concentrations is consistent with observations for the activity of Bogoslof in 2017 (Van Eaton et al., 2018), for which it has been suggested that plumes that do not extend into freezing temperature altitudes produce less discharge when they originate from submerged vents, compared to subaerial vents. This was directly associated to the water concentrations within the plume being higher for the submerged case. For Bogoslof, it was found that in plumes reaching freezing level conditions, the formation of ice crystals and hail can promote charging. Otherwise, the presence of water would hinder effective electrification. For plumes that reach freezing levels and originate from submerged vents, it was observed that they may generate fewer discharges close to the vent but more discharges once they reach freezing conditions at higher altitudes. The same effect had already been suggested for the 2010 activity of Eyjafjallajökull, where the plume showed a significantly higher number of discharges when reaching freezing levels in the atmosphere (Arason et al., 2011). Thus, water in liquid state may inhibit discharging, but once it is transformed into ice, it enhances it. Similarly, the eruption of Anak Krakatau in December 2018 was accompanied by an unprecedented large number of discharges. The rate of discharging increased by orders of magnitudes after the tsunamogenic collapse (i.e., after the change from a subaerial to submerged vent) as recorded by multiple long-range antenna systems (>100,000, e.g., Earth Networks Global Lightning Network). Analysis of satellite data suggests that vigorous updraft of sea-water vapor triggered by the phreatomagmatic activity generated a sustained and highly electrified ice-rich and ash-poor plume (Prata et al., 2019), possibly enhancing the amount of electrification.

5. Conclusions

In this study we have successfully reproduced volcanic jet electrification in the lab, constraining the influence of different initial temperatures and water contents on the resulting electrical activity. Our focus on triboelectrification and the over-pressurized region of the jet helps constraining the electrification mechanism at play in the jet region of an explosive, volcanic eruption in which water is the dominant volatile species.

We show that already small additions of water in the sample are sufficient to reduce the produced electrification by an order of magnitude. This is likely caused by multiple effects: (1) an increase of the jet opening angle with more water added, driving the particles and charges further apart; (2) an increase in the conductivity of the ash, enhancing charge transfer during collisions and thus hindering charge separation; and (3) an increase in conductivity of humid air. This agrees with and further constrains the observations made at recent eruptions of, for example, Eyjafjallajökull (2010), Bogoslof (2017–2018), or Anak Krakatau (2018–2019).

Temperature has a much more complex influence on charge generation and discharging. We observed smaller discharges at higher temperatures, with the mean magnitude decreasing linearly by an order of magnitude from RT to 220 °C. The total magnitude of charging and discharging is less affected by the temperature. It can be inferred that variations in the jet dynamics are the governing parameter determining changes in the electrical behavior of the jet in our experiments.

This study has yielded the first experimental constraints for the influence of water content and temperature on the electrification of volcanic plumes and offers direct applicability for natural cases. The water content, in particular, has a dramatic effect on the generated electrical activity and has to be considered as one of the most dominating parameters in natural phenomena.

Acknowledgments

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