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First In Situ Observations of Gaseous Volcanic Plume Electrification

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Abstract Volcanic plumes become electrically charged, often producing spectacular displays of lightning. Previous research has focused on understanding volcanic lightning, primarily the large electric fields produced by charging of ash particles. Here we report on the previously overlooked phenomenon of volcanic plume electrification in the absence of detectable ash. We present the first in situ vertical profile measurements of charge, thermodynamic, and microphysical properties inside predominantly gaseous plumes directly above an erupting volcano. Our measurements demonstrate that substantial charge (at least $\pm$8,000 pC/m^3) is present in gaseous volcanic clouds without detectable ash. We suggest that plume charging may be enhanced by the emission of radon gas from the volcano, which causes ionization. This presents a hitherto unrecognized, but likely to be common, mechanism for charge generation in volcanic plumes, which is expected to modulate plume characteristics and lifetime. This process is currently neglected in recognized mechanisms of volcanic plume electrification.

Key Points:
- We present the first in situ profiles of thermodynamic, microphysical, and charge properties in a gaseous volcanic plume above an erupting volcano
- Gaseous plumes with no detectable ash are, unexpectedly, significantly charged (up to $\pm$8,000 pC/m^3)
- A new mechanism for plume electrification related to radon emission from volcanic vents is proposed to explain the charge observations

Supporting Information:
- Supporting Information S1

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1. Introduction

Volcanic plumes become charged through a variety of mechanisms including fractoemission, triboelectrification, and hydrometeor-ash particle interactions (Aplin et al., 2016; Mather & Harrison, 2006). Fractoemission typically occurs close to the vent, where explosive activity causes fragmentation of magma (James et al., 2000), leading to the emission of photons, electrons, positive ions, and charged particulates (Lane et al., 2011). Triboelectrification is a type of contact electrification caused by charge transfer at the surfaces of particles (Houghton et al., 2013; Lacks & Levandovsky, 2007; Méndez-Harper & Dufek, 2016). Ice-ash particle interactions have been considered analogous to the ice-graupel interactions generating charge in thunderstorms (Arason et al., 2011; Williams & McNutt, 2005).

Understanding of the charging mechanisms above comes from laboratory experiments (Cimarelli et al., 2014; Méndez-Harper et al., 2018) detection of volcanic lightning from (1) lightning mapping arrays (Behnke et al., 2013), (2) global lightning detection networks (Bennett et al., 2010), or (3) high-speed imaging techniques (Aizawa et al., 2016; Cimarelli et al., 2016) and measurements of plume charge overhead from ground based electric field mills (James et al., 1998). The only direct measurements of volcanic plume charge have been made by collecting fallout ash particles (Gilbert et al., 1991; Miura et al., 2002) in a Faraday pail situated a few kilometers away from the eruptive vent. In situ plume charge measurements have, so far, been lacking due to the intrinsic difficulties of working in proximity to active volcanoes.

Renewed interest in understanding the behavior of charged aerosol clouds (including volcanic plumes and dust clouds) has arisen due to realization that existing long-range particle transport models do not accurately predict the transport of large particles (Ryder et al., 2013; van der Does et al., 2018; Weinzierl et al., 2017). Charging modifies the fall speeds of small particles in the atmospheric electric field, changes aggregation rates, and enhances the washout of particles by rainfall (Harrison & Carslaw, 2003). Charge may also act to prolong the transport of particles in substantial electric fields (Ulanowski et al., 2007). Despite all of the aforementioned potential effects of charge on the behavior of aerosols, the lack of in situ charge measurements in volcanic plumes means that the magnitude of particle charging is unquantified, and therefore,

the importance of such mechanisms, for example, long-range transport and particle fallout, is as yet unknown.

A major challenge in understanding the electrification of volcanic plumes is the separation of different charging mechanisms, due to the multiple ash processes simultaneously in action. The plumes studied here present the opportunity to study simplified volcanic plumes of gaseous vapor and liquid droplets in the absence of observable ash. Previous remotely sensed (using surface-based potential gradient [PG] measurements) gaseous plumes were observed to be charged only when substantial ash concentrations were present (Miura et al., 2002). The generally repeatable behavior of the PG perturbations as the various components of the volcanic plume pass overhead has led investigators to suggest that the gaseous component adopts a net positive charge, and the ash particles a net negative charge (e.g., Hatekayama & Uchikawa, 1952), with separation suggested to result from gravitational settling. The opposite charging of gas and ash particles is thought to originate from fractoemission, which occurs due to magma fragmentation, with lab experiments supporting the concept of positively charged gas and negatively charged ash particles (James et al., 2000). The only previous estimate of the charge in the gaseous region of such plumes is reported by Miura et al. (2002), who assumed a point charge geometry and derived 0.2 C from their surface PG measurements, which were made 2–5 km from the crater, when the plume was at an altitude of ~2 km above the sensor.

Here we used newly developed disposable sensors, which can be safely deployed to measure charge directly within a volcanic plume. These represent the first vertical profile measurements of thermodynamic, electrical, and microphysical properties inside a volcanic plume close to its source and provide new information about the magnitude, polarity, and vertical distribution of charge within prevalently gaseous volcanic clouds. Such measurements are relevant to refine our understanding of the electrical structure of volcanic plumes. Further, by focusing only on the gaseous component of volcanic plumes, we demonstrate that volcanic plumes do not require solid ash particles to become electrified, thereby providing evidence for an additional and hitherto unrecognized charging mechanism, which does not involve ash.

2. Stromboli Campaign

A measurement campaign at Stromboli, Italy (38.794°N, 15.211°E, 924 m above sea level [asl]) was conducted from 28 September to 4 October 2017. Normal Strombolian activity is characterized by frequent (about 10 per hour), intermittent, mild explosions (0.01–100 m^3 of tephra) generating weak plumes of a few tens to few hundred meters vertical extent. Passive degassing from the active crater terrace (~600 m asl) represents about 80% of the total SO_2 gas emissions (Tamburello et al., 2012) and substantially contributes to the composition of the persistent plume above Stromboli. During the campaign, the observed persistent plume was primarily gaseous and of small vertical and horizontal extents (positively buoyant up to 2 km above the craters). The persistent plume was mainly fed by passive degassing, punctuated by frequent small explosions (13 per hour on average) occasionally producing dense ash emissions. Instrument stations were deployed near the summit, overlooking the crater terrace and at a distance of ~300 m from the active vents (see Figure S1 in the supporting information) together with an Infratec Variocam thermal infrared camera recording the explosive activity at 60 fps from the summit of the volcano. Sensors deployed included an electrotatic detector developed by Biral ltd, UK. The detector (hereafter BTD (Bennett, 2018)) records displacement current induced on a 0.1-m radius spherical electrode on a 1.5-m mast. Measured current results from temporal fluctuations in the atmospheric electric field and is sampled at 100 Hz with a sensitivity of approximately 10 pA. In situ plume measurements were also obtained by VOLCLAB sensor packages designed at the University of Reading, deployed around the volcano's summit, as well as flown on free weather balloons fitted with standard meteorological radiosondes. The instrument packages consisted of a backscatter sensor (Harrison & Nicoll, 2014) for optical detection of aerosols, miniaturized SO_2 (Alphasense SO2-B4), and a charge sensor (Harrison et al., 2017). The radiosonde charge sensor responds to the displacement current induced on a small spherical electrode by changes in the electric field experienced during its ascent (e.g., Nicoll & Harrison, 2009), providing an average space charge density every ~5 m. No measurable electrostatic discharges were detected by the BTD in any of the plumes during the campaign.

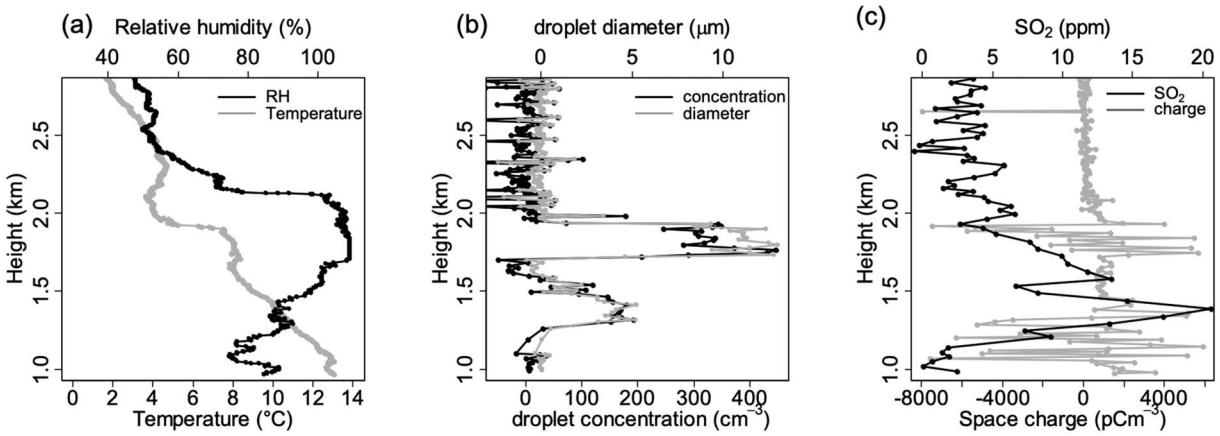

Figure 1. Vertical profiles through the volcanic plume at Stromboli at 11:40 UT on 30 September 2017 measured by a Vaisala RS92 radiosonde and the VOLCLAB sensor package (flight 1). (a) Temperature (±0 .5°C) and relative humidity (±5%), (b) droplet concentration (±100/cm^3) and diameter (±2 μm) measured by the optical backscatter sensor (errors quoted are two standard errors calibrated against the CAPS sensor; Baumgardner et al., 2001), (c) Space charge density (median error ± 500 pC/m^3) and SO_2 concentration (±3 ppm).

3. Vertical Profiles Through Volcanic Plumes

3.1. Near Vent Profile

A Vaisala RS92 radiosonde instrumented with the VOLCLAB sensor package was released from the summit directly through the persistent plume at 11:40:00 UT on 30th September 2017 (see Figure S1b for location of flight 1). At the time of launch the visibility over the launch site was reduced, mostly due to wind directing passive degassing from the vents toward the launch site. A ballistic-dominated explosion with vertically flying ejecta reaching a maximum height of 100 m occurred from 11:35:35 to 11:36:00 UT from the N2 crater, ending 4 min before the launch time (see Figure S2 and associated text for thermal infrared analysis of the explosion). This explosion is not believed to have influenced the large-scale structure of the main plume sampled by the balloon as sufficient time had elapsed for the material injected into the local environment from that event to have settled or been dynamically removed from the measurement area. Moreover, thermal infrared imaging shows a very small thermal anomaly, which confirms that the buoyant plume was mainly gaseous (Figure S2), a conclusion supported by our ground-based sensors which detected no ash when engulfed by the plume. Ash was therefore considered to be negligible as a source of additional charge during our measurements. Figure 1 shows vertical profiles from the instrumented balloon in the elevated plume. The thermodynamic parameters are shown in Figure 1a, droplet properties (including those condensed from gases in the volcanic plume to create, e.g., H_2O and H_2SO_4) measured by the optical sensor in (b), and charge and SO_2 measurements in (c).

Figure 1 demonstrates the presence of two distinct layers detected by the optical backscatter sensor (between 1.2–1.5 km and 1.7–2.0 km asl). The optical properties of the upper layer are characteristic of a meteorological cloud layer (with relative humidity [RH] >100% signifying saturation). Indeed, a broken layer of Stratocumulus (Sc) cloud was observed to form over Stromboli in the few hours before launch (see photographs in Figure S3), which formed only over the land, suggesting an orographic and potentially volcanic influence. From Figure 1a the temperature profile in the upper layer is very different to that within a normal Sc cloud, which typically cools with height through the layer, with a temperature inversion at cloud top. The region inside this cloud stays warm initially, with a sharp cooling toward cloud top. This suggests the entrainment of warm, buoyant (and very likely turbulent) air from the volcanic plume beneath. Figure S4 shows the ambient temperature and RH profile from the ERA5 reanalysis data set (for ~68 km east of Stromboli at 12 UT) compared with the balloon measurements over Stromboli. This demonstrates substantial differences between the two, with no cloud layer in ERA5, suggesting (assuming no model error) a significant influence of Stromboli on the thermodynamic properties of the atmosphere in the vicinity of the island, up to an altitude of ~2.5 km. Evidence for cloud formation directly over Stromboli is also provided in Figure S3, which shows contemporary satellite cloud measurements. A localized cloud is present over Stromboli but not in the surrounding region.

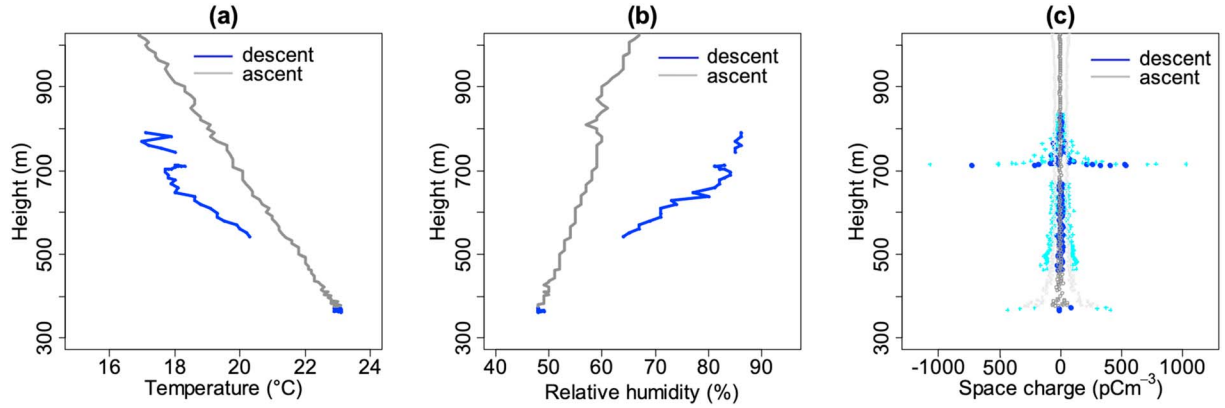

Figure 2. Vertical profiles through the volcanic plume at Stromboli at 11:55:15 UT on 4 October 2017 (flight 2) measured by a Vaisala RS92 radiosonde and the VOLCLAB sensor package. (a) temperature (±0.5°C), (b) relative humidity (±5%), (c) space charge density (the cyan "+" and light gray "x" denote errors in descent and ascent space charge data, respectively). The initial descent through the plume is shown in blue, and ascent in clear air in gray. For references to color in this figure, please see the online version of the article.

Conversely, the thermodynamic properties of the lower layer in Figure 1 display peculiar structure compared with a typical water cloud (e.g., in temperature transitions and gradients and RH <90%, indicating that the air is not saturated). The optical sensor indicates the presence of primarily small droplets <5 μm diameter, with fewer droplets than in the cloud layer above (200/cm^3 compared with 450/cm^3). Figure 1c also demonstrates a peak in SO_2 within this layer suggesting that it is volcanically generated, consisting of a mixture of common volcanic gaseous vapor (including water vapor, CO_2, SO_2, and HCl; Allard et al., 2008), some of which will condense to form small droplets (including H_2SO_4), producing the backscatter signal observed.

Within the upper cloud layer, the space charge profile demonstrates appreciable charge up to ±8,000 pC/m^3 (causing saturation of the charge sensor, so this is likely to be an underestimate), with well-defined layers of positive charge at cloud base and negative charge at cloud top. This is at least 80 times larger than the charge typically observed in normal stratiform water clouds (up to ±100 pC/m^3; Nicoll & Harrison, 2016), with the polarity at cloud top and bottom also reversed. The charge is strongly confined within the region of optical backscatter, suggesting that the charge is associated with the cloud droplets. Some charge separation is observed in this layer via differential gravitational settling as evidenced by the larger, positively charged droplets at the base underlying the smaller, negatively charged droplets at the layer top. In the lower cloud layer, the structure of the charge profile is complex, again with significant charge up to ±8,000 pC/m^3. The charge beneath this layer is highly variable in polarity, suggesting effective mixing within the gaseous plume (hindering any observable charge separation), which is expected given the close proximity (~300 m) to the active vents.

3.2. Profile Far From Vent

A second flight through the plume was made at 11:55 UT on 04/10/17, from halfway down the ridge on Stromboli's southern flank, ~400 m from the main craters (see Figure S1b for location of flight 2). Conditions at the summit were blustery (mean wind speed 7 m/s), and the primarily gaseous volcanic plume was generally swirling below the summit and occasionally up over the top of the ridge. The instrumented balloon was launched as the gaseous plume approached and was underinflated causing it to descend initially through the plume (from launch height of 800 m down to 350 m) and ascend in the clear air away from the volcano. Vertical profiles from flight 2 are shown in Figure 2, where there is evidence of the plume from 800 m down to 650 m (the UHF radiosonde signal, which requires line of sight, was lost intermittently during the descent). From Figure 2a, the temperature inside the plume is cooler than the ambient air, and the RH is considerably higher than ambient, but not fully saturated (maximum RH 87%) suggesting water vapor and gaseous constituents similar to the lower layer in flight 1 (SO_2 measurements were not available for flight 2). From Figure 2c a distinct layer of space charge was encountered as the balloon descended into the plume, up to ±600 pC/m^3 (the charge sensor saturation limit, itself more sensitive than the device used in flight 1). Both polarities of charge are present, with no obvious layering in the plume region where the

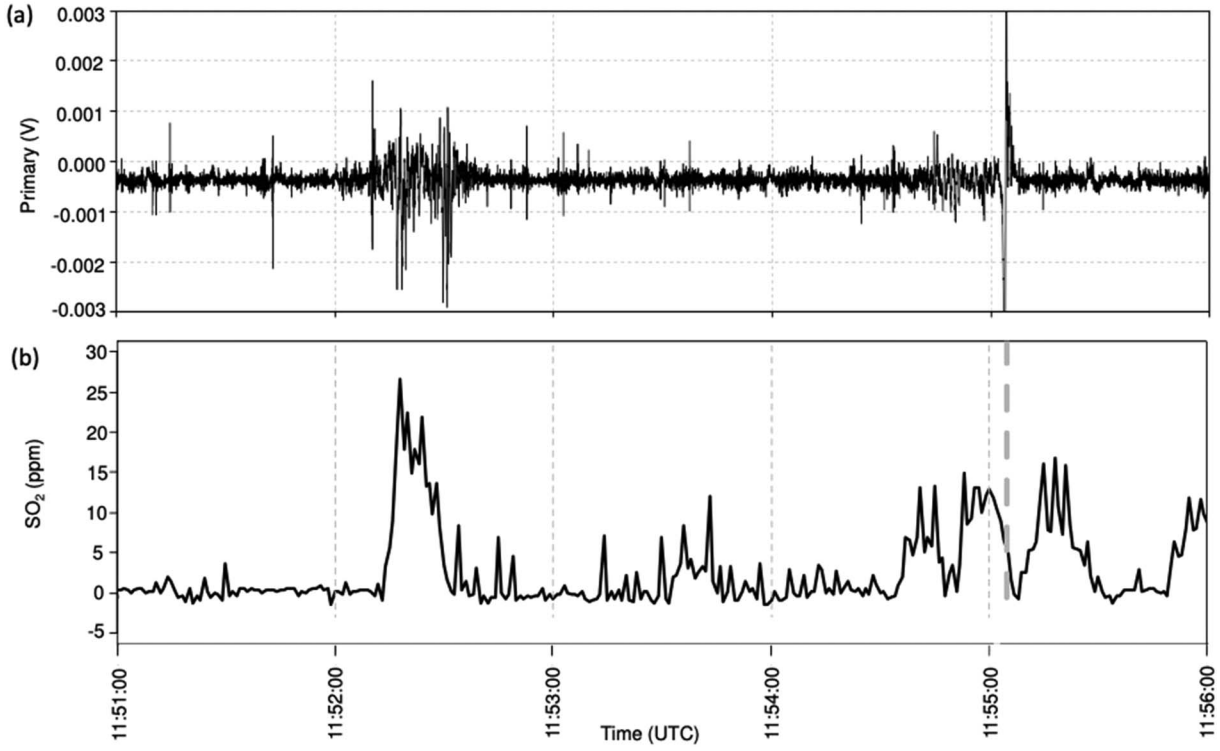

Figure 3. Time series around launch of flight 2 of (a) voltage measured by the primary antenna of the Biral BTD electrostatic sensor (±120 μV) and (b) SO_2 concentration (±3 ppm) measured by the VOLCLAB sensor package. Balloon launch time was at 11:55:15 and shown by the gray vertical dotted line.

data were obtained. Comparing this with the ascent profile in clear air, there is very little charge at the same height, supporting the conclusion that the space charge layer coincided with the volcanic plume.

A second measurement of space charge within the volcanic plume during flight 2 was made by the BTD. This was located ~50 m from the launch site and made alongside the SO_2 sensor from the VOLCLAB package. Figure 3 shows a time series of the voltage measured by the primary antenna from the BTD, for 5 min around the time of launch, alongside the SO_2 concentration at the same location. Several periods of enhanced variability in the BTD current (around 11:52:15 UT and 11:54:30 UT) coincide with enhanced SO_2 concentration when the charged plume surrounded the sensor. The large spike in BTD current at 11:55:15 UT is associated with the balloon launch.

We estimate the space charge density (i.e., the net charge) inside the plume measured by the BTD between 11:52 and 11:53 UT to be ±400 pC/m^3 (see supporting information Text S5 and Figure S5 for details). This is within a factor of 2 of the ±600 pC/m^3 found from the balloon sensor. The charge measurements by different methods therefore show consistency. Smaller charges observed on flight 2 compared with flight 1 may be due to more substantial mixing of the plume due to high wind speeds, increased distance from the vents, or reduced volcanic activity.

4. Discussion

4.1. Source of Plume Charging

These results, derived from independent measurement techniques, demonstrate for the first time that even weak volcanic plumes, produced by passive degassing and with negligible ash concentration, are nevertheless electrically charged. Fractoemission is the only known mechanism capable of producing substantial amounts of charge in a gaseous volcanic plume such as at Stromboli (e.g., Hatekayama and Ushikawa 1952, Lane and Gilbert 1992, James et al., 2000). The absence of detectable ash in the plumes observed at Stromboli suggests that the source of the observed charge was, however, not fractoemission, as the magnitude of the charge is expected to be proportional to both intensity and duration of ash generation and

ejection (James et al., 2000). We do not believe triboelectrification of ash particles plays a significant role here due to the lack of ash.

Previous research has demonstrated that the gaseous constituents of volcanic plumes can readily become charged by exposure to ionization sources (e.g., Ammann et al., 1993). It therefore follows that ash is not required in order to produce charge in a volcanic plume if a source of ionization is present. Volcanic plumes are typically a complicated mixture of high levels of acidic gases such as SO_2 and hydrogen halides accompanying the more abundant water vapor and CO_2. The high temperature of the volcanic environment is likely to lead to many complex interactions between gaseous molecules and liquid particles, which may result in ion generation. In this context chemical reactions and the ion species produced at magmatic temperatures remain largely unexplored and so too the effect of those on gas charging. To address this open question, many further gaseous plume measurements are required comprising simultaneous particle measurements, analysis of chemical species, and in situ ion and bulk space charge measurements. Condensation and sublimation of the gaseous vapor will give rise to liquid droplets, which are known to transfer charge during droplet-droplet collisions (e.g., Khain et al., 2004). Collisions between the substantial number of liquid droplets within the observed plume are highly likely to transfer charge between themselves. The presence of charge up to $\pm$7,000 pC/m^3 in the lowest region (<1 .2km) of the plume in Figure 1c in a region of no optically detectable droplets suggests, however, that droplet-droplet charging is not the dominant charging mechanism and that the charge exists primarily in the form of small ions, at least in this region.

An additional source of ionization within the observed plume could be radioactive decay of radon gas, which is emitted from the active vents and advected upward with the other gaseous constituents of the plume. Radon is emitted through the soil at Stromboli, with surveys reporting the largest radon concentrations at the summit, near active fractures (Cigolini et al., 2005; Cigolini et al., 2009). Atmospheric radon ionizes air through a well-understood decay chain, with most ionization from alpha and gammas from radon progeny. The volumetric ionization rate q is proportional to the radon concentration, for example, Omori et al. (2007) report $q = 4 \cdot \mathrm{cm}^{-3} \cdot \mathrm{s}^{-1}$ for an atmospheric radon concentration of 10 Bq/m^3. Cigolini et al. (2013) show that typical radon concentrations near the summit of Stromboli are 2×10^4 Bq/m^3, increasing to 1.25×10^6 Bq/m^3 during eruptions. In terms of the associated ion production rate q, this corresponds to $q = 8{,}000 \cdot \mathrm{cm}^{-3} \cdot \mathrm{s}^{-1}$ to $2 \times 10^5 \cdot \mathrm{cm}^{-3} \cdot \mathrm{s}^{-1}$. This is 2 to 4 orders of magnitude larger than typical near-surface values of $q \sim 5$–$10 \cdot \mathrm{cm}^{-3} \cdot \mathrm{s}^{-1}$ (Harrison & Tammet, 2008). In a convectively unstable environment such as a volcanic plume, the radon is well mixed (Beck, 1974), giving a steady radon concentration (and ionization rate) with height up to at least 1,000 m. Converting the ionization rates to space charge production rates, neglecting losses, gives 1,000 pC $\cdot$ m$^{-3} \cdot$ s^{-1} (for $q = 8{,}000 \cdot \mathrm{cm}^{-3} \cdot \mathrm{s}^{-1}$), increasing to 32,000 pC $\cdot$ m$^{-3} \cdot$ s^{-1} (for $q = 2 \times 10^5 \cdot \mathrm{cm}^{-3} \cdot \mathrm{s}^{-1}$), neither of which is inconsistent with the lower peak estimate of $\pm$8,000 pC/m^3 measured in the lowest 400 m of flight 1. While the mechanism for the source of the charge observed in the Strombolian plume cannot be known for certain without further detailed measurements, the known existence of radon at Stromboli and the order of magnitude agreement between the ion calculations and charge observations suggest that radon is a likely candidate.

4.2. Implications of Plume Charging

Understanding the processes contributing to volcanic lightning has been the focus of most studies of electrostatic phenomena at volcanoes; however, quantifying the effect of charge on ash and particle transport processes is an overlooked, although increasingly important problem (e.g., van der Does et al., 2018). Charging of droplets and particles can affect the fall speed of particles, changes aggregation rates, and enhances washout of particles, all of which are potentially important in accurately modeling the long-range transport of volcanic plumes. Particle transport models do not currently include the effects of charge, primarily due to a lack of understanding of the charging mechanisms involved as well as a lack of in situ charge measurements with which to develop theories and validate models.

This paper demonstrates that even the smallest and most benign volcanic plumes can be electrically charged and suggests a hitherto unrecognized source of charge generation in volcanic plumes in the form of radon emission. The likely contribution of radon, with a half-life (radon-222) of 3.8 days means that sources of charge may well remain within plumes as they are transported. This may explain previous observations of

charge in plumes distant from their source (Harrison et al, 2010). The ions continually produced by radioactive decay will attach to droplets and particles within the plume, potentially modifying their behavior from that of neutral droplets. Charge is known to affect cloud microphysics through modulating the collision efficiency of colliding droplets and aerosol particles, and affecting the condensation and evaporation of droplets (e.g., Harrison et al., 2015). Since water vapor dominates over other gaseous constituents in a volcanic plume (Allard et al., 2008), the effects of charge on water droplets will dominate over charge effects on droplets of different chemical composition, at least in the short temporal and spatial range. Geochemical reactions in volcanic plumes are complex processes involving the condensation and sublimation of gaseous vapors to form micron to submicron droplets and particles. The sulfur and chlorine-rich magma of Stromboli generates abundant acid gas and metals, which dominate over ash content in buoyant plumes drifted by the wind (Allard et al., 2000). Charge is known to stabilize droplet growth and enable droplet formation at lower critical supersaturations (Harrison et al., 2015); hence, highly charged gaseous volcanic plumes may form droplets more rapidly, or the droplets may persist for longer. Development of a new generation of particle transport models, which incorporate charge effects (including volcanic radon as a source of ionization as suggested here), will therefore lead to more accurate modeling of volcanic plume dispersion.

5. Conclusions

These first in situ measurements of charge inside gaseous plumes above an erupting volcano indicate that volcanic plumes can be substantially charged even if ash is not detected. Charge associated with gas emission adds an important component to the total charge budget of volcanic plumes that has been so far overlooked and needs to be taken into account in future work. The implications of gaseous plume charge include effects on the microphysics regulating the life of ash particles in volcanic plumes (aggregation, segregation and sedimentation) and the overall chemical reactivity of the plume (gas scavenging and longevity of chemical compounds), which are of paramount importance for the environmental impact of volcanic emission. This should be addressed in order to accurately model long-range volcanic plume dispersion, due to the overall societal importance of such events.

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