RESEARCH METHODS VOLCANICA

A gas-tight shock tube apparatus for laboratory volcanic lightning under varying atmospheric conditions

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Abstract

Explosive volcanic eruptions generate electrical discharges, a phenomenon termed volcanic lightning (VL). However volcanism has been active since Earth's origin. Thus, investigating VL under different atmospheric conditions is relevant for studies of early atmospheric chemistry, for studying volcanic lightning on other planets, and potential prebiotic reactions. We developed an experimental setup to investigate VL in varying atmospheres. We present the first experiments of laboratory discharges in particle-laden jets in varying atmospheric conditions. The new experimental setup is a mobile fragmentation bomb erupting into a gas-tight particle collector tank. This setup enables the testing of different atmospheric conditions, changes in the carrier gas of the jet, changes in the pressure within the tank, monitoring of the jet behavior, and sampling of the atmosphere together with the decompressed solid materials. We find that the number and magnitude of near-vent electrical discharge events are similar in CO_2-CO and air atmospheres.

Keywords: Volcanic Lightning; Shock tube apparatus; Early Earth; Experimental discharges.

1 Introduction

On Earth, volcanic lightning (VL) is a phenomenon observed in various volcanically active geological settings and over a wide range of magma composition [McNutt and Williams 2010]. In general, the electrical activity within a volcanic plume accompanying explosive, ash-rich volcanic eruptions is classified as 1) plume lightning, 2) vent discharges, or 3) near-vent lightning [Thomas et al. 2010; Behnke et al. 2013; Aizawa et al. 2016; Cimarelli et al. 2016; Van Eaton et al. 2020; Méndez Harper et al. 2021]. Previous studies have investigated and described the mechanisms that contribute to the electrical activity within the plume. The process of fracto-electrification [James et al. 2000] describes the buildup of charge due to the fragmentation of magma and consecutive fracturing of pyroclasts. The process of triboelectrification [Lacks and Levandovsky 2007] contributes to the buildup of charge by the frictional interaction of different-sized particles. The aforementioned mechanisms also contribute to other types of atmospheric electrification, for example, charge buildup in dust storms [Stow 1969; Esposito et al. 2016]. Thus, investigating fracto-electrification and triboelectrification is relevant for VL and other scenarios where particles interact in turbulent flows.

Lightning or discharges are one potential energy source for nitrogen fixation and are considered a potential prebiotic synthesis mechanism on early Earth [e.g. Miller 1953; Toupance et al. 1975; Chameides and Walker 1981; Stribling and Miller 1987; Bada 2023]. In recent years, the detection of VL, e.g. by the detection of radio-frequency impulses, has become important as a potential monitoring tool for a wide range of explosive volcanic eruptions [e.g. Vossen et al. 2021; Cimarelli and Genareau 2022; Vossen et al. 2022]. However, VL is also a phenomenon that has likely accompanied the Earth since its earliest beginnings and might have played a crucial role in the occurrence of the first organic molecules [Bada 2004; Navarro-González and Segura 2004; Bada 2023], synthesis of ATP components [Chu and Zhang 2023], and phosphorus reduction [e.g. Hess et al. 2021; Çalışkanoğlu et al. 2023]. Electrostatic discharges also represent an energy source to promote chemical reactions on the involved material such as, for example, the amorphization of S and Cl salts [Wang et al. 2020]. As the Earth's atmosphere has changed over time [e.g. Kasting 1993; Catling and Zahnle 2020], further understanding of how VL might have influenced prebiotic synthesis and the influence of the involved geomaterials requires a new experimental setup to investigate VL under varying atmospheric conditions.

One disputed aspect about prebiotic synthesis initiated by lightning activity is that the electrical current might not only produce molecules but also destroy them immediately thereafter as high temperatures are also destructive to most bioorganic compounds [Cleaves 2012]. Different zones of discharges characterize VL: in the near-vent region, the discharges are smaller in length and move smaller amounts of charge than meteorological lightning [Thomas et al. 2010; Aizawa et al. 2016; Méndez Harper et al. 2021; Cimarelli and Genareau 2022] and might represent a potential geological setting for the synthesis of first organic molecules. For electrical charging within the plume close to the vent, the interaction of the ash particles is essential for charge buildup.

The process of electrification is not uncommon in planetary atmospheres. Evidence for lightning exists, for example, on Jupiter, Saturn, Uranus, and Neptune and discharges are considered possible for Mars, Venus, and Titan [Desch et al. 2002; Aplin 2006]. Earth is not the only volcanically active planetary body, for example, Mars was volcanically active in its past [McCauley et al. 1972; Greeley and Spudis 1981; Horvath et al.

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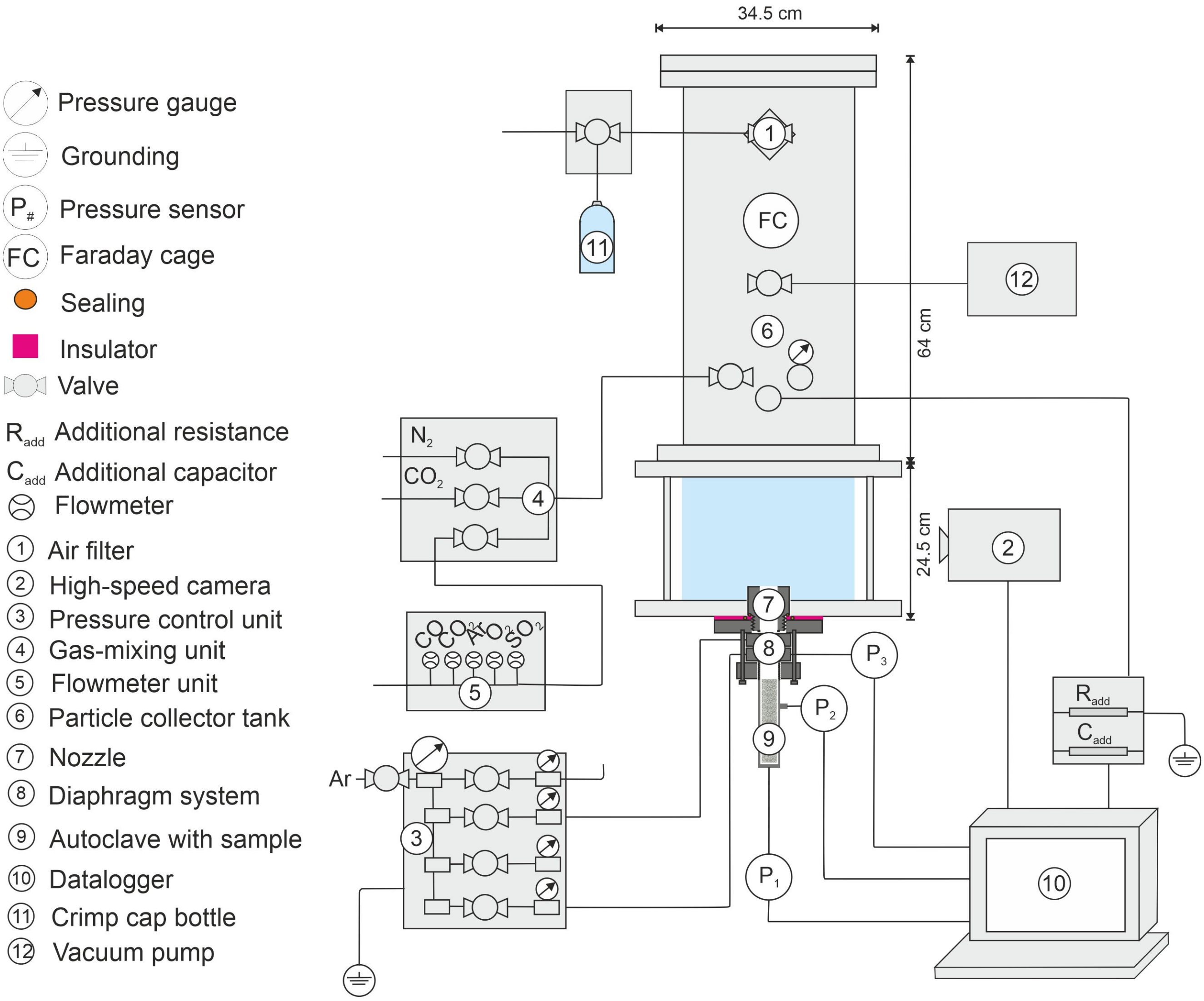

Figure 1: Experimental apparatus. The particle collector tank's upper part is insulated from the ground and the decompression system by a plastic flange (pink). The particle collector tank serves as a Faraday cage, and the datalogger records discharges from the jet to the nozzle. An additional resistor allows computing the electrical current (I) by measuring the voltage (V). The additional resistor decreases the amplitude of the signal and avoids saturation. The lower part of the particle collector tank is a transparent glass inlet, which enables the recording of the jet behavior during decompression with a high-speed camera.

2021] and Venus may be volcanically active at present [Herrick and Hensley 2023]. VL might occur in other volcanically active extra-terrestrial environments, and discharges might be generated by other dust-based charging mechanisms. In all these scenarios, oxygen is an unlikely major constituent of the atmosphere. In contrast, the influence of carbon volatiles on the charging and discharging behavior of particles could be of great importance for all these systems. For example, the Martian atmosphere consists mainly of CO_2 [Owen et al. 1977; Franz et al. 2017; Wordsworth et al. 2021; Sauterey et al. 2022], the main constituents of early Earth's atmosphere were CO_2 and N_2 [Catling and Zahnle 2020], and Venus' atmosphere also consists mainly of CO_2 [Basilevsky and Head 2003]. Other essential parameters that require further investigation to evaluate particle charging behavior on extra-terrestrial bodies are temperature, atmospheric conductivity, and atmospheric pressure. For example, Wurm et al. [2019] investigated the influence of pressure on potential charging mechanisms in Martian analogue dust. In their study, they investigated the effects of pressure on charging of monodisperse, spherical basalt grains and observed that the capability of building up charge is at a minimum in the range of Martian pressure [Wurm et al. 2019]. Additionally, the gas composition of the plume itself can significantly change the composition of the gas in which the discharge might occur and also needs to be addressed in future studies on VL [Navarro-González and Basiuk 1998].

A number of factors influencing VL have already been investigated experimentally as well as in the field, including mass eruption rate [Hargie et al. 2019], plume height [Bennett et al. 2010; Behnke et al. 2013; Vossen et al. 2021], water content and temperature [Stern et al. 2019; Méndez Harper et al. 2020] as well as grain size distribution [Gaudin and Cimarelli

2019; Springsklee et al. 2022b]. The impact of an oxygen-free atmosphere on VL, however, has not yet been investigated experimentally.

2 Methods and materials

To enable the investigation of controlled atmosphere VL studies, we present a new experimental setup ('Tiny Tank') which is a smaller, mobile version of the shock tube apparatus first developed by Alidibirov and Dingwell [1996]. The shock tube apparatus originally developed by Alidibirov and Dingwell [1996] has been used successfully for decades to investigate and characterize the central mechanisms of explosive volcanism. The setup was iteratively improved to enhance the understanding of concepts such as the fragmentation threshold, the speed of the fragmentation front, fragmentation efficiency, resulting grain size distribution, the role of permeability, and the effects of vent geometry [Scheu and Dingwell 2022, and references therein]. The newest versions of the setup were further developed and improved to analyze experimentally generated VL [Cimarelli et al. 2014; Gaudin and Cimarelli 2019; Stern et al. 2019]. The setup enables (Figure 1) the simulation of VL in the lab under varying atmospheric conditions (so far CO_2, CO-CO_2 mixtures, and air), with different transporting gases (to date, Ar and N_2) and varying atmospheric pressures (from 250 mbar to 4 bar). The setup enables us to detect the magnitude and the number of discharges and to monitor and record the particles' eruption behavior with a high-speed camera through a transparent glass section of the tank. The pressure within the investigated atmosphere is recorded by a pressure gauge that further allows the measurement of pressure increase inside the tank/atmosphere caused by the rapid decompression of the particle-laden jet into the particle collector tank (⑥ in Figure 1). The small size of the apparatus allows the transport of the setup to different external locations (e.g. an explosion-safe chamber to test more explosive gas mixtures). The ash can be sampled after the experiment from the bottom of the particle collector tank, and the fine particles are filtered from the atmosphere within the tank by a filtering system in the exhaust air exit. Sampling the gas is also possible by attaching crimp cap bottles to an extra line of the exhaust air exit (⑪ in Figure 1). The particle collector tank is designed as a Faraday cage, which enables us to determine the net electric charge associated with the particle gas mixture along with the number and intensity of discharges. The upper part of the setup, the particle collector tank, became a Faraday cage by insulating it from the grounded autoclave (⑨ in Figure 1). The new setup enables us to measure the discharges and sample the ash and gas-phase pre- and post-experiment.

The autoclave contains the sample and is pressurized by a system of imprinted diaphragms (⑧ in Figure 1). The diaphragms (in combination with the diaphragm holders) form individual chambers in which pressure can be built up to a certain empirically determined burst pressure of the diaphragms. The experiment is triggered by a systematic failure of the uppermost diaphragm; instigated by a pressure increase. The stability of the individual diaphragms is designed in a way that the failure of the uppermost diaphragm causes the rupture of all other diaphragms. To date, we have used argon and nitrogen gas to pressurize the sample chamber. After rupturing the diaphragms, the gas and the gas-particle mixture within the autoclave are ejected through the diaphragm system and the nozzle (⑦ in Figure 1) into the particle collector tank (86 cm in length and 27 cm in diameter). The diameter of the autoclave used in our experiments is 26 mm, and the nozzle diameter is 28 mm. Two diaphragm holders with iron diaphragms were used to pressurize the sample to 10 MPa. The pressure control unit (③ in Figure 1) supplies the autoclave system with pressure and is equipped with three gas lines to supply up to three diaphragm holders with gas. The applied pressure pressurizes the sample and the gas phase within the autoclave.

As the sample is ejected from the autoclave into the particle collector tank, the eruption behavior can be recorded with a high-speed camera (Vision Research Phantom V711) (② in Figure 1) as the lower part of the particle collector tank is composed of transparent glass (17.5 cm in height). A precision digital pressure gauge (model CPG 1500 by WIKA) is attached to the particle collector tank to determine the pressure before and after each experiment. The autoclave can be flushed with the pressurizing gas phase before performing the experiments to remove the initial air. The gas composition within the particle collector tank is determined by the gas mixing unit and a flowmeter unit (Bronkhorst Deutschland Nord GmbH) (⑤ in Figure 1) with flowmeters for CO, CO_2, argon, oxygen, and SO_2. The flowmeter unit enables a highly precise, constant gas flow. The software used to control the flowmeter unit is provided by Bronkhorst (`FLOWDDE`, `FLOWVIEW`, and `FLOWPLOT`). The particle collector tank is conditioned by a vacuum pump before performing experiments. We performed experiments in CO_2 atmosphere in two different modes: (1) by purging the tank three times without evacuation in-between or (2) vacuuming the particle collector tank between the three times of purging before filling the particle collector tank with the desired gas composition for an experiment. The final gas composition before decompression was analyzed for its gas composition. Gas samples were analyzed using a Thermo *Trace 1310* gas chromatograph equipped with an Agilent *CP-Sil 5 CB* column and a thermal conductivity detector (TCD). The temperature program for separation started with a plateau at 50 °C for 2 minutes. The temperature was then raised to 100 °C at a rate of 20 °C min^{-1}. A constant flow of 2 mL min^{-1} of helium was used as a carrier and reference gas.

The gas mixing unit is used to switch between the gas flow of the flowmeter and a simple supply of N_2 and CO_2. Here, the only way to measure the gas inlet is the pressure increase within the particle collector tank, monitored by the pressure gauge. The particle collector tank is sealed gas-tight during the experiments, and an exhaust air outlet can release gas through a Whatman Grade EPM 2000 Air Sampling Filter. A sampling system for crimp cap bottles is attached to the exhaust air outlet. Before sampling, the bottles are evacuated and subsequently filled with helium gas three times. We insulated the particle collector tank from the ground and the diaphragm system in the tradition of previous setups. The autoclave is electrically grounded, whereas the particle collector tank is grounded during the experiments with an additional

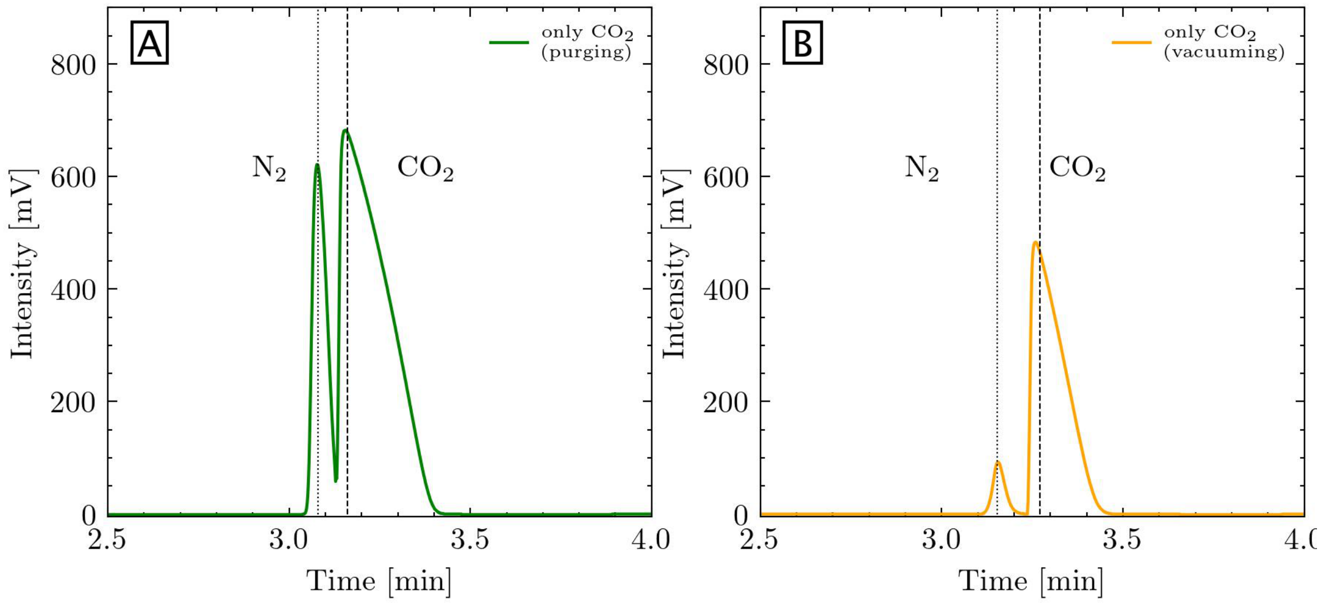

Figure 2: Separation of carbon dioxide and remaining components of air by GC. Gas samples were analyzed using a Thermo Trace 1310 gas chromatograph equipped with an Agilent CP-Sil 5 CB column and a thermal conductivity detector (TCD). Nitrogen and CO_2 show separate peaks, and the amount of air within the sample was determined by calibration. In [A] the gas within the collector tank purged with CO_2 was analyzed, whereas in [B] the gas composition within the particle collector tank conditioned by vacuum was analyzed.

resistor set between the Faraday cage and the ground. The total resistance of this additional resistor (2500 Ω) and the datalogger (Yokogawa WE7000), and the voltage measurement allows us to compute the electrical current. Also, as described by Gaudin and Cimarelli [2019], the additional resistor is used with an extra capacitor to decrease the signal amplitude and avoid saturation in the recording. For the data evaluation, the data processing code developed by Gaudin and Cimarelli [2019] was implemented, and the capacitance for the Faraday cage of this setup was determined as described by Gaudin and Cimarelli [2019].

For the experiments, we used spherical synthetic soda-lime glass beads (GB) as sample materials. The synthetic soda-lime glass beads were purchased from the MHG Strahlanlagen GmbH as fractions of 150–250 µm and 40–70 µm. The grain size fraction of 40–70 µm was ground in a disc mill and the powder obtained was sieved to achieve a grain-size fraction <25 µm. The glass beads have an average matrix density of 2.45 g cm^{-3}.

The grain size distribution (GSD) of the samples was analyzed with a Bettersizer S3 plus (3P Instruments GmbH & Co. KG). The unprocessed glass beads (grain size fraction 150–250 and 40–70 µm) exhibit a very high sphericity whereas the ground particles (grain size fraction <25 µm) are highly angular shards.

Each sample used in the experiments is a mixture of fine (<25 µm or 40–70 µm) and coarse loose sample powder (150–250 µm) that were mixed in varying compositions. The sample was mixed in a sample container and poured into the autoclave using a funnel. Each experiment contains a total mass of ~113 g, such that the sample mixture's weight and volume were approximately constant.

3 Results

In total, we performed 39 experiments in atmospheres of varying amounts of CO_2-CO and air and with two different pressurizing gases: argon and nitrogen.

Two different modes of supplying the particle collector tank with gas were tested: either purging the particle collector tank or purging and vacuuming the tank in between. We tested the discharge behavior for both operational modes in a CO_2 atmosphere to investigate potential differences by changing the atmosphere's composition. The gas composition within the tank was sampled by crimp cap bottles. The gas samples were analyzed using a Thermo Trace 1310 gas chromatograph equipped with an Agilent CP-Sil 5 CB column and a TCD. This enabled us to distinguish between CO_2 and the remaining components of N_2 (Figure 2).

Quantification of the remaining air was obtained by calibration with different partial pressures of air added to CO_2. The analysis of the gas composition within the particle collector tank before decompression shows that purging the tank three times with the desired gas phase is not sufficient to remove all the initial air within the tank.

We measured a remaining volume of ~24 vol.% in the samples for experiments conducted with purging only. Evacuating the chamber before filling the desired gas composition for the experiment is a necessity as this procedure reduced the remaining volume of air to ~7 vol.%.

We performed two experiments (EXP266 and EXP276) with a mixture of the coarse grain size of 150–250 µm and

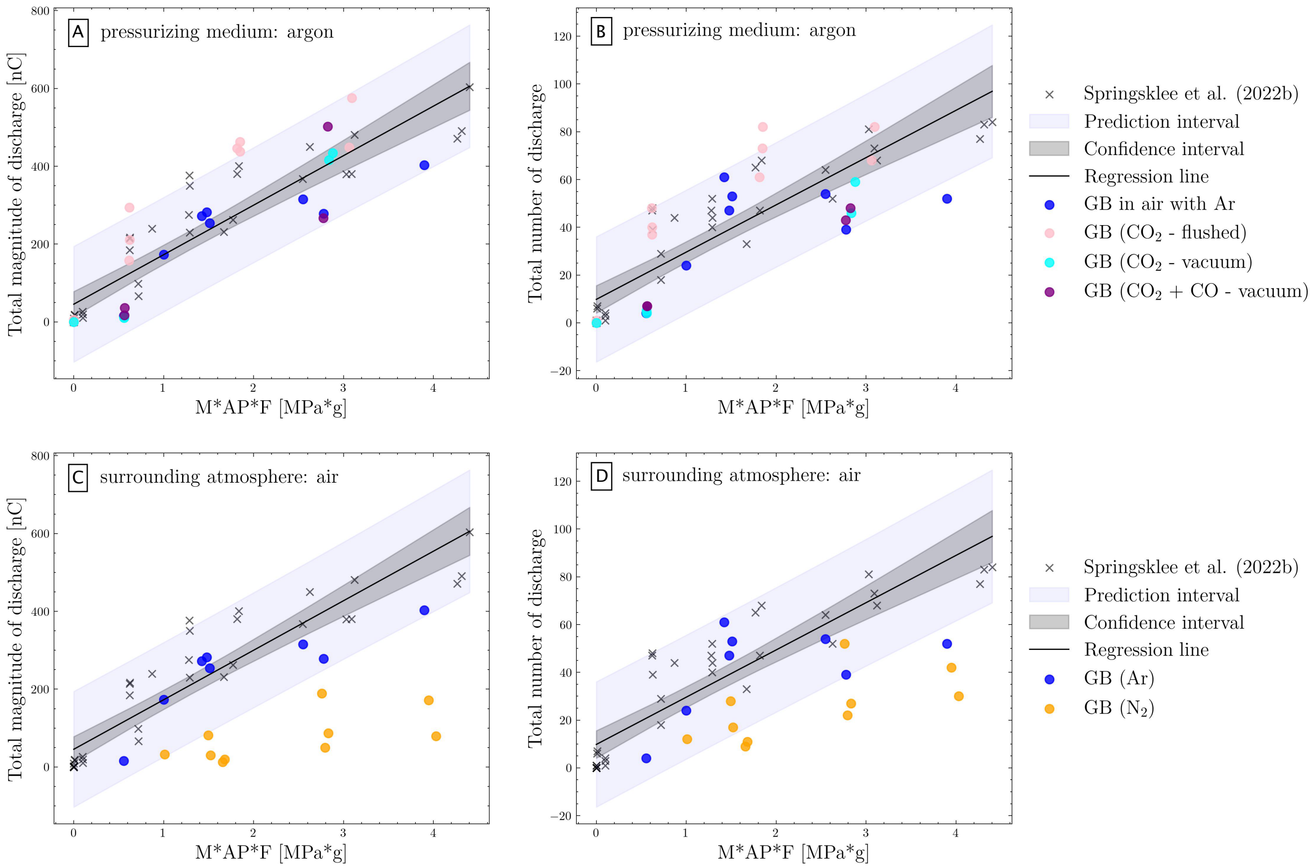

Figure 3: Results of the experimental generation of discharges during decompression with glass beads (GB). The prediction interval, the confidence interval and the regression line are based on the dataset in Springsklee et al. [2022a]. In [A] and [B], the pressurizing gas was always argon, and the surrounding atmosphere was changed. In 2c and 2d, the surrounding atmosphere was air, whereas the pressurizing gas was either argon or nitrogen. The total magnitude of discharge is displayed as a function of mass (*M*) times autoclave pressure (*AP*) times the percentage of very fines (*F*) (< 10 µm) in [A] and [C], whereas the total number of discharges as a function of mass times autoclave pressure times the percentage of very fines (<10 µm) is displayed in [B] and [C]. All experiments of this study represent a mixture of the coarse grain size fraction (150–250 µm) mixed with varying amounts of a fine grain size fraction <25 µm or 40–70 µm.

fine grain size of 40–70 µm in a CO_2 atmosphere. Those two experiments produced no discharges. The same behavior is observable in experiments conducted in air, emphasizing the importance of very fine particles in the initial GSD for generating discharges during decompression [Springsklee et al. 2022a; b]. All other experiments contain a mixture of a coarse grain size fraction and a fine fraction of <25 µm. We conducted all experiments with an applied pressure of ~10 MPa.

We compared our experimental data with a previous dataset [Springsklee et al. 2022a], where the same experiments were performed in air only (Figure 3). As already investigated by Gaudin and Cimarelli [2019], the magnitude and the number of discharges increase with the increasing content of fines within the bimodal grain size distribution.

Comparison of the experiments conducted in atmospheres containing air, CO_2, or CO_2-CO reveal no significant differences in magnitude and number of discharges (Figure 3A and 3B). The measured magnitude and number of discharges fall within the prediction interval for experiments conducted in an air atmosphere. As the experiments containing a CO-bearing atmosphere were not conducted in an explosion-safe laboratory; therefore, the content of CO was held below 5 vol.% for safety reasons.

The discharge behavior of the sample does not change when the surrounding atmosphere composition changes. We changed the pressurizing medium to investigate changes in the transporting gas-phase and exchanged the argon gas as a carrier gas with nitrogen (Figure 3C and 3D). The comparison of the carrier gases, argon and nitrogen, shows that in both cases, the particles are charging during the eruption, demonstrated by an increase in the net charge of the Faraday cage, but discharges are less commonly observed in a jet with nitrogen as a carrier gas. Figure 4 shows the different discharge behavior of an experiment with argon as carrier gas (EXP371) and an experiment with nitrogen as carrier gas (EXP370). For both experiments, following decompression, there is an increase in charge of the Faraday cage, but for the experiment with argon as a carrier gas, the discharges cause a decrease in the net charge of the Faraday cage which does not occur in the jet containing nitrogen as a carrier gas.

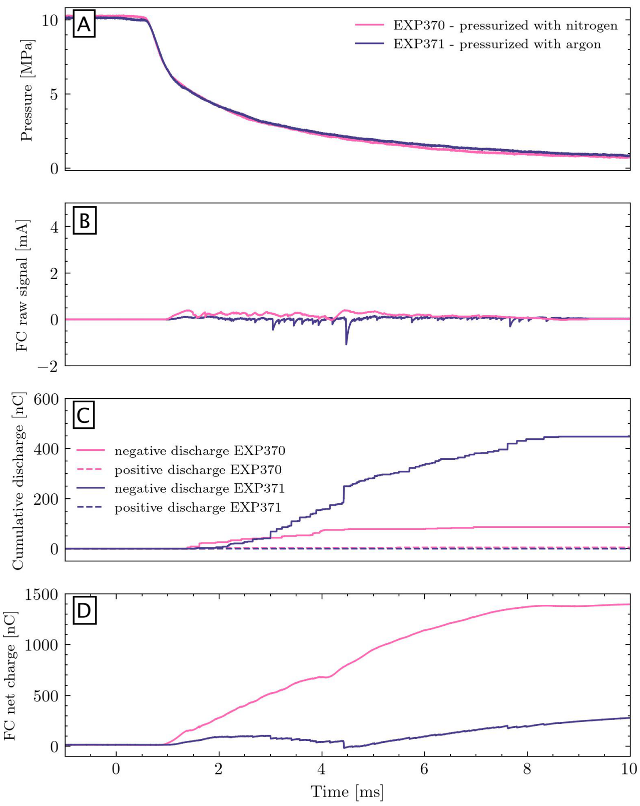

Figure 4: Signals from experiment 370 (carrier gas N_2) and experiment 371 (carrier gas Ar). Both experiments were conducted under similar conditions (10 MPa pressure within the autoclave before decompression) but, depending on the carrier gas composition, a different build-up in net charge is observable.

4 Discussion

The experiments show that near-vent discharges are possible in an oxygen-free atmosphere and the mechanism seems similarly effective at causing discharges as in air. Under the present experimental conditions, the discharge experiments demonstrate that the surrounding atmosphere does not significantly affect the magnitude and number of discharges in experimental near-vent lightning, as it is observable by comparing experiments conducted in air and CO_2. This observation comes with a caveat that there is high variability within the experiments themselves and limitations in our ability to observe all discharge events. In general, the electrical breakdown threshold between two charged objects where a discharge occurs is described by the Paschen law and depends on the pressure of the enveloping gas and the distance between the charged objects. Because the Paschen curve of air and CO_2 differ [e.g. Figure 1 in Helling et al. 2013], we expected to see differences in the magnitude or number of the discharges. Although we can observe optically by a high-speed camera the jet behavior and individual discharges during the experiments, many discharges in our experiments are hidden in the turbulent suspension of the jet. Therefore, a quantitative and qualitative evaluation of the length of individual discharges is impossible at this time.

Méndez Harper and Dufek [2016] investigated the effect of dynamics on the triboelectrification of volcanic ash and concluded that the surface charge density seems to be capped by atmospheric conditions, specifically by the breakdown characteristics of the gas. The charge density of particles fluidized in an argon and nitrogen atmosphere was investigated to test this idea. They observed that the peak of the surface charge density distribution in nitrogen occurs at a value twice as high as in argon. This difference in charge density distribution does not stem from properties of the particles but instead supports the premise that atmospheric properties, not dynamics, dominate the saturation charge in a granular flow [Méndez Harper and Dufek 2016]. Therefore, the charging of the particles should vary with the properties of the surrounding gas. Compared to their experimental setup, we have to consider the interaction of the expanding jet (particles and argon/nitrogen gas) with a surrounding atmosphere. The jet can be described as a gas-particle mixture where the distribution of the particles within the jet is governed by the tendency of particles to become coupled to the transporting gas phase. The frictional interaction of the small grains is based on turbulent collisions with the coarse grain size fraction and is most likely the charging mechanism causing the charging of the particles within our experimental setup. The jet behaves turbulently as eddies are observable in the high-speed images (recordings of the experiments conducted in air with argon are available in the supplementary material in [Springsklee et al. 2022b]. This behavior is more pronounced in the upper region of the jet than close to the nozzle. The results of the experiments demonstrate that gas entrainment of the atmospheric gas phase is less efficient closer to the nozzle as in the upper parts of the jet. The results also illustrate the importance of the transporting gas phase for near-vent discharges.

5 Conclusions

The new experimental setup presented in this study enables further advances in rapid decompression experiments, allowing the investigation of electrified dusty flows under controlled atmospheres conditions. The ability to control both gas carrier phase and ambient atmosphere further extends the range of gases investigated, which is relevant for prebiotic synthesis experiments, including those relevant for volcanic plumes. Our results emphasize the importance of the carrier gas phase for the case of prebiotic synthesis triggered by near-vent volcanic lightning, with relevance for reducing gases in the volcanic jets. Additionally, our setup opens new opportunities for the study of electrical discharges in other planetary atmospheres. Experiments conducted with nitrogen as a carrier gas compared to those with argon are less efficient in generating discharges following the Paschen law for those two gases. Therefore, the influence and interaction of the surrounding atmosphere and the volatile content of a plume for several eruption types should be investigated to provide deeper insights into volcanic eruptions and their accompanying VL. The influence of atmospheric pressure on such processes also requires further investigation.

Author Contributions

B.S. and C.S. designed the study and the experimental setup. B.S. and M.M. provided supervision. D.G., B.S. and C.C. provided the methodology. D.G. and C.C. designed the processing code to analyze the raw signal. C.S., B.S. and M.M. performed the experiments, C.S. analyzed, and visualized the results, prepared the samples, sampled the solid sample and gas mixtures, and wrote the original draft. C.SE. and O.T. analyzed the gas samples. All authors contributed to the discussion and revised the manuscript. B.S. and D.B.D. acquired funding.

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Data Availability

Experimental data are online at the GFZ Data Services [Springsklee et al. 2023].

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