



Spatial and Temporal Variations in SO₂ and PM_{2.5} Levels Around Kīlauea Volcano, Hawai'i During 2007–2018

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Whitty RCW, Ilyinskaya E, Mason E, Wieser PE, Liu EJ, Schmidt A, Roberts T, Pfeffer MA, Brooks B, Mather TA, Edmonds M, Elias T, Schneider DJ, Oppenheimer C, Dybwad A, Nadeau PA and Kern C (2020) Spatial and Temporal Variations in SO₂ and PM_{2.5} Levels Around Kīlauea Volcano, Hawai'i During 2007–2018. Front. Earth Sci. 8:36. doi: 10.3389/feart.2020.00036 Among the hazards posed by volcanoes are the emissions of gases and particles that can affect air quality and damage agriculture and infrastructure. A recent intense episode of volcanic degassing associated with severe impacts on air quality accompanied the 2018 lower East Rift Zone (LERZ) eruption of Kilauea volcano, Hawai'i. This resulted in a major increase in gas emission rates with respect to usual emission values for this volcano, along with a shift in the source of the dominant plume to a populated area on the lower flank of the volcano. This led to reduced air quality in downwind communities. We analyse open-access data from the permanent air quality monitoring networks operated by the Hawai'i Department of Health (HDOH) and National Park Service (NPS), and report on measurements of atmospheric sulfur dioxide (SO₂) between 2007 and 2018 and PM_{2.5} (aerosol particulate matter with diameter <2.5 µm) between 2010 and 2018. Additional air quality data were collected through a community-operated network of low-cost PM_{2.5} sensors during the 2018 LERZ eruption. From 2007 to 2018 the two most significant escalations in Kilauea's volcanic emissions were: the summit eruption that began in 2008 (Kīlauea emissions averaged 5-6 kt/day SO2 from 2008 until summit activity decreased in May 2018) and the LERZ eruption in 2018 when SO₂ emission rates reached a monthly average of 200 kt/day during June. In this paper we focus on characterizing the airborne pollutants arising from the 2018 LERZ eruption and the spatial distribution and severity of volcanic air pollution events across the Island of Hawai'i. The LERZ eruption caused the most frequent and severe exceedances of the Environmental Protection Agency (EPA) PM_{2.5} air quality threshold (35 µg/m³ as a daily average) in Hawai'i in the period 2010-2018. In Kona, for example, the maximum 24-h-mean mass concentration of $PM_{2.5}$ was recorded as 59 μ g/m³ on the twenty-ninth of May 2018, which was one of eight recorded exceedances of the EPA air quality threshold during the 2018 LERZ eruption, where there had been no exceedances in the previous 8 years as measured by the

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HDOH and NPS networks. SO₂ air pollution during the LERZ eruption was most severe in communities in the south and west of the island, as measured by selected HDOH and NPS stations in this study, with a maximum 24-h-mean mass concentration of 728 μ g/m³ recorded in Ocean View (100 km west of the LERZ emission source) in May 2018. Data from the low-cost sensor network correlated well with data from the HDOH PM_{2.5} instruments, confirming that these low-cost sensors provide a robust means to augment reference-grade instrument networks.

Keywords: Kīlauea, volcano, PM_{2.5}, SO₂, emissions, air quality, Hawai'i

1. INTRODUCTION

Volcanic clouds are complex, evolving mixtures of volcanic and atmospheric gases, primary and secondary aerosol particles, ash and dust (Oppenheimer and McGonigle, 2004; Pfeffer et al., 2006b; von Glasow et al., 2009; Oppenheimer et al., 2010; Langmann, 2014). As well as the potential for global climatic consequences from explosive or large-scale volcanic emissions, low altitude volcanic clouds can have important impacts on air quality, human and animal health, and the environment on the local to regional scale (Hansell and Oppenheimer, 2004; Barsotti et al., 2010; Mather, 2015; Schmidt et al., 2015; Andronico and Del Carlo, 2016; Tam et al., 2016; Ilyinskaya et al., 2017).

Gaseous sulfur dioxide (SO_2) is usually highly concentrated in volcanic emissions compared to the background atmosphere and is often the focus of gas emission monitoring due to the relative ease of its measurement and its important environmental and air quality consequences (Cadle et al., 1971; Lambert et al., 1988; Loughlin et al., 2012; Schmidt et al., 2015). Population sub-groups including children, asthmatics and cardiac- or respiratory-compromised individuals are particularly vulnerable to exposure to SO₂ (ATSDR, 1998; CRI, 2004). For example, exposure to mass concentrations of $1,310 \,\mu\text{g/m}^3$ SO₂ for 3 min can induce respiratory attacks in asthmatic individuals (Balmes et al., 1987; ATSDR, 1998). In 2010 the U.S. Environmental Protection Agency (EPA) set the National Ambient Air Quality Standard (NAAQS) for SO2 mass concentration exposure limits at 195 µg/m³ as an hourly average (EPA, 2010). Persistent volcanic SO₂ emissions on the Island of Hawai'i led the state of Hawai'i being designated as unclassifiable for the EPA 2010 NAAQS, and as such Hawai'i uses the pre-2010 EPA SO₂ exposure limit of $366 \,\mu\text{g/m}^3$ as 24-h average (EPA, 2013). The European Commission (EC) air quality standards recommend a SO_2 mass concentration threshold of 350 µg/m³ for a 3-h average, and $125 \,\mu\text{g/m}^3$ as a daily average (EC, 2018).

Particulate matter (PM) in volcanic clouds is also significant in the context of environment and health. The chemical composition of volcanogenic PM_{2.5} (PM with diameter <2.5 μ m) is highly heterogeneous. Typical chemical species include sulfates (primary emissions or formed via oxidation of sulfur gases) (Cadle et al., 1971; Stockwell and Calvert, 1983; Allen et al., 2002; Mather et al., 2003; Langmann, 2014) and halides, with an array of metals and metalloids including environmentally-harmful species such as lead and cadmium (Longo, 2013; Langmann, 2014). PM_{2.5} is a well-established indicator for air quality, since it commonly includes particulates derived from transport and industrial sources, fine wind-blown mineral dust, ambient matter, and volcanic material (Lim et al., 2012; Tam et al., 2016; Holgate, 2017; Butwin et al., 2019). It has been estimated that the health burden due to exposure to ambient PM_{2.5} globally amounts to more than three million premature deaths each year (Lim et al., 2012), and is especially linked to increases in death from cardiovascular and respiratory diseases in vulnerable individuals (Holgate, 2017). EPA NAAQS thresholds recommend a PM_{2.5} mass concentration exposure limit of 35 µg/m³ as a daily average (EPA, 2013). This is higher than the 24-h mean exposure guideline of 25 µg/m³ established by the World Health Organization (WHO, 2005).

2. KĪLAUEA ERUPTIVE ACTIVITY, 2007–2018

Kīlauea volcano on the Island of Hawai'i consists of a summit caldera at 1,200 m a.s.l. and rift zones to the south-west and east. From 1983 until 2008, activity at Kīlauea was concentrated on the middle East Rift Zone (ERZ), primarily near the Pu'u 'Õ'ō vent (Elias and Sutton, 2007; Poland et al., 2008) (**Figure 1A**). Between 2002 and 2006, average SO₂ flux from the ERZ was 1.7 ± 0.7 kt/day, while emissions from the summit were low at 0.1 kt/day (Elias and Sutton, 2007). From November 2007 to March 2008, SO₂ emissions at the summit increased to levels 10 times the long-term background (Wooten et al., 2009) (**Figure 2A**). On the twelfth of March 2008, a new vent opened within the Halema'uma'u summit crater, leading to sporadic explosive eruptions and increased degassing of SO₂.

Kīlauea's SO₂ emissions peaked in the summer of 2008, when a total emission rate of up to 20 kt/day was measured by satellite sensors (Beirle et al., 2014). At this time, emissions from both the ERZ and the summit were significant, with the two sources contributing variable amounts to the total degassing rate (Elias and Sutton, 2012). For the period 2009–2017, the dynamic activity at Kīlauea was reflected in variable emissions, with a longterm average of 5–6 kt/day based on satellite and ground-based measurements (Eguchi et al., 2011; Elias and Sutton, 2012; Carn et al., 2016; Elias et al., 2018) (**Figure 2A**). Lava was first observed in the Halema'uma'u summit crater in September 2008, with a permanent lava lake visible from February 2010 until May 2018 (Patrick et al., 2013; Neal et al., 2019).

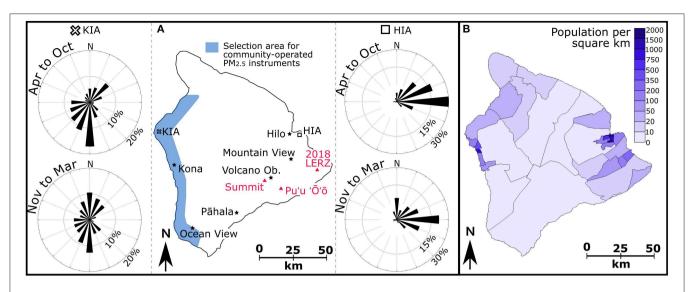


FIGURE 1 | Island of Hawai'i. (A) Ground-based sampling networks; HDOH and NPS stations indicated with black stars. Kilauea summit, Pu'u 'Õ'ō vent and 2018 lower East Rift Zone eruption site indicated with red triangles. Selection area of community-operated PM_{2.5} instruments indicated with blue highlight. Wind directions at Kona International Airport (KIA, empty black cross) and Hilo International Airport (HIA, empty black square) displayed as seasonal wind roses for period 2007–2018 (NOAA, 2019). Note "Volcano Ob." stands for Volcano Observatory. (B) Population density per square kilometer on the Island of Hawai'i, based on 2010 Census data. Highest population density is located in the east-coast city of Hilo (population ~45,700) and the west-coast city of Kailua-Kona (population ~28,500) (Hawai'i Department of Business Economic Development and Tourism, 2011).

The 2018 Kilauea eruption in the lower East Rift Zone (LERZ) began following the collapse of the Pu'u 'O'o vent on the thirtieth of April (Neal et al. 2019; HVO, 2018). Twenty-four fissures opened over a distance of 6.8 km in the vicinity of Leilani Estates (Figure 1A). During the first week of the LERZ eruption, spattering activity at individual fissures was typically short-lived (minutes to hours in duration) and lava was viscous with spatter deposition within tens of meters of individual fissures. On the eighteenth of May, the eruptive style evolved to less viscous lava and resulted in fast-moving lava flows which reached the ocean 2 days later (HVO, 2018). By the end of May 2018, activity had become focused at Fissure 8, and this remained the dominant fissure for the remainder of the LERZ eruption (Neal et al., 2019). Lava fountains from Fissure 8 reached heights of 80 m, and lava effusion rates ranged from 50 to 200 m³/s (Neal et al., 2019). Lava from Fissure 8 flowed in a semi-stable channel to the ocean and eventually covered an area of land 35.5 km² in size (Neal et al. 2019; HVO, 2018). This eruption was the largest along Kilauea's LERZ in the last two centuries and had farreaching impacts around the Island of Hawai'i. With the collapse of the Pu'u 'O'o vent and draining of Kilauea's summit magma reservoir, the dominant source of volcanic SO₂ became the LERZ eruptive vents. SO₂ emissions reached an average of 200 kt/day in June 2018 (Kern et al. 2019), severely impacting island-wide air quality. The eruption declined rapidly at the end of July and lava effusion ceased on the fourth of August 2018 (Neal et al., 2019).

2.1. Downwind Processes and Impacts

Since initiation of intermittent fountaining activity at Kīlauea in 1983, SO_2 emissions have been a health concern among downwind communities on the Island of Hawai'i. As emissions

from Kilauea are dispersed downwind, communities are exposed to volcanic smog, locally known as vog, predominantly composed of SO₂ and fine particles of sulfuric acid aerosol (Longo, 2009; Longo et al., 2010; Halliday et al., 2015; Tam et al., 2016; Elias and Sutton, 2017). Prevailing trade winds from the north-east, particularly during the period from April to October, carry Kilauea's emissions over the communities to the south and west (Longo et al., 2005, 2008; Michaud et al., 2007; Tam et al., 2016; Elias and Sutton, 2017) (Figure 1A). The Island of Hawai'i has relatively low population density in the south (Figure 1B), with ~4,400 residents in Ocean View and ~1,300 residents in Pāhala. Trade winds from the north-east are influenced by the high topography of Mauna Loa and Mauna Kea, generating more localized air movement in the lee of the island on the west coast (Michaud et al., 2007) (Figure 1A). This wind shadow allows a potentially longer residence time for air pollutants (volcanogenic or otherwise) along the densely-populated western coastline (Figure 1B). During the winter months (November to March), the trade winds weaken and southerly and westerly winds may distribute vog toward the densely-populated eastern coastline of the island (Wyrtki and Meyers, 1976; Mannino et al., 1996; Michaud et al., 2004) (Figure 1B).

Numerous studies have investigated the impact of Kīlauea's SO_2 emissions on the health of island residents, even at the relatively low levels of degassing prior to the emergence of the lava lake in 2008. Mannino et al. (1996) reviewed the frequency of visits to emergency departments and hospitalizations for respiratory issues during periods of continuous and discontinuous SO_2 emissions throughout the 1980s. Communities on the western side of the island frequently exposed to vog were found to have higher rates of

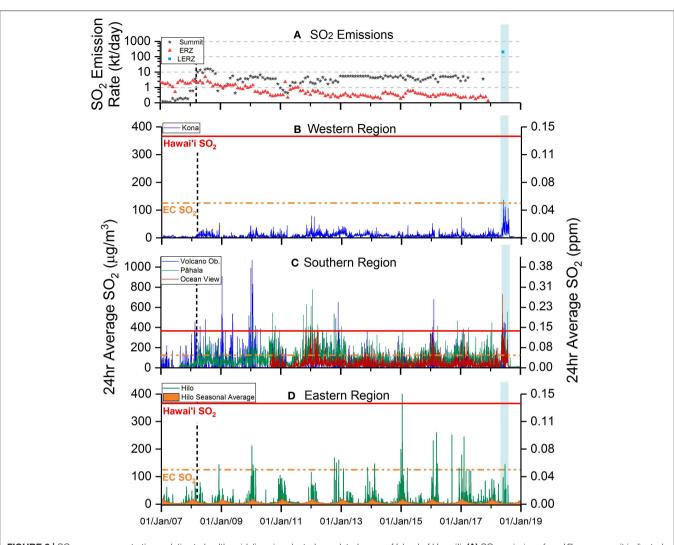


FIGURE 2 | SO₂ mass concentrations relative to health guidelines in selected populated areas of Island of Hawai'i. (**A**) SO₂ emissions from Kilauea summit indicated with black stars; from ERZ indicated with red triangles; from 2018 LERZ eruption indicated with blue square. Note logarithmic scale. (**B–D**) SO₂ air quality (24-h mean) in communities from 2007 to the end of 2018. Solid red line: Hawai'i Standard air quality threshold (24-h mean of 366 μ g/m³); dashed orange line: European Commission (EC) air quality threshold (24-h mean of 125 μ g/m³). Dashed black line: 2008 initiation of summit activity; blue highlight: 2018 LERZ eruption duration. (**A**) SO₂ emissions data sourced from Elias and Sutton (2012), Beirle et al. (2014), Elias et al. (2018), and Elias et al. (in preparation). Summit emissions for 2008–2012 are from Beirle et al. (2014). LERZ data-point represents the average SO₂ emission from the LERZ eruption for June 2018, taken from Kern et al. (2019). (**B**) Note that "Volcano Ob." stands for Volcano Observatory; (**D**) seasonal average for Hilo between 2007 and 2017 indicated by orange bars.

hospitalizations for chronic obstructive pulmonary disease than the east-coast city of Hilo, which is rarely exposed to vog. Periods of weakened north-easterly trade winds coincided with a 15 % increase in emergency department visits for asthma in Hilo (Mannino et al., 1996). In 2004, the health of Hawai'i residents in vog-exposed and -unexposed communities was surveyed (Longo et al., 2008; Longo, 2009). Those in exposed communities were found to have a significantly increased prevalence of cough, phlegm, sinus congestion, rhinorrhoea, wheezing, eye irritation, and bronchitis than those in unexposed communities. Following the increase in SO₂ flux from Kīlauea's summit in 2008, Longo (2013) reassessed the vog-related health impacts on the residents of the Island of Hawai'i. The magnitude of cardio-respiratory issues in vog-exposed communities was found to have increased as compared to 2004 (Longo et al., 2008), with the risk factor of acute cardiac events in persons aged >50 years increased by 12 % (Longo, 2013). A study by Tam et al. (2016) investigated the effects of vog on the respiratory health of school children across Hawai'i, finding that chronic exposure to vog was associated with increased prevalence of cough and potential decrease in lung function, but not with the prevalence of asthma or bronchitis. The unprecedented emission rates of the 2018 LERZ eruption has presented a continued motivation to further characterize the severity and distribution of volcanic air pollution during elevated volcanic activity.

The 2018 Kīlauea LERZ eruption provided a unique opportunity to study the impacts arising from a large low-altitude cloud rich in SO_2 in a populated and well-instrumented part

of the world. Here we used open-access data from a network of reference-grade instruments in populated areas around the Island of Hawai'i to determine the severity of SO_2 and $PM_{2.5}$ impact on air quality from the LERZ eruption. We compare air quality during the LERZ eruption to that from a lower emission period, defined to be January 2007 to December 2017 for SO_2 and January 2010 to December 2017 for $PM_{2.5}$. We examine a network of community-operated $PM_{2.5}$ instruments and compare their measurements to those from reference-grade instruments. From these data, we demonstrate that SO_2 and $PM_{2.5}$ mass concentrations during the 2018 LERZ eruption in selected communities around the island were of a higher magnitude than during volcanic activity from Kīlauea during 2007 to 2017.

3. DATA AND METHODS

3.1. Continuous SO₂ and PM_{2.5} Air Quality Monitoring

Hawai'i Department of Health (HDOH) ambient air quality stations continuously monitor SO_2 and $PM_{2.5}$ mass concentrations around the Island of Hawai'i (**Figure 1A**). Automated SO_2 monitoring stations have been operational since 1997 in Hilo, 2005 in Kona, 2007 in Pāhala and 2010 in Ocean View. A National Park Service (NPS) ambient air quality station monitors SO_2 inside of Hawai'i Volcanoes National Park at the Volcano Observatory. $PM_{2.5}$ has been autonomously monitored since 2005 in Mountain View and Kona, 2008 in Hilo and Pāhala and since 2010 in Ocean View.

SO₂ is measured by a pulsed fluorescence spectroscopy analyzer (model 43i manufactured by Thermo Scientific) that is designated by the EPA for measurements in the range of 0-1,000 ppb, with a lower detectable SO₂ limit of 0.5 ppb and a precision of 1 ppb (Thermo Scientific 2010; EPA, 2016). FEM-designated instruments (Forum for Environmental Measurements), such as this SO₂ analyzer, promote consistency in measurements and laboratory conditions ensuring that the instruments are of reference-grade quality (EPA, 2016). Following EPA regulations, the analyzers undergo in-situ calibration checks weekly, with a multi-point calibration run every 6 months. PM_{2.5} mass concentrations are measured by a Beta Attenuation Monitor (BAM) with a 60-min sampling rate. The BAM instrument (model BAM-1020, manufactured by Met One Instruments) is FEM-designated for measurements of particles in the size range of $0-1,000 \,\mu\text{m}$ (with PM_{2.5} being a small subset of the measured particle size range), with a resolution of 1 µm particle diameter and a lower detection limit of $4 \mu g/m^3$ (MetOne 2008; EPA, 2016). The BAM instruments undergo calibration and auditing every 6 months. Permanent HDOH ambient air quality stations are kept in air-conditioned enclosures to maintain long-term stability. Data from the air quality stations are streamed in near-real time to the HDOH website, which is open-access and publicly-available (HDOH, 2019).

Data from the HDOH and NPS station networks used in this study have been categorized into regions for the purpose of data

analysis. The western region includes Kona HDOH station on the west coast of the Island of Hawai'i. The southern region includes Volcano Observatory NPS station and Pāhala and Ocean View HDOH stations. The eastern region includes the Hilo and Mountain View HDOH stations. We compare the HDOH and NPS SO₂ timeseries data against the European Commission 24-h air quality threshold ($125 \mu g/m^3$) and the Hawai'i 24-h ambient air quality standard ($366 \mu g/m^3$). The HDOH PM_{2.5} timeseries data is compared to the World Health Organization 24-h exposure limit ($25 \mu g/m^3$) and the Environmental Protection Agency NAAQS 24-h limit ($35 \mu g/m^3$).

3.2. Community-Operated PM_{2.5} Instruments

PurpleAir (Utah, USA) instruments are low-cost (approximately \$250 per unit) particulate sensors that are purchased and operated by individuals and provide open access data online (PurpleAir, 2019). PurpleAir instruments contain Plantower PMS5003 nephelometer sensors, which use a small fan to draw air through a laser-induced light, and a photo-diode detector converts 90°-scattered light into a voltage pulse (Kelly et al., 2017). PMS5003 sensors have a 10 s response time and detect particles between 0.3 and 10 µm in diameter (Kelly et al., 2017; Sayahi et al., 2019). The maximum consistency error of the sensors is stated by the manufacturer to be $\pm 10 \,\mu\text{g/m}^3$ between 0 and $100 \,\mu\text{g/m}^3$ (Plantower, 2016). The instruments are factory calibrated prior to sale (PurpleAir, 2019). PM mass concentration measurements are calculated using an atmospheric calibration factor, details of which are not provided by the manufacturer (Kelly et al., 2017; Zheng et al., 2018; Sayahi et al., 2019). PurpleAir instruments contain two Plantower PMS5003 sensors mounted in one housing, allowing self-consistency checks to alert when significant differences are reported between the internal sensors. An ESP8266 wireless chip is included in PurpleAir instruments to upload data via WiFi to an online cloud database, which is open-access (Sayahi et al., 2019).

Prior to the 2018 LERZ eruption, six community-operated PurpleAir instruments were located on the Island of Hawai'i, three of which were in the Kona area in the western region. Installation dates ranged from August 2017 to February 2018. Following the onset of the LERZ eruption and island-wide increase in atmospheric pollutants, the number of PurpleAir instruments increased, with a further twenty instruments installed across the western region of the island over the course of May to July 2018.

We carried out fieldwork during the 2018 LERZ eruption and co-located PurpleAir instruments with HDOH ambient air quality stations at Kona and Ocean View (**Figure 1A**). Two PurpleAir instruments were installed at the Kona station, one from the fifth of June to September 2018 and a second from the nineteenth of July to the third of August, and one PurpleAir instrument was installed at the Ocean View station from the nineteenth of July to the third of August 2018. The PurpleAir instruments were installed close to the inlet for the BAM instruments, on the roofs of the air quality shelters and away from obstructions. Other than the coordinates of the instrument, no environmental information is provided in association with the PurpleAir instruments. For this reason, measurement uncertainty arising from factors such as installation in proximity to potential contamination sources in community-operated PurpleAir instruments should be considered significantly higher than those placed in optimal conditions near HDOH stations. Community-operated PurpleAir instruments were selected for analysis along the region highlighted in blue in **Figure 1A**.

4. RESULTS

4.1. SO₂ Mass Concentrations 2007–2018 4.1.1. Western Region: Kona

During 2007–2017, the west-coast city of Kailua-Kona was commonly exposed to low mass concentrations of SO₂, with a maximum recorded 24-h-mean mass concentration of 79 µg/m³ recorded at the Kona HDOH station (**Table 1**). SO₂ air pollution in Kona increased with the onset of summit activity in 2008 (2008–2017 average mass concentrations were 7.7 µg/m³ compared to $3 \mu g/m^3$ in 2007) (**Figure 2B**). During 2007–2017 there were no 24-h periods where SO₂ mean mass concentrations at the HDOH Kona station exceeded Hawai'i or EC recommended thresholds (**Figure 2B**).

Kona experienced elevated SO₂ mass concentrations during the 2018 LERZ eruption, with a peak 24-h-mean mass concentration of 136 μ g/m³ measured at the Kona station. During the 2018 LERZ, SO₂ mass concentrations in Kona did not exceed the Hawai'i SO₂ threshold of 366 μ g/m³ (**Figure 2B**), but did exceed the 125 μ g/m³ EC threshold on one occasion (**Table 1**).

4.1.2. Southern Region: Volcano Observatory, Pāhala and Ocean View

Following initiation of Kilauea's summit activity in 2008, the HDOH-operated Pāhala station and the NPS-operated Volcano Observatory station routinely recorded high concentrations of SO_2 (Figure 2C), with SO_2 mass concentrations exceeding the Hawai'i 366 µg/m³ 24-h-mean threshold 0.7 % of the time at both Volcano Observatory (twenty-eight exceedance events) and Pāhala (thirty exceedance events) (Table 1). The maximum 24h-mean mass concentration recorded by the NPS station at Volcano Observatory during 2007-2017 was 1,068 µg/m³, and by the HDOH station in Pāhala was 776 μ g/m³. The Ocean View HDOH station is located farther to the south-west than Pāhala and Volcano Observatory, at a greater distance from Kilauea's summit and the ERZ (Figure 1A). During the period 2010–2017, SO₂ mass concentrations recorded at Ocean View exceeded the Hawai'i 24-h-mean threshold 0.1 % of the time (three exceedance events). The maximum 24-h-mean mass concentration recorded at Ocean View was 403 µg/m³, significantly lower than measured at Volcano Observatory and Pāhala (Table 1).

In comparison, during the 3-months of the 2018 LERZ eruption, SO_2 mass concentrations exceeded the Hawai'i 366 µg/m³ threshold 2.1 % of the time at Volcano Observatory (two exceedance events), 5.3 % of the time at Pāhala (five exceedance events) and 4.2 % at Ocean View (four exceedance events). Maximum 24-h-mean mass concentrations at Volcano

Observatory and Pāhala were lower than those measured during 2008–2017 (450 and 555 μ g/m³, respectively), but the relative frequency of exceedance events increased (**Table 1**). During the 2018 LERZ eruption, the Ocean View station recorded a peak 24-h-mean mass concentration of 728 μ g/m³, almost double the previous peak measurement of 403 μ g/m³ recorded at that station in January 2016.

4.1.3. Eastern Region: Hilo

During the period 2007–2017, SO₂ mass concentrations in Hilo followed a distinct seasonality (Figure 2D). Peak SO₂ mass concentrations were commonly observed in Hilo in November to March (average monthly concentration of $11 \,\mu g/m^3$) with low mass concentrations in the intervening months of April to October (average monthly mass concentration of $3.6 \,\mu\text{g/m}^3$, as calculated from 2007 to 2017) (Figure 2D). Exceedances of the EC 24-h-mean threshold $(125 \,\mu g/m^3)$ rarely occurred outside this peak season. In the period 2007-2017 there were twenty-one exceedance events during November to March, compared with just three between April to October. The seasonal variations in SO₂ mass concentrations observed in Hilo can be explained by the strong prevalence of northeasterly trade winds during April to October (Wyrtki and Meyers, 1976) (Figure 1A). During these months, emissions from Kilauea's summit and the ERZ were dispersed predominantly to the south-west of the Island. The trade winds weaken between November to March, allowing SO2 to be dispersed to the east of the island (Mannino et al., 1996; Michaud et al., 2004; Elias and Sutton, 2017).

During the third of May to fourth of August LERZ eruption, the HDOH station in Hilo recorded a maximum 24-h-mean mass concentration of $144 \,\mu\text{g/m}^3$, which was recorded on the twenty-first of June (**Table 1**), and was the only exceedance of the EC 24-h-mean threshold during the 3-month eruption. The SO₂ mass concentrations measured during the LERZ eruption were lower than the average measurements during the 2007–2017 period. SO₂ mass concentrations in Hilo are usually low during the months when the LERZ eruption occurred. Nevertheless, during the 2018 LERZ eruption, SO₂ mass concentrations in Hilo rose significantly above the average for the season (average 24-h-mean SO₂ mass concentration during 2018 LERZ eruption was 6.9 µg/m³, in comparison to the usual seasonal average of 3.6 µg/m³).

4.2. PM_{2.5} Mass Concentrations 2010–2018 4.2.1. Western Region: Kona

In the period 2010–2017, $PM_{2.5}$ recorded by HDOH Kona station never exceeded the EPA 24-h-mean threshold of 35 µg/m³ (**Figure 3A**). The WHO 24-h-mean guideline of 25 µg/m³ was exceeded 1.2 % of the time at the Kona site (33 exceedance events). The maximum $PM_{2.5}$ 24-h-mean mass concentrations was 33 µg/m³, recorded in April 2016 (**Table 1**).

During the 2018 LERZ eruption, $PM_{2.5}$ exceeded the 24h-mean 35 µg/m³ EPA threshold 8.4 % of the time at the Kona station (eight exceedance events). The lower guideline of 25 µg/m³ established by the WHO was exceeded 34.7 % of the time at the Kona site (33 exceedance events). The maximum

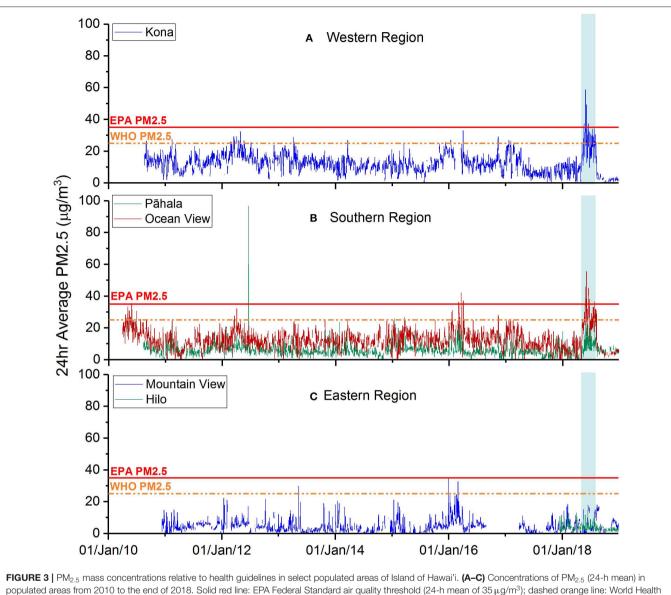
Station location Hilo Elevation: 121 m asl 40 km NE of summit 35 km NW of Fissure 8	Measurement period	Sulfur dioxide (SO ₂)						Particulates (PM _{2.5})
		(SO₂) (μg/m³) 24-h mean		Exceedance Hawai'i threshold (24-h > 366 μg/m³)	Exceedance EC threshold (24-h > 125 μg/m ³)	(PM _{2.5}) (µg/m ³) 24-h mean		Exceedance EPA threshold (24-h > 35 μ g/m ³)	Exceedance WHO guideline (24-h > 25 μg/m³)
		Average	7			Average	_		
		σ 19	- 1 day 0%	24 days 0.6%	σ	_	[No Data]	[No Data]	
		Max	403	3		Max	-	-	
	High Emission 2018 LERZ **	Average	6	0 days	1 day 1.1%	Average	5	0 days 0%	0 days 0%
		σ	21			σ	2		
		Max	144	_		Max	15		
Mountain View Elevation: 426 m asl 26 km NE of summit 23 km NW of Fissure 8	Low Emission ***	Average	_	[No Data]	[No Data]	Average	4	1 day 0%	3 days 0.1%
		σ	-			σ	4		
		Max	_			Max	35		
	High Emission 2018 LERZ **	Average	-	 [No Data]	[No Data]	Average	7	O days	0 days 0%
		σ	-			σ	6		
		Max	-	-		Max	18		
Volcano Observatory Elevation: 1,161 m asl 2 km NW of summit 35 km W of Fissure 8	Low Emission *	Average	24	28 days	204 days 5.1%	Average	_	 [No Data]	[No Data]
		σ	67			σ	-		
		Max	1068	-		Max	-		
	High Emission 2018 LERZ ***	Average	56	2 days	10 days 10.5%	Average		- [No Data]	[No Data]
		σ	106			σ	-		
		Max	450	_		Max	-		
Pāhala Elevation: 320 m asl 30 km SW of summit 66 km SW of Fissure 8	Low Emission *	Average	81	30 days 0.7%	729 days 18.1%	Average	6	2 days 0.1%	3 days 0.1%
		σ	72			σ	5		
		Max	776			Max	97		
	High Emission 2018 LERZ **	Average	129	5 davs	34 days 35.8%	Average	10	O days 0%	0 days 0%
		σ	108			σ	5		
		Max	555	—		Max	24		

(Continued)

TABLE 1 | Continued

Station location	Measurement period	Sulfur dioxide (SO ₂)					Particulates (PM _{2.5})				
		(SO ₂) (μg/m ³) 24-h mean		Exceedance Hawai'i threshold Exceedance EC threshold $(24-h > 366 \ \mu g/m^3) \qquad (24-h > 125 \ \mu g/m^3)$		(PM _{2.5}) (μg/m ³) 24-h mean		Exceedance EPA threshold (24-h > 35 μ g/m ³)	Exceedance WHO guideline (24-h > 25 μ g/m ³)		
		Average	26	_		Average	12				
Ocean View Elevation: 862 m asl 61 km W of summit 100 km W of Fissure 8	Low Emission ***	σ	39	3 days 0.1%	93 days 3.5%	σ	5	4 days 0.1%	25 days 0.9%		
		Max 403	-		Max	42	-				
	High Emission 2018 LERZ **	Average	117	4 days	27 days 28.4%	Average	26	- 10 days 10.5%	44 days 46.3%		
		σ	114			σ	8				
		Max	728	_		Max	56				
		Average	7			Average	12				
Kona Elevation: 517 m asl 67 km W of summit 106 km W of Fissure 8	Low Emission *	σ	7	0 days	0%	σ	5	0 days	33 days 1.2%		
		Max	79			Max	33				
	High Emission 2018 LERZ **	Average	39			Average	24				
		σ	25	0 days 0%	1 days 1.1%	σ	9	8 days 8.4%	33 days 34.7%		
		Max	136			Max	59				

All units in µg/m³. Exceedances of air quality standards are indicated for 24 h means, and calculated as percentage of total measurement duration. Distances between emission points and measurements sites are straight line distances; the emissions will not always follow the most direct route from near- to far-field. Note: no data available for PM_{2.5} at Volcano Observatory, or Hilo for 2010–2017; no data available for SO₂ at Mountain View. *Low emission period for SO₂ from the first of January 2007 to the thirty-first of December 2017 and for PM_{2.5} from the first of September 2010 to the thirty-first of December 2018. LERZ period for SO₂ and PM_{2.5} from the third of May 2018 to the sixth of August 2018. ***Exceptions due to data availability: Mountain View low emission for PM_{2.5} from the first of December 2010 to the thirty-first of December 2017; Volcano Observatory high emission 2018 LERZ period for SO₂ from the third of May 2018 to the third of July 2018; Ocean View low emission for SO₂ from twenty-third of August 2010 to the thirty-first of December 2017.



Organization (WHO) air quality guideline (24-h mean of $25 \,\mu$ g/m³); blue highlight: 2018 LERZ eruption duration.

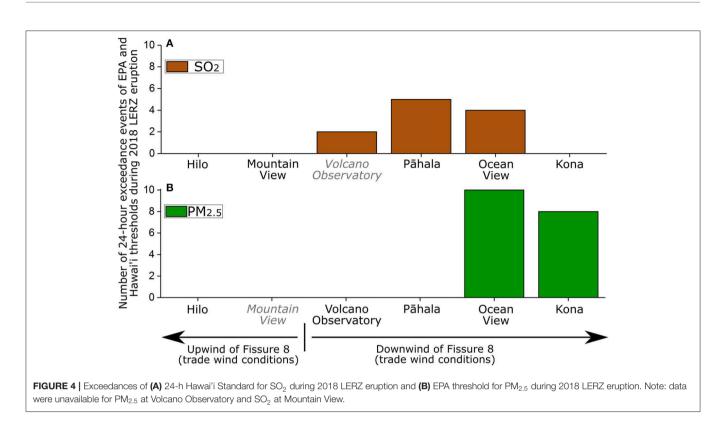
 $PM_{2.5}$ 24-h-mean mass concentration at the site was recorded as 59 µg/m³ on the twenty-ninth of May 2018.

4.2.2. Southern Region: Pāhala and Ocean View

During the period 2010–2017, the southern region of the Island of Hawai'i experienced variable levels of $PM_{2.5}$ (**Figure 3B**). The maximum $PM_{2.5}$ 24-h-mean mass concentration recorded in Ocean View was 42 µg/m³, recorded in March 2016. In Pāhala the maximum recorded $PM_{2.5}$ 24-h-mean mass concentration was 97 µg/m³, recorded on the eighteenth of June 2012, and coincident with two brush fires in the vicinity of Pāhala which burned approximately 5,600 acres (Hawai'i Emergency Management Agency, 2018). The EPA 35 µg/m³ PM_{2.5} threshold was exceeded 0.1 % of the time at both Ocean View and Pāhala (four exceedance events and two exceedance events, respectively).

The lower $PM_{2.5}$ guideline of 25 µg/m³ established by the WHO was exceeded 0.9 % of the time at Ocean View (25 exceedance events) and 0.1 % of the time at Pāhala (three exceedance events).

Mean PM_{2.5} mass concentrations in Pāhala during the 2018 LERZ eruption were higher than the 2010–2017 average ($10 \mu g/m^3$ with respect to $6 \mu g/m^3$) (**Table 1**), however there were no 24-h periods which exceeded either the EPA or WHO 24-h-mean thresholds. During the 2018 LERZ eruption, PM_{2.5} recorded in Ocean View exceeded the 35 $\mu g/m^3$ EPA threshold 10.5 % of the time (ten exceedance events) and exceeded the WHO 24-h-mean guideline 46.3 % of the time (44 exceedance events). In mid-June 2018, the Ocean View HDOH station recorded 3 consecutive days where 24-h-mean mass concentrations exceeded 35 $\mu g/m^3$, unprecedented in the period 2010–2017. The maximum 24-h-mean mass concentration



recorded in Ocean View during the 2018 LERZ eruption was $56 \,\mu g/m^3$, somewhat higher than the peak 24-h-mean mass concentration recorded at Ocean View in 2016 (42 $\mu g/m^3$).

4.2.3. Eastern Region: Hilo and Mountain View

HDOH stations in the eastern region of the Island of Hawai'i recorded variable levels of PM_{2.5} during 2010–2017 (**Figure 3C**). The maximum PM_{2.5} 24-h-mean mass concentration in Mountain View was $35 \,\mu\text{g/m}^3$, which was recorded in December 2015, and was the only exceedance of the $35 \,\mu\text{g/m}^3$ 24-h-mean EPA threshold during the period 2010–2017 (**Figure 3C**). The lower PM_{2.5} 24-h-mean guideline of $25 \,\mu\text{g/m}^3$ established by the WHO was exceeded 0.1 % of the time at Mountain View (three exceedance events).

During the 2018 LERZ eruption, $PM_{2.5}$ mass concentrations in Mountain View were higher than the average mass concentrations for 2010–2017 (7 and 4 µg/m³, respectively) (**Table 1**). However, during the 2018 LERZ eruption, $PM_{2.5}$ mass concentrations did not exceed either the EPA threshold of 35 µg/m³ or the WHO guideline of 25 µg/m³.

5. DISCUSSION

5.1. Dispersal of Volcanic Emissions

Time series of SO_2 mass concentrations were analyzed in five populated areas on the Island of Hawai'i; Hilo, Volcano Observatory, Pāhala, Ocean View and Kona, for the period 2007– 2018 (**Figure 2**). Time series of $PM_{2.5}$ were analyzed in Hilo, Mountain View, Pāhala, Ocean View, and Kona for the duration 2010–2018 (**Figure 3**). Significant escalations in emissions from Kīlauea volcano can be identified (**Figure 2A**), which were registered by the air quality monitoring instruments around the Island of Hawai'i (**Table 1**, section 4.1 and 4.2).

High mass concentrations of SO2 and PM2.5 generally occurred in the southern and western parts of the Island of Hawai'i. Prevailing trade winds from the north-east dispersed SO2 emissions from Kilauea volcano toward communities in the south and west of the island, as reported in previous studies (Longo et al., 2005, 2008; Michaud et al., 2007; Tam et al., 2016). During the 2018 LERZ eruption, the EPA 24-h-mean threshold for $PM_{2.5}$ (35 µg/m³) and the Hawai'i threshold for SO_2 $(366 \,\mu\text{g/m}^3)$ were exceeded in the south and west of the island (Figure 4). HDOH stations in the south of the island, 35–100 km away from Fissure 8, recorded 24-h events where SO₂ exceeded Hawai'i thresholds (note that there were no HDOH or NPS permanent monitoring stations for SO₂ in proximal location to the 2018 LERZ eruption site), but PM2.5 EPA exceedance events only occurred at HDOH stations 100 km or further away from Fissure 8 (Figure 4). This spatial variance between distribution of PM2.5 and SO2 is well-documented and thought to reflect the timescale of oxidation of sulfur dioxide gas into sulfate aerosol during dispersion (Cadle et al., 1971; Stockwell and Calvert, 1983; Porter et al., 2002; Ilyinskaya et al., 2017).

Volcanic emissions at source commonly consist of a mixture of silicate ash particles, various gases and non-silicate aerosol (Oppenheimer and McGonigle, 2004; von Glasow et al., 2009; Langmann, 2014). The lifetime of SO_2 in the lower troposphere is generally considered to be on the order of 1–3 days to a week (Allen et al., 2002; Rotstayn and Lohmann, 2002; Pfeffer et al., 2006a; Pattantyus et al., 2018), the rate of conversion

depending on relative humidity and temperature, the availability of oxidants, and interaction with cloud or fog (Saxena and Seigneur, 1987; Oppenheimer et al., 1998). However, the SO₂ oxidation pathways in a volcanic cloud are not necessarily the same as under background conditions (Galeazzo et al., 2018). Through a variety of reaction pathways (including oxidation with the hydroxyl radical, OH, and with hydrogen peroxide, H_2O_2 , and O_3), SO₂ in volcanic clouds is gradually converted to sulfate aerosol (Stockwell and Calvert, 1983; Allen et al., 2002), which is a dominant component of volcanic PM_{2.5} (Tam et al., 2016; Pattantyus et al., 2018). The conversion rate of SO₂ to sulfate aerosol is important for estimating the potential hazard of volcanic PM_{2.5} to human health and the downwind environments (Kroll et al., 2015).

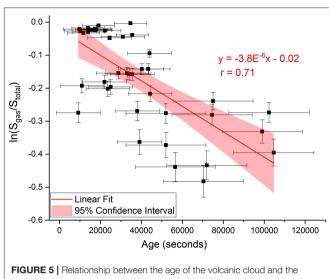
HDOH stations measure the ambient air, which contains SO₂ and PM_{2.5} derived from anthropogenic sources as well as natural non-volcanic and volcanic sources. In order to determine the influence of the volcanic eruption on the measured SO₂ and aerosol abundances, it is first necessary to calculate the volcanic component of the HDOH measurements. During the 2018 LERZ eruption, Fissure 8 was the dominant source of volcanic SO₂ emissions on the island, and following the decline of the eruption, the SO₂ and PM_{2.5} mass concentrations decreased to below pre-LERZ eruption levels at all HDOH stations analyzed in this study (Figures 2, 3). The mass of pollutants recorded at HDOH stations during this post-LERZ eruption period (mid-August 2018 to the first of February 2019) are therefore used to define the background abundances arising from all other nonvolcanic sources. During this time there was some SO₂ emitted from Kilauea's summit but at the lowest rate measured in decades at 0.1 kt/year (Nadeau et al., 2019). The volcanic component of the HDOH measurements was calculated by subtracting the average PM2.5 and SO2 mass concentration following the end of the 2018 LERZ eruption for each station from the mass concentrations measured during the LERZ eruption, to estimate the volcanogenic component. The sulfate aerosol component within the volcanic PM2.5 mass concentration was then estimated to be in the range 77-92 %, following the methods of Mather et al. (2012) and Kroll et al. (2015), of PM2.5 composition from Kīlauea.

Estimating the conversion rate of SO₂ to sulfate from SO₂ and SO₄²⁻ datasets is not straightforward because several processes can occur simultaneously, including SO₂ oxidation to sulfate, dispersion-dilution and deposition of SO₂ and/or SO₄²⁻ to the surface.

Here, a first-order decay constant for SO_2 is estimated by the relationship between volcanic components of SO_2 and SO_4^{2-} , as follows;

$$\ln\left(\frac{S_{gas}}{S_{total}}\right) = -kt \tag{1}$$

where S_{gas} is the sulfur component of the volcanic SO₂ mass concentration (µg/m³), S_{total} is the sum of sulfur components of the volcanic SO₄²⁻ and SO₂ mass concentrations (µg/m³), *t* is the age of the volcanic cloud (seconds) and -k is a first-order decay constant.



volcanic sulfur components measured at Pāhala, Ocean View and Kona HDOH ambient air quality stations. S_{gas} is the sulfur component of the volcanic SO_2 mass concentration, S_{total} is the sum of sulfur component of the volcanic SO_2^{-2} and SO_2 mass concentrations. Age is derived from HYSPLIT back-trajectory simulations (llyinskaya et al., in preparation). Y axis error bars indicate range of SO_4^{-2} component in volcanic $PM_{2.5}$ composition from Kilauea. X axis error bars indicate one standard deviation of HYSPLIT back-trajectory results specific to Kona, Ocean View and Pāhala. Correlation trendline based on the data points, with a first-order decay constant of $3.8 \times 10^{-6} \text{ s}^{-1}$ with a 95 % confidence interval of $\pm 1.26 \times 10^{-6} \text{ s}^{-1}$.

The age of the volcanic cloud is here considered to be the time between emission of the cloud at the LERZ eruption source point and subsequent measurement at the HDOH station. From back-trajectory HYSPLIT simulations run between the HDOH ambient air quality stations and Fissure 8, the average age of the emissions and dispersal distance was calculated for dates between the eighteenth of July to the second of August 2018 (Ilyinskaya et al., in preparation). An estimate of the average first-order decay constant from our data-set is indicated in Figure 5. Although there is considerable scatter in the data, a broad trend of decreasing S fraction in the gas phase is apparent and a linear fit allows us to estimate a first-order rate constant of 3.8 imes 10⁻⁶ s⁻¹ with a 95 % confidence interval of \pm 1.26×10^{-6} s⁻¹. This first-order decay constant for SO₂ relative to total sulfur can represent an estimate of the average rate of SO₂ oxidation to sulfate only if negligible sulfur deposition has occurred. Nevertheless, our value is similar to the SO₂ oxidation rate calculated by Kroll et al. (2015) from direct measurements of sulfur in gas and particle phase in Kilauea's emission cloud from the summit to Pāhala. Kroll et al. (2015) identified a diurnal cycle in measured sulfate as a fraction of total sulfur, from which they calculated a noontime instantaneous SO2 oxidation rate of $2.4 \times 10^{-6} \text{ s}^{-1}$.

5.2. Reliability Assessment of Community-Operated PM_{2.5} Instruments

A subset of the community-operated PurpleAir instruments on the Island of Hawai'i were selected for intercomparison

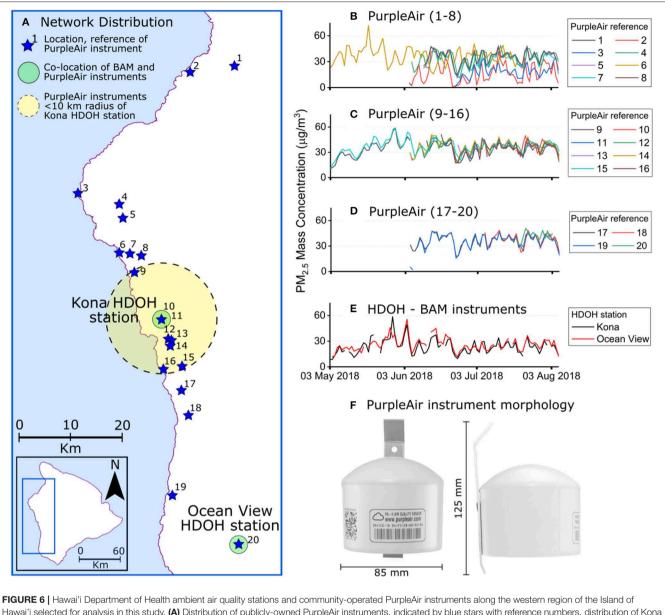
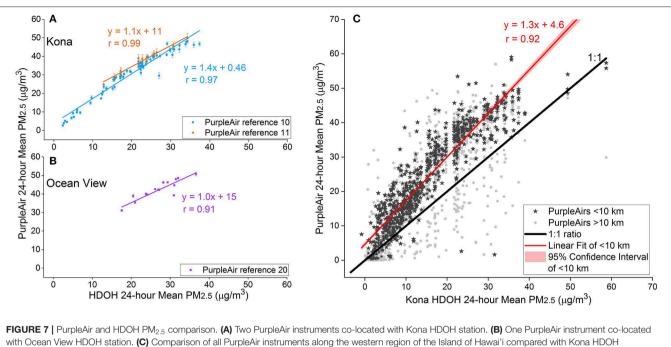


FIGURE 6 Hawa'l Department of Health ambient air quality stations and community-operated PurpleAir instruments along the western region of the Island of Hawai'l selected for analysis in this study. (A) Distribution of publicly-owned PurpleAir instruments, indicated by blue stars with reference numbers, distribution of Kona and Ocean View HDOH stations indicated by green circles. PurpleAir instruments within 10 km radius of Kona HDOH station indicated by dashed yellow circle. (B–D) 24-h average mass concentration measurements from 20 PurpleAir instruments during the 2018 LERZ eruption; (E) 24-h average PM_{2.5} from BAM instruments at Kona and Ocean View HDOH ambient air quality stations during the 2018 LERZ eruption. (F) Size and morphology of the PurpleAir instruments, seen from the front (left image) and side (right image). Photos by kind permission of PurpleAir LLC.

with the established institutional data-sets across the western region of the island. The low-cost and portable nature of the PurpleAir instruments (**Figure 6F**) facilitated installation of a monitoring network across the western region (**Figure 6A**), with a high spatial resolution of measurements in comparison to the locations of HDOH ambient air quality sites (**Figure 6A**). This can be advantageous to capture the effects of local topographic and meteorological factors, which may influence dispersion of and deposition from volcanic plumes. Mass concentrations from PurpleAir instruments and HDOH PM_{2.5} instruments during the course of the 2018 LERZ eruption are presented in Figures 6B–E.

Small differences were found between individual PurpleAir instruments in the same location. Two PurpleAir instruments (**Figure 6A**, PurpleAir references 10 and 11) were co-located at the Kona HDOH station, and the PurpleAir instruments ran together for 16 days. During this time, the maximum absolute differences in 24-h average measurements between the PurpleAir instruments was $2.3 \,\mu\text{g/m}^3$; which was 6 % of the total



with Ocean View HDOH station. **(C)** Comparison of all PurpleAir instruments along the western region of the Island of Hawai'i compared with Kona HDOH measurements. PurpleAir instruments within 10 km of Kona HDOH indicated by dark gray stars, PurpleAir instruments at greater distances are indicated by light gray circles. Linear regression and 95 % confidence interval fit to instruments <10 km from Kona HDOH ambient air quality station. Measurement periods vary with earliest PurpleAir data from August 2017 and most recent from January 2019. **(A,B)** PurpleAir error bars show measurement variations between the two sensors within a single PurpleAir instrument, and therefore indicate the minimum error.

measured concentration. Correlation between the two PurpleAir instruments was very strong (Pearson's r = 0.99). Similar results were found by Malings et al. (2019) with co-location of nine PurpleAir instruments at a site in Pennsylvania for a period of 66 days (Pearson's r > 0.9). The high correlation between co-located individual PurpleAir instruments indicates high standardization. With this being the case, relative PM_{2.5} mass concentrations measured by PurpleAir instruments over a wider geographical area should be comparably reliable.

Three PurpleAir instruments were co-located with HDOH PM_{2.5} instruments (BAM) to determine the accuracy of PurpleAir instrument PM2.5 measurements in relation to reference-grade instruments (Figure 6A, PurpleAir references 10, 11, and 20). The co-located PurpleAir measurements correlated well with the BAM measurements, with Pearson's r values of 0.99, 0.97, and 0.91 (Figures 7A,B). However, PurpleAir instruments did record higher mass concentrations of PM_{2.5} in comparison to the BAM analyzers (Figures 7A,B). PurpleAir reference 10 (co-located with Kona HDOH station) (Figure 6A) recorded the greatest measurement off-set, with up to 40 % higher mass concentrations of PM2.5 relative to the Kona BAM analyzer measurements (Figure 7A). This trend was also found when the community-operated PurpleAir instruments across the western region of the island were compared to the BAM at Kona HDOH station (Figure 7C), with strong correlation between PurpleAir instruments and BAM (Pearson's r = 0.92) but an average 30 % higher mass concentrations measured by PurpleAir instruments within 10 km of the Kona BAM instrument. Previous testing of PurpleAir instruments in Pennsylvania and California has yielded similar findings (AQ-SPEC, 2017; Malings et al., 2019), with over-estimation of PM_{2.5} mass concentrations measured by PurpleAir instruments relative to BAM reference-grade analyzers.

Some discrepancy between PurpleAir and BAM measurements may be expected, as it is well-known that low-cost instruments measuring PM2.5 with light scattering methods have not historically agreed with measurements obtained from reference grade instruments with different operating principles (Watson et al., 1998; Wilson et al., 2002; Chow et al., 2008; Burkart et al., 2010). In comparison with sensors of other operating principles, light-scattering optical particle sensors have been shown to suffer effects of relative humidity (Wang et al., 2015; Crilley et al., 2018), since the operating principle relies on indirect measurement of particle size and shape based on scattered light, and an assumed particle shape and refractive index. Conversely, BAM instruments measure direct changes in aerosol mass concentrations based on the loss of electrons on a filter which the aerosol has been deposited on (Watson et al., 1998; Manikonda et al., 2016), and so are not influenced by the hygroscopic growth of individual particles. Zheng et al. (2018) analyzed the performance of Plantower PMS3003 (an earlier version of the Plantower PMS5003 housed in PurpleAir instruments used in this study) against a reference grade scattered light spectrometer (with good correlation of $R^2 = 0.8$) and a BAM instrument (with lower correlation of $R^2 = 0.5$). They concluded that a likely explanation contributing to the discrepancy is the potential for hygroscopic growth of aerosol particles due to ambient humidity, which alters the light-scattering properties of the aerosol and therefore the measurement made by the optical sensor (Watson et al., 1998; Cabada et al., 2004; Spinetti and Buongiorno, 2007; Jayaratne et al., 2018). In this instance, Zheng et al. (2018) found that the low-cost light-scattering sensor correlated best with the reference-grade instrument of the same operating principle, finding a lower correlation against the reference-grade instrument operating on principles other than light-scattering. The PurpleAir instrument over-estimation of PM_{2.5} relative to the BAM reference-grade instrument may therefore be due to influences of humidity acting on the light scattering (Zheng et al., 2018).

An additional consideration in explaining the overestimation of PM2.5 by the PurpleAir instruments relative to the referencegrade BAM is the particle density of the measured particulates. The Plantower PMS5003 sensors contained within the PurpleAir instruments provide mass concentration measurements which are calculated by an unreported atmospheric calibration factor (Zheng et al., 2018). We assume that this calibration factor uses an average particle density, likely similar to that for an urban environment, such as 1.65 g cm⁻³ (Pitz et al., 2003; Liu et al., 2015; Crilley et al., 2018). If the Plantower PMS5003 sensor measures ambient air with a particle density dissimilar to the average particle density used in the atmospheric calibration factor, the resulting sensor output would be biased. For example, introduction of volcanic aerosol, primarily sulfate with an average particle density of 1.77 g cm⁻³ (Sarangi et al., 2016), into the air measured by the sensor could result in sensor output bias as a result of dissimilarity between the real and assumed particle density.

Despite the PM_{2.5} over-estimation, the strong correlation between the PurpleAir and BAM instruments indicates that the PurpleAir instruments provided qualitatively valuable measurements of the atmospheric conditions. The high degree of intra-instrument performance, similar to findings by Malings et al. (2019), indicates that the PurpleAir instruments are reliable for determining relative variations in PM_{2.5}. The dense network of instruments with a high spatial resolution along the western region of Hawai'i during the 2018 LERZ eruption gives a good indication of the relative amounts of PM2.5 across the region, at a finer spatial resolution than available from the sparselylocated BAM instruments (Figure 6A). Low-cost communityoperated networks, such as the PurpleAir instruments across the western region of Hawai'i during the 2018 LERZ eruption, can therefore be invaluable in providing insights into smallerscale heterogeneities in air quality across a regional area at a scale inaccessible by the usually more disperse reference-grade instruments (Figures 6B-E). Additionally, as far as the authors are aware, this is the first validation of PurpleAir instruments in a volcanic environment. Their strong correlation to the BAM instruments along the west coast of the Island of Hawai'i during the 2018 LERZ eruption indicates that they are suitable for augmenting reference-grade instrument networks in periods of volcanic unrest. Considering that these instruments also provide a source of open-access data to the public, they present an opportunity to improve community awareness and inclusion of the general public in hazard assessment of downwind volcanic PM_{2.5} air pollution.

6. CONCLUSIONS

Kilauea's 2018 eruption was the largest LERZ eruption in the last two centuries. SO₂ emissions reached a monthly average of 200 kt/day during June (Kern et al. 2019), significantly exceeding emissions from Kilauea during 2008-2017, which averaged 5-6 kt/day (Eguchi et al., 2011; Beirle et al., 2014; Carn et al., 2016; Elias et al., 2018). During the 2018 LERZ eruption, SO₂ mass concentrations exceeding the Hawai'i 24-h-mean threshold $(366 \,\mu\text{g/m}^3)$ primarily occurred in the south of the island, at Volcano Observatory and in Pāhala and Ocean View (2.1, 5.3, and 4.2 % of the time during the 3-month long eruption, respectively). SO₂ mass concentrations were elevated at the HDOH Kona station (average 24-h-mean mass concentration of $39 \,\mu\text{g/m}^3$ during the LERZ eruption, relative to $7 \,\mu\text{g/m}^3$ during 2007-2017), but mass concentrations were highest in Ocean View, Pāhala and at Volcano Observatory (average 24-h-mean mass concentrations of 117, 129, and $56 \,\mu\text{g/m}^3$, respectively). The Hawai'i 24-h-mean threshold was exceeded five times in Pāhala and twice at Volcano Observatory, but peak mass concentrations did not exceed those from the period 2007-2017. In Ocean View, the Hawai'i 24-h-mean threshold was exceeded four times and 24-h-mean mass concentrations peaked at 728 μ g/m³, almost double the previous peak measurement of $403 \,\mu\text{g/m}^3$ recorded at that station in January 2016.

 $PM_{2.5}$ mass concentrations recorded at HDOH stations around the island from 2010 to 2017 rarely exceeded the EPA 24h-mean threshold of $35 \,\mu\text{g/m}^3$ (0.1 % of the time at both Ocean View and Pāhala). The lower 24-h-mean $PM_{2.5}$ limit ($25 \,\mu\text{g/m}^3$) set by the World Health Organization was exceeded with greater frequency, particularly in Kona and Ocean View (1.2 and 0.9 % of the time, respectively). During the 2018 LERZ eruption, PM_{2.5} air pollution was significantly higher than 2010–2017 levels in Kona and Ocean View, exceeding WHO guidelines 34.7 and 46.3 % of the time, respectively. Peak 24-h-mean mass concentrations in Ocean View were recorded at 56 $\mu\text{g/m}^3$, and 3 consecutive days in June were recorded with mean 24-h mass concentrations exceeding 35 $\mu\text{g/m}^3$. The Kona HDOH stations recorded eight 24-h periods which exceeded EPA thresholds, unprecedented in the 2010–2017 period.

Following the decline of the 2018 LERZ eruption, mass concentrations of both SO_2 and $PM_{2.5}$ measured at the HDOH stations decreased to below pre-LERZ eruption levels, indicating that a large proportion of the air quality anomalies measured during the eruption were volcanogenic. The post-LERZ HDOH measurements are here assumed to be representative of the background (non-volcanically-perturbed) atmosphere, and subtracting these abundances from those recorded during the LERZ eruption provides an estimate of the purely volcanogenic $PM_{2.5}$ and SO_2 . The sulfate aerosol component within the volcanic $PM_{2.5}$ is calculated as between 77 and 92 %, following Mather et al. (2012). HYSPLIT back-trajectory

simulations provide an estimate of emission age following dispersion from source to measurement point (Ilyinskaya et al., in preparation), and a first-order SO₂ decay constant is estimated at 3.8×10^{-6} s⁻¹.

Community-operated PurpleAir instruments provided a high spatial resolution network across the western region of the island, informing the public regarding PM2.5 mass concentrations in their locality. Low measurement variability (Pearson's r = 0.99) was found between co-located individual PurpleAir instruments, indicating a high level of intrainstrument performance. Observations recorded by co-located PurpleAir and BAM instruments correlated well (Pearson's r = 0.99, 0.97, and 0.94), but PurpleAir instruments were found to overestimate the PM25 mass concentration by up to 40 %, relative to the BAM instrument. This likely reflects inherent differences in instrument operating principles and may be associated with changes in optical-properties of aerosol arising from hygroscopic growth in ambient humidity. Nevertheless, the PurpleAir instruments are suitable for providing a lowcost network to augment reference-grade instruments, and contribute an open-access source of readily-available information to the public leading to development of community awareness toward air quality.

This study has assessed the impacts to air quality in downwind communities around the Island of Hawai'i from 2007 to 2018. Spatial variability of air quality around the island during the 2018 LERZ eruption was comparable to patterns identified over the previous decade, but $PM_{2.5}$ and SO_2 pollution levels resulting from the 2018 LERZ eruption were significantly higher in western and southern regions of the island. A study of the potential health burden of these significant impacts on air quality might further illuminate aspects of the dose-response to volcanogenic emissions in addition to the impacts of public health protection measures put in place during the eruption.

DATA AVAILABILITY STATEMENT

The datasets analyzed for this study can be found in the Hawai'i Department of Health Ambient Air Quality Data repository [http://health.hawaii.gov/cab/hawaii-ambientair-quality-data/], and in the PurpleAir sensor list repository [https://www.purpleair.com/sensorlist].

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AUTHOR CONTRIBUTIONS

RW performed the data analysis and wrote the original draft. EI, AS, TR, MP, and TM contributed to data interpretation. All co-authors contributed to draft review and editing.

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Conflict of Interest: AD was employed by company PurpleAir LLC.

The remaining authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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