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Impact of the 2021 La Palma volcanic eruption on air quality: Insights from a multidisciplinary approach

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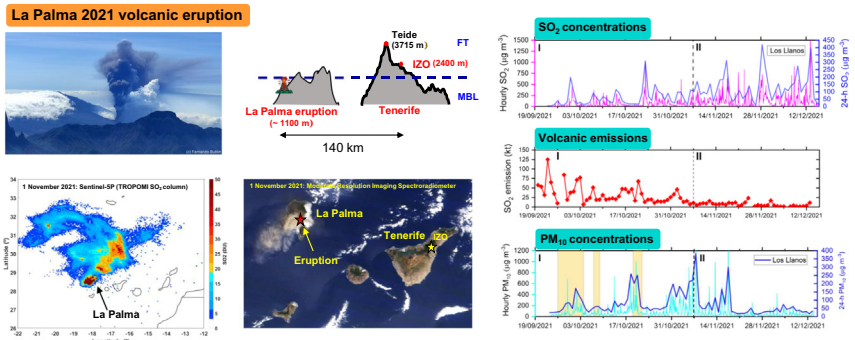
HIGHLIGHTS

- La Palma 2021 volcanic eruption had a large impact on air quality.
- Ground, satellite and height-resolved measurements provide crucial information.
- SO_2 concentrations at ground-level showed opposite temporal trend to SO_2 emissions.
- Volcanic and desert dust aerosols both impact lower troposphere during the eruption.
- Atmospheric vertical stratification plays key role in eruption impact on air quality.

GRAPHICAL ABSTRACT



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ABSTRACT

The La Palma 2021 volcanic eruption was the first subaerial eruption in a 50-year period in the Canary Islands (Spain), emitting ~1.8 Tg of sulphur dioxide (SO_2) into the troposphere over nearly 3 months (19 September-13 December 2021), exceeding the total anthropogenic SO_2 emitted from the 27 European Union countries in 2019. We conducted a comprehensive evaluation of the impact of the 2021 volcanic eruption on air quality (SO_2, PM_{10} and $PM_{2.5}$ concentrations) utilising a multidisciplinary approach, combining ground and satellite-based measurements with height-resolved aerosol and meteorological information. High concentrations of SO_2, PM_{10} and $PM_{2.5}$ were observed in La Palma (hourly mean SO_2 up to ~2600 μg m^{-3} and also sporadically at ~140 km distance on the island of Tenerife (> 7700 μg m^{-3}) in the free troposphere. PM_{10} and $PM_{2.5}$ daily mean concentrations in La Palma peaked at

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~380 and 60 μg m^{-3}. Volcanic aerosols and desert dust both impacted the lower troposphere in a similar height range (~ 0–6 km) during the eruption, providing a unique opportunity to study the combined effect of both natural phenomena. The impact of the 2021 volcanic eruption on SO_2 and PM concentrations was strongly influenced by the magnitude of the volcanic emissions, the injection height, the vertical stratification of the atmosphere and its seasonal dynamics. Mean daily SO_2 concentrations increased during the eruption, from 38 μg m^{-3} (Phase I) to 92 μg m^{-3} (Phase II), showing an opposite temporal trend to mean daily SO_2 emissions, which decreased from 34 kt (Phase I) to 7 kt (Phase II). The results of this study are relevant for emergency preparedness in all international areas at risk of volcanic eruptions; a multidisciplinary approach is key to understand the processes by which volcanic eruptions affect air quality and to mitigate and minimise impacts on the population.

1. Introduction

The release of gases and aerosols during volcanic eruptions can have many impacts on the atmosphere, including on air quality, aviation and climate (Langmann et al., 2012; Gettelman et al., 2015; Ilyinskaya et al., 2017) and on the biosphere, for example on terrestrial, marine and freshwater ecosystems (e.g. Caballero et al., 2022; Nogales et al., 2022; Román et al., 2022; Weiser et al., 2022). Volcanic gases mainly include water vapour (H_2O), carbon dioxide (CO_2), sulphur dioxide (SO_2), hydrogen sulphide (H_2S), carbon monoxide (CO) and hydrogen halogenides (mainly HCl and HF), some of which are reactive in the atmosphere. Volcanic aerosols include both primary particulate emissions, of different size fractions including ash (defined as particles with diameter < 2 mm) and secondary particulates (e.g. sulphates, metal-bearing aerosols) (Stewart et al., 2021). The impact of volcanic air pollution on human health has recently been reviewed by Stewart et al. (2021) who report studies confirming that exposure to volcanic air pollution can have adverse effects on both respiratory and cardiovascular health as well as other effects such as ocular and dermal irritation.

Air quality and atmospheric impacts of volcanic eruptions have been identified locally (e.g. Searl et al., 2002; Gíslason et al., 2015; Andronico and Del Carlo, 2016; Ilyinskaya et al., 2017; Cazorla and Herrera, 2020; Tang et al., 2020; Whitty et al., 2020; Carlsen et al., 2021; Crawford et al., 2021; Trejos et al., 2021) and have also been reported hundreds or thousands of kilometres from the eruption location (e.g. Tu et al., 2004; Newnham et al., 2010; Schäfer et al., 2011; Schmidt et al., 2015; Twigg et al., 2016). For example the April–May 2010 eruption of Eyjafjallajökull Volcano, Iceland caused widespread disruption throughout Europe due to volcanic ash in the atmosphere affecting aviation safety (Thomas and Prata, 2011; Langmann et al., 2012). However, despite these studies, there are many locations affected by volcanic eruptions worldwide where air quality measurements are not available (Stewart et al., 2021). It has been estimated that over a billion people live in the proximity of an active volcano (Freire et al., 2019), therefore it is important to understand the air quality impacts of volcanic eruptions.

Numerous studies have identified the adverse effects of air pollution on human health (e.g. WHO, 2013, 2016, 2018; Landrigan et al., 2018; Orellano et al., 2020, 2021), both from natural and anthropogenic sources, and in response, the World Health Organisation (WHO) set Air-Quality Guidelines (AQGs) for particulate matter (PM) (PM_{10} and $PM_{2.5}$, PM with aerodynamic diameter < 10 μm and < 2.5 μm, respectively), sulphur dioxide (SO_2), nitrogen dioxide (NO_2), and ozone (O_3). A global update of the WHO AQGs was provided in 2005 (WHO, 2006) and in 2021, new WHO AQGS were published (WHO, 2021) on the basis of evidence that adverse effects of air pollution are observed at lower concentration levels than previously understood. The European Commission (EC) air quality standards for air pollutants in ambient air sets various target and limit values for different pollutants and different averaging times (EC, 2008).

In this study, we examine the air quality impacts of the most recent eruption in the Canary Islands archipelago, which occurred in La Palma between 19 September and 13 December 2021 (lasting 85 days and 8 h) (PEVOLCA, 2021; González, 2022). This was the first subaerial eruption in a 50-year period in the Canary Islands, following the 1971 Teneguia and 1949 San Juan eruptions (also in La Palma). La Palma is the most volcanically active island in the Canary Islands in historical times, hosting six out of the twelve most recent subaerial eruptions (Hernández-Pacheco and Valls, 1982; Longpré and Felpeto, 2021). Volcanic unrest has been detected at Cumbre Vieja volcano since 2017 (Torres-González et al., 2020), but this unrest accelerated only eight days prior to the eruption (Longpré, 2021). The La Palma 2021 volcanic eruption, located in a zone known as Hoya de Tajogaite, was characterised by extensive lava emissions, (12.4 km^2 lava field), and a very dynamically evolving eruptive style with simultaneous lava fountains (>100 m high), strombolian and/or vulcanian activity, alternating between numerous vents on the eruptive fissure (PEVOLCA, 2021; Castro and Feisel, 2022). The eruption caused considerable destruction and damage to homes, buildings, crops and other infrastructures. La Palma has a population of 83,380 inhabitants (ISTAC, 2021), and a large proportion of the population live in towns and villages on the western side of La Palma close to the 2021 volcanic eruptive centre.

The Canary Islands are located in the sub-tropical eastern North Atlantic, and this region has some distinct features which affect the air quality impacts of the La Palma volcanic eruption, including for example the strong vertical stratification of the lower troposphere (Carrillo et al., 2016; Barreto et al., 2022a). In addition, this region is in the pathway of the long-range transport of mineral dust exported from the Sahara-Sahel region to the North Atlantic, which can expand westward to the Americas (Prospero and Mayol-Bracero, 2013; Tsamalis et al., 2013; Rodríguez et al., 2020; Prospero et al., 2021; Barreto et al., 2022a). This desert dust export from western Africa establishes a dust-laden air layer that is referred to as the Saharan Air Layer (SAL), this dust layer can periodically affect the Canary Islands and increase the particulate load significantly (Viana et al., 2002; Alonso-Pérez et al., 2007; Dominguez-Rodriguez et al., 2020; Rodríguez et al., 2020; Barreto et al., 2022b).

During the 2021 eruption, an unprecedented level of instrumentation and observational capacity were both already in place and deployed as part of the emergency response (ACTRIS, 2021; García et al., 2022; Lopez et al., 2022), involving collaboration among many public organizations and research groups. Observations were obtained from both ground and satellite-based instruments and numerical models using advanced scientific instrumentation and techniques in a manner that was not technically possible in previous subaerial eruptions in the Canary Islands. This level of observational capacity and multidisciplinary approach provides a unique possibility to investigate the 2021 volcanic eruption in La Palma and further develop the understanding of air quality impacts of volcanic eruptions. In this study we evaluate the local and regional impacts of the La Palma 2021 volcanic eruption, concentrating on the air quality impacts of SO_2 and PM concentrations (PM_{10} and $PM_{2.5}$), as they are considered the most important parameters for population-scale impacts of volcanic air pollution on a longer-term (Stewart et al., 2021).

2. Materials and methods

2.1. Study area

La Palma in the Canary Islands archipelago is located ~ 445 km west of the African continent in the North Atlantic (Fig. 1a). La Palma is crossed by a north-south volcanic ridge, reaching a maximum height of 2423 m a.s.l. at Roque de los Muchachos. The 2021 volcanic eruption was located on the western slopes of La Palma (Fig. 1c), at a pre-eruption

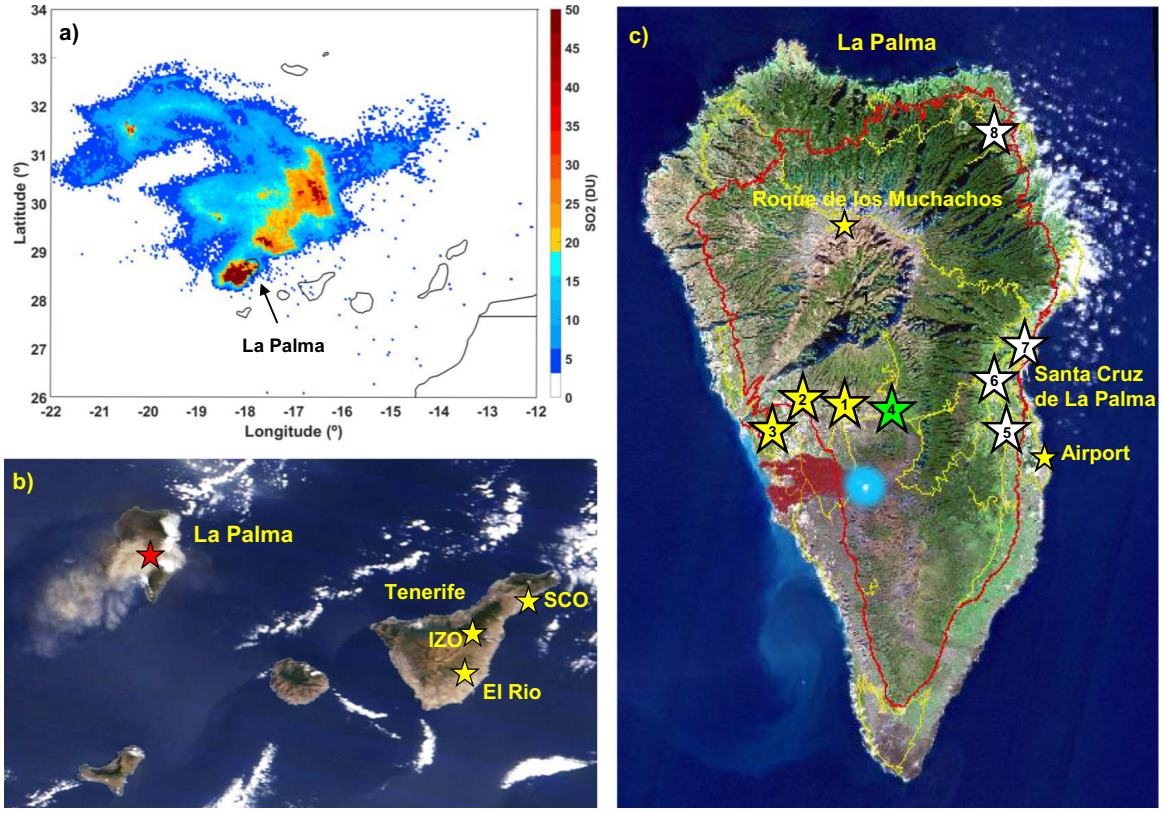

Fig. 1. Study site location. a) Satellite column integrated Sulphur Dioxide (SO_2) from Copernicus Sentinel-5P TROPOMI on 1 November 2021 indicating La Palma. b) Image for 1 November 2021 obtained by the Moderate Resolution Imaging Spectroradiometer (MODIS) sensor aboard the Terra satellite showing the western Canary Islands including La Palma and Tenerife, the location of the volcanic eruption is marked with a red star. The location of the Izaña Observatory (IZO), Santa Cruz Observatory (SCO) and the El Rio regional background measurement site are marked with yellow stars. c) Plan view of La Palma indicating the location of the volcanic eruption (blue circle), the lava flow on 17/12/2021 marked in dark red, the air-quality measurement sites, (1) El Paso, (2) Los Llanos de Aridane, (3) Tazacorte, and (4) the AEMET meteorological station (C126A). In addition, the permanent air-quality measurement sites on the eastern side of La Palma are also shown, (5) San Antonio, (6) La Grama, (7) El Pilar and (8) Las Balsas. The red and yellow lines mark roads. Credits: a) Copernicus Sentinel-5P TROPOMI (https://maps.s5p-pal.com/so2/), b) We acknowledge the use of imagery provided by services from NASA's Global Imagery Browse Services (GIBS), part of NASA's Earth Observing System Data and Information System and c) Instituto Geográfico Nacional (IGN) (https://ign-esp.maps.arcgis.com/).

altitude of ~ 900 m a.s.l. The maximum altitude of the volcanic edifice during the eruption was 1131 m a.s.l. (PEVOLCA, 2021).

The Canary Islands archipelago is generally dominated by the influence of the Azores anticyclone and the quasi-permanent subsidence conditions modulated by the descending branch associated with the Hadley cell. This causes the atmosphere in this region to be characterised by a strong vertical stratification of the lower troposphere, with a strong and stable temperature inversion layer, present for most of the year, with predominance of low-level north-easterly trade winds in the Marine Boundary Layer (MBL) and north-westerly winds above the inversion layer (Carrillo et al., 2016; Azorin-Molina et al., 2018; Barreto et al., 2022a). This predominant atmospheric scenario of background conditions (low aerosol concentration) is interrupted sporadically by desert dust outbreaks (higher aerosol loading) as detailed in the long-term study of aerosol characterization in the sub-tropical eastern North Atlantic (Barreto et al., 2022b).

2.2. Air quality measurements

Immediately after the La Palma volcanic eruption began on 19/9/2021, air quality monitoring stations, additional to the existing Canary Islands Air Quality Monitoring Network (AQMN), were deployed by the Canary Islands Government as part of the emergency response, with the collaboration of several public organizations including the Izaña Atmospheric Research Centre (IARC) which is part of the State Meteorological Agency of Spain (AEMET), and the Spanish National Research Council (CSIC).

In this study we analyze air quality data from the following three measurement sites which were deployed to aid the monitoring of the volcanic eruption, located on the western side of La Palma: 1) El Paso and 2) Los Llanos de Aridane (herein denoted as Los Llanos), measurements started on 23/9/2021 (sites operated by Canary Islands Government) and 3) Tazacorte, measurements started on 24/9/2021 (site operated by IARC-AEMET). The population of the three municipal districts of Los Llanos de Aridane, El Paso and Tazacorte, in which these three measurement sites are located, is ~ 33,000 inhabitants (ISTAC, 2021). Details of the location and further details of all the measurement stations utilised in this study are given in Table 1.

The Canary Islands AQMN had four permanent air quality monitoring stations in La Palma, prior to the volcanic eruption. They are all located on the eastern side of La Palma (Fig. 1c), separated from the eruptive centre on the western slope by the Cumbre Vieja volcanic rift which reaches a height of ~1450 m a.s.l due east of the eruptive centre. These four sites are: 1) San Antonio, Breña Baja, 2) La Grama, Breña Alta, 3) El Pilar, Santa Cruz de La Palma and 4) Las Balsas, San Andres and Sauces (Table 1).

In addition to the measurement sites in La Palma, we present data from the Izaña Observatory (IZO) in Tenerife and El Rio regional background measurement site, a Canary Islands AQMN site (Fig. 1b). The Izaña Observatory in Tenerife is a high-mountain (2373 m a.s.l.), remote background, permanent observatory located in the free troposphere (FT) ~ 140 km from the eruptive centre, managed by IARC-AEMET. The Izaña Observatory is a World Meteorological Organisation (WMO) Centennial observatory

Table 1
Location and details of measurement sites utilised in this study.

Site name and location	Latitude (° N)	Longitude (° W)	Altitude (m a.s.l.)	Distance from eruptive centre (km)	Site type	Data utilised in this study[a]	Institution
La Palma (west)							
El Paso	28.65	17.88	647	~ 4.4	Temporary[b]	SO_2, PM_{10}, $PM_{2.5}$	CIG AQMN
Los Llanos	28.66	17.91	351	~ 6.2	Temporary[b]	SO_2, PM_{10}, $PM_{2.5}$	CIG AQMN
Tazacorte	28.64	17.93	108	~ 6.9	Temporary[b]	SO_2, PM_{10}, $PM_{2.5}$, lidar profiles	IARC-AEMET
El Paso met station	28.65	17.85	844	~ 4.6	Meteorological station	Wind speed & wind direction at 10 m a.g.l.	AEMET
La Palma (east)							
San Antonio	28.65	17.77	208	~ 10	Suburban background	SO_2, PM_{10}, $PM_{2.5}$	CIG AQMN
La Grama	28.67	17.78	245	~ 11	Suburban industrial	SO_2, PM_{10}, $PM_{2.5}$	CIG AQMN
El Pilar	28.69	17.76	52	~ 13	Urban industrial	SO_2, PM_{10}, $PM_{2.5}$	CIG AQMN
Las Balsas	28.81	17.78	364	~ 23	Rural background	PM_{10}	CIG AQMN
Tenerife							
Izaña Observatory	28.31	16.50	2373	~ 140	High-mountain, remote background	SO_2	IARC-AEMET
Santa Cruz Observatory	28.47	16.24	52	~ 160	Urban background	Lidar profiles	IARC-AEMET
El Rio	28.15	16.52	500	~ 140	Regional background	PM_{10}, $PM_{2.5}$	CIG AQMN
Güímar station	28.32	16.38	105	~ 150	Radiosonde Station	Radiosonde vertical profiles of pressure, temperature & relative humidity	AEMET

Canary Islands Government (CIG), Air Quality Monitoring Network (AQMN), Izaña Atmospheric Research Centre (IARC), State Meteorological Agency of Spain (AEMET).
[a] Measurement sites have more data available but not employed in this study.
[b] Temporary measurement sites deployed to monitor eruption during emergency response.

which performs high-quality, long-term atmospheric composition measurements (Cuevas et al., 2022).

Measurements of SO_2 concentrations were conducted using ultraviolet (UV) fluorescence analysers at El Paso (Teledyne API T100) and Los Llanos (Teledyne API T100 until 14/12/2021 and Thermo 43i from 14/12/2021 onwards). Measurements of PM_{10} and $PM_{2.5}$ were conducted at Los Llanos with an Optical Particle Sizer (OPS) (Palas FIDAS 200, Karlsruhe, Germany), an aerosol spectrometer developed specifically for regulatory air pollution control, and in El Paso and Tazacorte, measurements were conducted from 20/11/2021 onwards with an OPS within the air quality instrument (Palas AQ Guard Smart, Germany). The measurements were conducted following European reference methods: EN-14212:2013/AC:2014 for SO_2 and EN-16450:2017 for PM_{10} and $PM_{2.5}$. In Tazacorte and Izaña Observatory, SO_2 concentrations were also measured with UV fluorescence analysers, Thermo 43C and Thermo 43C-TL (Trace Level), respectively. During the volcanic eruption, the calibration of the analyser in Tazacorte was verified against the same secondary standard used by the SO_2 analysers of El Paso and Los Llanos. For further details of measurement protocols utilised in Izaña Observatory see González (2012) and Cuevas et al. (2022).

2.3. Meteorological parameters: observations and numerical modelling

Ground-based meteorological data was obtained from the El Paso AEMET automatic meteorological station (C126A), which is situated at a distance of 2.7 km from the El Paso air quality station (Fig. 1c). Data included hourly measurements of wind speed and wind direction at 10 m a.g.l., air temperature at 2 m a.g.l. and accumulated precipitation, measurements are conducted following AEMET specifications and WMO guidelines (WMO, 2018).

Radiosonde observational data of vertical profiles of atmospheric pressure, temperature, and relative humidity was obtained from the AEMET automatic radiosonde station in Güímar, Tenerife (Global Climate Observing System, Upper-air Network station 60018). The radiosondes (Vaisala, RS41, Finland) are launched daily at 00:00 and 12:00 UTC, we used the values at 12:00 UTC in this study. The trade wind inversion (TWI) layer base height was derived from these radiosonde observations following Carrillo et al. (2016).

Daily vertical profiles of wind speed and direction have been retrieved from the AEMET numerical weather prediction (NWP) model HARMONIE-AROME (Hirlam Aladin Regional/Meso-scale Operational NWP in Euromed) Version 43 h2.1.1 (Bengtsson et al., 2017). The spatial resolution of the model is 2.5 × 2.5 km^2, with 65 vertical levels and a temporal output of 3 h and forecasts until 72 h. The forecast sounding of the grid point closest to the eruptive centre has been used.

As an additional tool, forward trajectories were computed with the FLEXible TRAjectories model (FLEXTRA) (Stohl et al., 1995; Stohl and Seibert, 1998) using ERA5 reanalysis from the European Centre for Medium-Range Weather Forecasts (ECMWF) (Hersbach et al., 2020) obtained through flex extract (v7.1) interface running in gateway mode as an ECMWF member-state user. FLEXTRA input data every 1 h were extracted to a horizontal resolution of 0.28125° x 0.28125°, covering a geographical domain between 18° N to 36° N latitude and 27° W to 0° longitude, with 137 vertical levels. FLEXTRA was configured with the starting point at the eruptive centre with coordinates 28.61° N, 17.87°W, and at pressure levels of 950, 900, 800, 700, 600 and 500 hPa. We obtained a forward trajectory coordinate every 20 min with a 3D wind configuration.

2.4. Auxiliary data

2.4.1. Satellite derived volcanic SO_2 emissions

Daily volcanic SO_2 emissions were obtained from the TROPOspheric Monitoring Instrument (TROPOMI), onboard the Copernicus Sentinel-5 Precursor (S-5P) platform, which provides complete daily global coverage of key atmospheric air quality and climate components. TROPOMI (launched in October 2017) is a push-broom imaging spectrometer measuring earthshine radiances at eight spectral bands covering from ultraviolet to shortwave infrared wavelengths (Theys et al., 2017), with a high spectral and unprecedented spatial resolution (5.5 × 3.5 km^2 at nadir since August 2019). Here, the daily SO_2 emissions for the 2021 La Palma eruption were obtained from the volcano monitoring platform MOUNTS (Monitoring Unrest from Space; www.mounts-project.com) (for further details of MOUNTS see Valade et al., 2019). MOUNTS downloads the TROPOMI level 2 near-real-time products and converts the vertical column densities of pixels (a 500 × 500 km frame) into SO_2 mass. The daily SO_2 emission corresponds to the sum of the masses of the pixels flagged as SO_2 contaminated (L2-product flag "sulfurdioxide_detection_flag", Romahn et al., 2022).

2.4.2. Lidar aerosol vertical distribution observations

A micro-pulse lidar (MPL-4B) was installed on 15/10/2021 in the Tazacorte measurement site as part of the emergency deployment to characterise the volcanic eruption (for further details see ACTRIS, 2021;

Sicard et al., 2022). In addition, the lidar system was used to identify Saharan dust episodes during the volcanic eruption. Lidars are considered the most efficient technique for characterization and continuous monitoring of aerosol vertical distribution, especially if polarization lidar options are available (Ansmann et al., 2012). This technique has been used in previous studies to characterise the evolution of the vertical and seasonal dust distribution in the subtropical North Atlantic region (Berjón et al., 2019; Barreto et al., 2022a). The lidar system in Tazacorte measurement site was used from 15/10/2021 onwards, while before this date, the MPL-4B system operating in Santa Cruz de Tenerife Observatory (SCO) was used. These MPLs are eye-safe elastic lidar operating at 532 nm with depolarization capability, and are the standard lidar system within the global NASA Micro-Pulse Lidar Network (MPLNET, https://mplnet.gsfc.nasa.gov) (Welton and Campbell, 2002). See Cuevas et al. (2022) for more details of the lidar system in Santa Cruz de Tenerife.

2.4.3. Tephra deposition measurements

Fresh tephra samples deposited from the eruptive plume were collected in plastic trays covered with paper sheets over measured time intervals. A permanent trap was installed from 20/10/2021 at the IARC-AEMET Tazacorte station, for 24-48 h collection of tephra samples. Additional traps were sporadically set for shorter durations (5–300 min) at different locations in the Aridane Valley between 1 and 8 km from the vents. The sporadic measurements in the Aridane Valley represent "instantaneous" rates, they do not integrate the accumulation of a whole day or several days as in the measurements in Tazacorte station. The tephra samples were stored in air tight plastic bags, later dried (for 24 h at 50 °C) and weighed with a precision balance (Mettler Toledo XS105) in the IZO laboratory.

3. Results and discussion

3.1. Air quality surface concentrations

3.1.1. SO_2 concentrations

The temporal evolution of SO_2 surface concentrations measured at the three measurement stations in the Aridane Valley in La Palma are presented for the nearly three months of eruption (19/9/2021–13/12/2021) and for three months post-eruption (14/12/2021–15/03/2022) (Fig. 2a-c), clearly demonstrating that the SO_2 concentrations were strongly impacted by the volcanic eruption, with values well above the typical long-term background values. The measurements also demonstrate that the SO_2 surface concentrations in La Palma generally increased as the volcanic eruption progressed.

During the La Palma volcanic eruption, mean hourly SO_2 surface concentrations at El Paso, Los Llanos and Tazacorte in La Palma were 40 ± 126 (Standard Deviation (SD)), 95 ± 133 and 56 ± 103 μg m^{-3}, respectively (Fig. 2a-c, Table 2). Maximum hourly mean SO_2 concentrations at El Paso, Los Llanos and Tazacorte were 2593, 1677 and 1623 μg m^{-3}, respectively. SO_2 concentrations exceeded the EC air quality hourly threshold of 350 μg m^{-3} at El Paso, Los Llanos and Tazacorte for a total of 24 h (1.2 % of the eruption period), 82 h (4.0 %) and 37 h (1.8 %), respectively (Table 2). Filonchyk et al. (2022) also report SO_2 and PM concentrations during the La Palma 2021 eruption, covering the first month of the eruption (22/9/2021–17/10/2021) and reported that SO_2 hourly concentrations in this first month often exceeded 400 μg m^{-3}.

Mean and median hourly SO_2 concentrations at Izaña Observatory in Tenerife, ~140 km from the volcanic eruptive centre, during the volcanic eruption, were 33 ± 296 μg m^{-3} and 0.33 μg m^{-3}, respectively (Fig. 2d). SO_2 concentrations at the Izaña Observatory FT background site

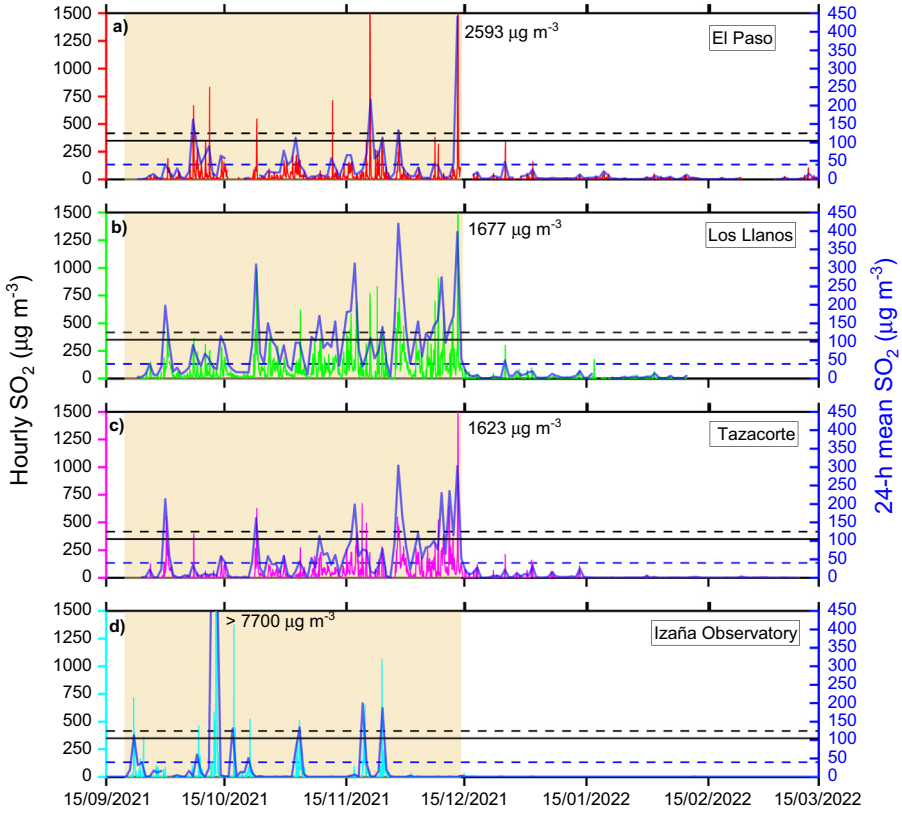

Fig. 2. Concentrations of SO_2 at three measurement sites in La Palma: a) El Paso, b) Los Llanos and c) Tazacorte and d) Izaña Observatory, the free troposphere site in Tenerife (15/9/2021–15/3/2022). The left axis indicates hourly mean concentrations and the right axis daily (24-h) mean concentrations. The black horizontal and dashed lines show the European Commission (EC) hourly (350 μg m^{-3}) and daily (24-h mean) (125 μg m^{-3}) air quality thresholds, respectively. The blue dashed line shows the WHO 2021 24-h mean SO_2 air quality guideline (40 μg m^{-3}). The shaded area marks the volcanic eruption period (19/9/2021–13/12/2021).

Table 2
Impact of the 2021 volcanic eruption on hourly mean concentrations of SO_2, PM_{10} and $PM_{2.5}$ in Aridane Valley (La Palma) measurement stations and Izaña Observatory (IZO) (Tenerife), 19/9/2021–13/12/2021.

Parameter	Mean (μg m^{-3})	SD (μg m^{-3})	Max (μg m^{-3})	Exceedance air quality thresholds and guidelines		
				EC threshold h > 350 μg m^{-3}	EC threshold 24-h > 125 μg m^{-3}	WHO 2021 guideline 24-h > 40 μg m^{-3}
SO_2						
El Paso	40	126	2593	24 (1.2 %)	4 (5 %)	23 (27 %)
Los Llanos	95	133	1677	82 (4.0 %)	20 (24 %)	55 (65 %)
Tazacorte	56	103	1623	37 (1.8 %)	8 (9 %)	38 (45 %)
IZO	33	296	> 7700	28 (1.4 %)	6 (7 %)	10 (12 %)
				EC threshold 24-h > 50 μg m^{-3}	WHO 2021 guideline 24-h > 45 μg m^{-3}	
PM_{10}						
Los Llanos	77	144	2836	38 (45 %)	48 (56 %)	
					WHO 2005 guideline 24-h > 25 μg m^{-3}	WHO 2021 guideline 24-h > 15 μg m^{-3}
$PM_{2.5}$						
Los Llanos	20	22	399		20 (24 %)	44 (52 %)

Measurements started on 23/9/2021 in El Paso and Los Llanos and on 24/9/2021 in Tazacorte, percentages calculated for 85 days of eruption. Standard Deviation (SD).

are normally very low, in the region of 0.2 μg m^{-3} (Cuevas et al., 2022). Maximum hourly mean SO_2 concentrations at IZO exceeded the maximum detection range of the analyser at 7700 μg m^{-3} on 12/10/2021 at 23:00 UTC (the maximum detection range was exceeded for 90 min in total). SO_2 concentrations exceeded the EC hourly threshold of 350 μg m^{-3} in IZO for a total of 28 h (1.4 % of the eruption period).

The impact of the volcanic eruption on SO_2 concentrations at Izaña (> 7700 μg m^{-3} at 140 km distance) is in the range of recorded air quality impacts of the 2014–2015 flood lava eruption at Bárðarbunga (Iceland), one of the largest tropospheric volcanic eruptions worldwide in terms of SO_2 emissions. Total SO_2 emissions over the 6-month Bárðarbunga eruption were estimated between 11 Tg (Gíslason et al., 2015) and 19 Tg (Gauthier et al., 2016). Maximum hourly SO_2 concentrations of 3050 μg m^{-3} at ~80 km distance were reported by Gíslason et al. (2015); 1509 μg m^{-3} at 123 km, 1451 μg m^{-3} at 86 km and 247 μg m^{-3} at 2755 km distance by Schmidt et al. (2015) and 1700 μg m^{-3} at 100 km distance by Ilyinskaya et al. (2017).

With respect to daily (24-h) values, mean SO_2 concentrations during the volcanic eruption period at El Paso, Los Llanos, Tazacorte and IZO were 39 ± 61, 93 ± 85, 55 ± 68 and 33 ± 118 μg m^{-3}, respectively (Table 3). Maximum 24-h mean SO_2 concentrations at El Paso, Los Llanos and Tazacorte were 439, 420 and 304 μg m^{-3}, respectively (Fig. 2a-c). Maximum 24-h mean SO_2 concentrations at IZO exceeded 800 μg m^{-3} (Fig. 2d), an exact maximum 24-h value cannot be given because the SO_2 concentration exceeded the maximum detection range of the analyser. In comparison, Whitty et al. (2020) reported maximum 24-h mean SO_2 values of 728 μg m^{-3} at a distance of 100 km during the May–August 2018 eruption of Kılauea Volcano, Hawaii.

SO_2 concentrations exceeded the EC 24-h mean air quality threshold of 125 μg m^{-3} at El Paso, Los Llanos and Tazacorte for a total of 4 days (5 % of days of the eruption period), 20 days (24 %) and 8 days (9 %), respectively. In IZO the EC threshold was exceeded for a total of 6 days (7 % of days of the eruption period). It is striking that the maximum hourly mean SO_2 concentration recorded at IZO, in Tenerife, on 12/10/2021 at 23:00 UTC (> 7700 μg m^{-3}) was three times higher than the maximum hourly mean SO_2 concentration recorded in the three measurement stations in the Aridane Valley in La Palma (2593 μg m^{-3}) and the maximum 24-h mean SO_2 at IZO also surpassed the La Palma values. This is due to the nearly unperturbed volcanic plume being transported in the similar height range as Izaña Observatory in Tenerife (~ 3 km a.s.l.) and therefore directly impacting the troposphere and the observatory at this altitude, particularly in the first phase of the eruption. In contrast, the measurement stations in the Aridane Valley in La Palma that we utilise in this study were not typically impacted directly by the volcanic plume, as they are located at altitudes lower than the volcanic eruptive centre and are also not downwind of the predominant wind direction during the 2021 eruption (see Section 3.3.4 for more details).

SO_2 concentrations exceeded the more stringent WHO 2021 24-h mean AQG of 40 μg m^{-3} at El Paso for 23 days (27 % of eruption period), at Los Llanos for 55 days (65 %), in Tazacorte for 38 days (45 %) and in IZO for

Table 3
Impact of the 2021 volcanic eruption on 24-h mean concentrations of SO_2, PM_{10} and $PM_{2.5}$ in Aridane Valley (La Palma) measurement stations and Izaña Observatory (IZO) (Tenerife), 19/9/2021–13/12/2021.

Parameter	Background[a]		Eruption		Volcanic enhancement	
	Mean ± SD (μg m^{-3})	Max (μg m^{-3})	Mean ± SD (μg m^{-3})	Max (μg m^{-3})	Mean ± SD (μg m^{-3})	Max (μg m^{-3})
SO_2 (El Paso)	2.7 ± 3.5	16	39 ± 61	439	36 ± 61	436
SO_2 (Los Llanos)	4.2 ± 4.4	14	93 ± 85	420	89 ± 85	416
SO_2 (Tazacorte)	0.6 ± 0.7	3.0	56 ± 68	304	55 ± 68	304
SO_2 (IZO)	0.23 ± 0.04	0.3	33 ± 118	> 800	32 ± 117	>800
PM_{10} (Los Llanos)	16 ± 12	72	76 ± 72	382	61 ± 70	367
$PM_{2.5}$ Los Llanos	7 ± 4	22	20 ± 13	58	14 ± 11	52

[a] Background data for SO_2: El Paso and Tazacorte, 15/02–15/03/2022 (post-eruption), for Los Llanos, 1–8/2/2022 as measurements end on this date and for IZO, 1–18/9/2021 (pre-eruption). Background (non-volcanic) concentrations of PM_{10} and $PM_{2.5}$ taken from San Antonio measurement site during eruption, 19/9/2021–13/12/2021. Standard Deviation (SD).

10 days (12 %). Here we note there were more days of the WHO SO_2 AQG exceeded in all three measurement sites of La Palma than in IZO. Although the maximum peak SO_2 concentration in IZO (Tenerife) was larger, the duration and frequency of peak values at IZO were less compared to in La Palma (see Fig. 2), which explains the greater exceedance of the WHO SO_2 AQG values in La Palma. Furthermore, we observe that the last major peak of SO_2 recorded at IZO during the eruption period, was on 24/11/2021 (24-h mean: 187 μg m^{-3}) (Fig. 2d). After this date, in the latter 18 days of the volcanic eruption, there were only a few, much smaller, briefer spikes in SO_2 concentration registered at IZO. However, in this latter period of the volcanic eruption in La Palma we observe how the SO_2 24-h mean concentrations, particularly in Los Llanos and Tazacorte, show a trend of increasing concentrations with persistent elevated values (largely >40 μg m^{-3}) (Fig. 2b and c). In summary, the SO_2 concentration data, both near to the eruptive centre in La Palma, and further away in the free troposphere in Tenerife, highlight the strong impact of the La Palma 2021 volcanic eruption and its effect on SO_2 ambient concentrations.

3.1.2. PM surface concentrations

The temporal evolution of PM_{10} and $PM_{2.5}$ surface concentrations measured in the Aridane Valley in La Palma during the volcanic eruption (19/9/2021–13/12/2021) is presented in Fig. 3a and b, demonstrating the high variability in PM concentrations and the high hourly and daily maximums that were reached. Of the three sites set up to monitor the volcanic eruption in the Aridane Valley, on the western side of La Palma, measurements of PM_{10} and $PM_{2.5}$ were only conducted for the whole eruption period at the Los Llanos measurement site. During the volcanic eruption period, mean hourly PM_{10} and $PM_{2.5}$ concentrations at Los Llanos were 77 ± 144 and 20 ± 22 μg m^{-3}, respectively (Table 2). Maximum hourly mean PM_{10} and $PM_{2.5}$ concentrations at Los Llanos were 2836 and 399 μg m^{-3}, respectively. Regarding daily values, mean 24-h PM_{10} and $PM_{2.5}$ concentrations during the volcanic eruption period at Los Llanos were 76 ± 72 μg m^{-3} and 20 ± 13 μg m^{-3}, respectively (Table 3). Maximum 24-h mean PM_{10} and $PM_{2.5}$ concentrations at Los Llanos were 382 μg m^{-3} (Fig. 3a) and 58 μg m^{-3} (Fig. 3b), respectively. For comparison, Whitty et al. (2020) reported maximum 24-h mean $PM_{2.5}$ values of 59 μg m^{-3} at a distance of 106 km during the May–August 2018 eruption of Kılauea Volcano, Hawaii. PM_{10} concentrations at Los Llanos exceeded the EC 24-h mean air quality threshold (50 μg m^{-3}) for 38 days (45 % of eruption period) and the WHO 2021 24-h mean PM_{10} AQG (45 μg m^{-3}) for 48 days (56 %) (Fig. 3a). $PM_{2.5}$ concentrations at Los Llanos exceeded the WHO 24-h mean PM2.5 2005 AQG (25 μg m^{-3}) and 2021 AQG (15 μg m^{-3}) for 20 days (24 % of eruption period) and 44 days (52 %), respectively (Table 2).

It is important to highlight that in the case of SO_2, the dominant source of SO_2 emission in the study period in La Palma was the volcanic eruption. However, this is not the case for PM_{10} and $PM_{2.5}$ concentrations, which were a net result of all contributing PM sources during the eruption. There were multiple sources of PM emissions in La Palma during the eruption: there were substantial primary PM emissions (ash particles among the tephra emitted); other PM emissions which arose indirectly from the volcanic eruption, from e.g. fires as the lava advanced on buildings, banana plantations and other vegetation; the formation of secondary particulates (e.g. sulphates), resuspension of volcanic PM and other aerosols produced when the lava flow came into contact with the ocean. In addition to volcanic PM, there were other anthropogenic sources of PM in La Palma such as on-road traffic emissions and there was also the contribution of mineral dust during Saharan air mass episodes.

Lidar vertical profiles at SCO, back trajectory information and reanalysis data were utilised to identify the Saharan air mass episodes that occurred during the volcanic eruption period, in order to examine the contribution of mineral Saharan dust to the PM concentrations. Three Saharan air mass episodes (covering 13 days in total) affecting La Palma during the 85 days of the volcanic eruption, were identified (see Fig. 3 and Fig. S1). The SAL episodes occurred during the following dates: 1) 26/9–3/10/2021, 2) 7–8/10/2021 and 3) 19–21/10/2021. These dust events have been observed to be clearly confined between ~ 1–7 km a.s.l. during the first episode and up to 4 km a.s.l. during the following two episodes, in agreement with the expected seasonal evolution of the SAL vertical extension shown by Barreto et al. (2022a). Median lidar ratio (one-layer method) ranged between 26 and 61 sr, in agreement with results published by Berjón et al. (2019). Polarization measurements provide an important piece of information to characterise the shape of the backscattering particles, ranging from near-zero Volume Depolarization Ratio (VDR) values in the case of spherical particles to the highest VDR values found for non-spherical particles (Ansmann et al., 2012), and consequently provide useful information about the aerosol type. Typical values for VDR and Particle Depolarization Ratio (PDR) for desert dust are given by Freudenthaler et al. (2009) and Ansmann et al. (2012), and references therein. We therefore expect VDR values for desert dust close to 0.20 with PDR values ranging between 0.30 and 0.35. However, higher PDR values are expected for volcanic ash by these authors (0.34), based on 2–3 day transported volcanic plumes, and importantly, lower PDR (0.02) for fine-mode aerosols (such as those sulphates originating from volcanic SO_2 emissions). Fresh volcanic aerosols could present lower PDR values due to a balanced mixing stage

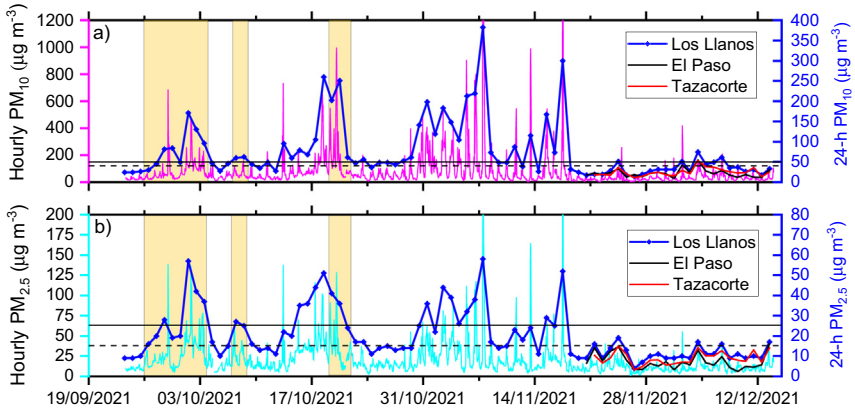

Fig. 3. Concentrations of a) PM_{10} and b) $PM_{2.5}$ at Los Llanos, El Paso and Tazacorte, 19/9/2021–13/12/2021. The left axis indicates hourly mean concentrations and the right axis daily (24-h) mean concentrations. Measurements in El Paso and Tazacorte were only available from 20/11/2021 onwards. The black horizontal and dashed lines in the top panel show the European Commission (EC) 24-h mean PM_{10} air quality threshold (50 μg m^{-3}) and the WHO 2021 24-h mean PM_{10} air quality guideline (45 μg m^{-3}), respectively. The black horizontal and dashed lines in the lower panel show the WHO 2005 24-h mean $PM_{2.5}$ (25 μg m^{-3}) and 2021 (15 μg m^{-3}) air quality guidelines, respectively. The shaded areas mark the Saharan Air Layer episodes affecting La Palma during the eruption period.

between coarse and fine volcanic particles. Indeed, lidar measurements carried out close to the Mount Etna volcano reported a wide range of PDR values (0.16–0.45) (Pisani et al., 2012; Scollo et al., 2012). This is the case of the PDR mean maximum daily values of 0.26 ± 0.08 obtained during the La Palma volcanic eruption by Córdoba-Jabonero et al. (2023). We observe how the three SAL air mass episodes in September and October 2021 coincide with increasing PM surface concentrations (Fig. 3). However, the maximum PM concentrations observed during the volcanic eruption in November 2021 did not coincide with a SAL episode, therefore these high PM concentrations are attributed to volcanic aerosols.

3.2. Volcanic enhancement of surface SO_2 and PM concentrations

In this section, we estimate the volcanic "enhancement" or increment in surface concentrations attributed to the volcanic eruption, in order to further quantify the impact of the La Palma volcanic eruption on air quality levels.

3.2.1. SO_2 concentrations

On the eastern side of La Palma, there were SO_2 measurements prior to the eruption at the permanent air quality monitoring stations in the Canary Islands Air Quality Monitoring Network, therefore to give some context on the volcanic enhancement we examined the SO_2 concentrations in the pre-eruption period. Annual mean SO_2 concentrations at two of the permanent air quality monitoring stations, San Antonio and El Pilar, prior to the volcanic eruption, ranged between 3 and 5 μg m^{-3} in 2019–2020 (CIG, 2020, 2021). There were no exceedances of the EC hourly or daily SO_2 limit values at the permanent air quality monitoring stations in La Palma in the years prior to the 2021 volcanic eruption.

The volcanic enhancement of surface SO_2 concentrations (ΔSO_2) during the eruption was estimated by subtracting the background concentrations for each measurement site. The mean background SO_2 concentration of the post eruption period in La Palma (mid-February-mid March 2022) was subtracted from the daily (24-h) mean SO_2 concentrations during the volcanic eruption to obtain the daily values of ΔSO_2 (Table 3). We utilised the post-eruption period of mid-February-mid March 2022 as in the preceding two months (mid December 2021-mid February 2022), although the volcanic eruption had ceased, there was still some degassing from the volcanic vents which was detected by the La Palma measurement sites as elevated SO_2 concentrations (see Fig. 2a-c).

As expected, we observe a strong volcanic enhancement on both mean and maximum SO_2 concentrations measured during the volcanic eruption. The background SO_2 concentrations are low, at the three measurement stations in the Aridane Valley in La Palma, ranging between 0.6 and 4.2 μg m^{-3} (Table 3) and confirm that there is very little contribution from local sources of SO_2 (which include a power plant and the Santa Cruz de La Palma harbour on the eastern side of La Palma). The dominant source of SO_2 emission in the study period in La Palma was the volcanic eruption, the volcanic SO_2 emissions dominate any small contribution from SO_2 anthropogenic emissions. Values of ΔSO_2 (volcanic enhancement of SO_2) as a mean 24-h value in La Palma range between 36 μg m^{-3} in El Paso to 89 μg m^{-3} in Los Llanos (Table 3). The mean ΔSO_2 concentrations are up to 93 times larger compared to background concentrations in La Palma and maximum ΔSO_2 24-h concentrations also increase by up to 2 orders of magnitude (e.g. in Tazacorte the maximum background concentration is 3 μg m^{-3} compared to maximum ΔSO_2 of 304 μg m^{-3}, Table 3). In Izaña Observatory, where background SO_2 concentrations are very low (0.23 ± 0.04 μg m^{-3}, Table 3), representative of the free troposphere and far from the influence of anthropogenic sources, the volcanic enhancement is even greater, with the mean ΔSO_2 (32 μg m^{-3}) and maximum ΔSO_2 (> 800 μg m^{-3}) concentrations being >2 and 3 orders of magnitude greater than the background concentrations, respectively.

3.2.2. PM concentrations

Estimating the volcanic enhancement of surface SO_2 concentrations during the La Palma volcanic eruption is quite straightforward as atmospheric background concentrations of SO_2 are low in this region. Particulate matter in this subtropical eastern North Atlantic region, however, has a larger atmospheric background and demonstrates high variability and therefore the volcanic enhancement needs to be discriminated from this variable background concentration. This challenge for PM is similar to the challenge in deriving volcanic enhancements for CO_2 as described in Butz et al. (2017). In this region, concentrations of PM_{10} and $PM_{2.5}$ in the absence of Saharan dust events, are usually low, with typical 24-h mean values of ~20 and ~ 10 μg m^{-3}, respectively (Dominguez-Rodriguez et al., 2020), but concentrations of PM_{10} and $PM_{2.5}$ increase from tens to hundreds (μg m^{-3}) during Saharan dust outbreaks, depending on the severity of the outbreak (see Fig. S2).

To take account of this variability in the PM background, we utilised the PM_{10} and $PM_{2.5}$ concentrations at the San Antonio suburban background measurement site on the eastern side of La Palma as representative of the daily background or "non-volcanic" concentrations. These non-volcanic concentrations include other contributing PM sources during the eruption such as any contribution of mineral dust during Saharan air mass episodes.

We obtained the daily net volcanic enhancement (ΔPM) in PM_{10} and $PM_{2.5}$ at Los Llanos measurement site, by subtracting the daily background (non-volcanic) concentration at San Antonio from the PM_{10} and $PM_{2.5}$ concentrations in Los Llanos. Only the enhancement at the Los Llanos station is evaluated because its measurement series covers the entire volcanic eruption and the other stations do not. The choice of San Antonio for the background site provides a conservative estimate of the volcanic eruption enhancement as there were days when the San Antonio measurement site was also affected to some degree by the volcanic eruption. However, on examining the data, there were only four days (20–23/11/2021) when the PM_{10} and $PM_{2.5}$ concentrations at the San Antonio measurement site were affected to a greater degree by the volcanic eruption than the Los Llanos site.

As an additional check on the representativeness of the background (non-volcanic) concentration, we also utilised the PM_{10} and $PM_{2.5}$ concentrations at the El Rio regional background measurement site in Tenerife. This measurement site has been used for various studies and characterisations of Saharan air mass episodes in the Canary Islands (Viana et al., 2002; Alonso-Pérez et al., 2007; Milford et al., 2020) and is also the measurement site utilised to quantify the contribution of natural dust to daily PM concentrations during dust episodes in the Canary Islands at a national level (e.g. MITECO, 2021). The PM_{10} and $PM_{2.5}$ concentrations at San Antonio and El Rio measurement site are very similar (see Fig. S2), confirming the San Antonio concentration series as representative of the background PM variability in this region during the volcanic eruption.

We observe a strong volcanic enhancement on both mean and maximum PM_{10} and $PM_{2.5}$ concentrations measured during the volcanic eruption (Fig. 4). Background or "non-volcanic" PM_{10} and $PM_{2.5}$ concentrations in La Palma during the eruption are 16 ± 12 μg m^{-3} and 7 ± 4 μg m^{-3}, respectively (Table 3). Values of ΔPM_{10} and $\Delta PM_{2.5}$ as a mean 24-h value in La Palma are 61 ± 70 μg m^{-3} and 14 ± 11 μg m^{-3}, respectively. The maximum 24-h value of ΔPM_{10} and ΔPM2.5 in La Palma are 367 μg m^{-3} and 52 μg m^{-3}, respectively. The mean and maximum ΔPM_{10} are 4 and 5 times the background concentrations, respectively, while the mean and maximum $\Delta PM_{2.5}$ concentrations are twice the background concentrations. The volcanic enhancement relative to background concentrations is larger in the PM_{10} fraction than the $PM_{2.5}$ fraction.

Although the volcanic enhancement in PM_{10} and $PM_{2.5}$ concentrations is substantial, the enhancement in PM in the subtropical eastern North Atlantic region from Saharan air mass episodes can be larger than the observed volcano enhancement, in terms of PM surface concentrations. This is demonstrated by measurements in January 2022 where there were two very intense Saharan air mass episodes with daily mean (24-h) PM_{10} peaking at Los Llanos and San Antonio at 560 and 647 μg m^{-3}, respectively, on 16/1/2022, and at 431 and 472 μg m^{-3}, respectively on 30/1/2022 (Fig. S2). Daily mean (24-h) $PM_{2.5}$ peaked at Los Llanos and San Antonio at 142 and 155 μg m^{-3}, respectively, on 16/1/2022, and at 116 and 113 μg m^{-3}, respectively on 30/1/2022. In winter and early

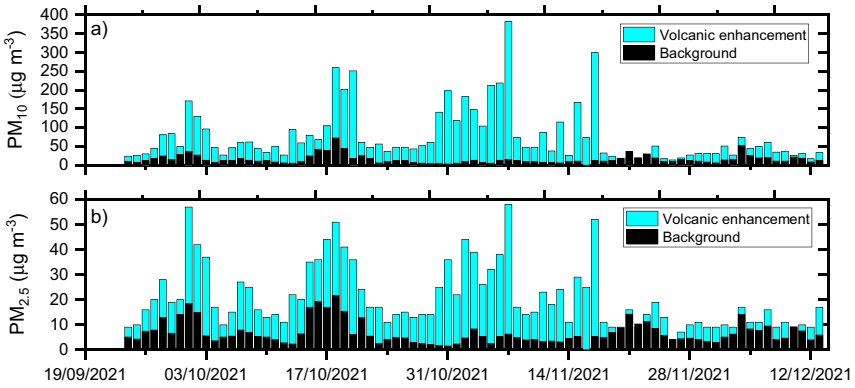

Fig. 4. Daily (24-h mean) volcanic enhancement in Los Llanos and background (non-volcanic) concentrations at San Antonio (La Palma-suburban background) of a) PM_{10} and b) $PM_{2.5}$ (19/9/2021–13/12/2021).

spring, the Saharan air mass episodes over the subtropical eastern North Atlantic are usually confined to low altitudes (< 2 km a.s.l.) while in summer, the SAL and dust transport occurs at higher altitudes (~ 1–6 km a.s.l.) (Barreto et al., 2022a,b). The dust episodes in wintertime in this region are more intense in terms of surface PM concentrations than those in summer (Viana et al., 2002); for example, Milford et al. (2020) observed an increment of 84 and 26 μg m^{-3} in PM_{10} and $PM_{2.5}$ mean (24-h) concentrations during winter intense dust outbreaks and 35 and 13 μg m^{-3} in summer intense dust outbreaks in this region.

The volcanic PM enhancement was substantial and although the desert dust PM enhancement in the subtropical eastern North Atlantic region can be larger, the duration of the volcanic eruption (85 days), was much longer than the typical Saharan dust episode duration (3–5 days).

3.3. Temporal evolution of SO_2 and PM concentrations: modulating factors

In this section, we examine the temporal evolution of SO_2 and PM concentrations during the 2021 La Palma volcanic eruption and explore the various factors, both volcanic and atmospheric, that modulate these concentrations. The temporal evolution of various parameters during the eruption period is presented in Fig. 5, these include SO_2 surface concentrations in La Palma (Fig. 5a), daily SO_2 volcanic emissions (according to satellite constrained estimates) (Fig. 5b), measurements of the volcanic eruptive column top height and Trade Wind Inversion (TWI) layer base height (Fig. 5c), PM_{10} and $PM_{2.5}$ surface concentrations in La Palma (Fig. 5d), tephra deposition (Fig. 5e) and the $SO_2/PM_{2.5}$ mass ratio (Fig. 5f).

3.3.1. Volcanic SO_2 emissions

The temporal evolution of SO_2 surface concentrations in La Palma during the eruption period demonstrates that the SO_2 concentrations generally increased as the volcanic eruption progressed (Fig. 5a), however, this is in contrast to the temporal trend of the SO_2 volcanic emissions (Fig. 5b). At the start of the volcanic eruption, the daily SO_2 volcanic emissions (according to satellite constrained estimates) were at their greatest, fluctuating in the range ~ 10–85 kt and reaching a maximum of 125 kt on 23/9/2021 (Fig. 5b).

As the eruption progressed, the SO_2 emission rates decreased, with a notable decline from approximately 7/11/2021 onwards. We have denoted this first period of the volcanic eruption with greater SO_2 emission rates, as Phase I. The daily mean and accumulated SO_2 volcanic emission during Phase I were 34 kt and 1.59 Mt., respectively. It is striking to observe the largest peaks in SO_2 surface concentrations in La Palma at the end of the eruption (Fig. 5a, Phase II) when the daily SO_2 emissions were at their lowest; the daily mean and accumulated SO_2 emission during Phase II were 7 kT and 0.25 Mt., respectively. Although Phase I is roughly 55 % of the eruption duration, the total SO_2 mass emitted during Phase I was >6 times larger than in Phase II.

Total SO_2 emitted from the La Palma 2021 volcanic eruption was estimated as ~ 1.84 Tg from satellite constrained estimates (TROPOMI) provided by the MOUNTS volcano monitoring platform (see Section 2.4.1 for more details), this is in agreement with the value provided by PEVOLCA of ~ 2 Tg (PEVOLCA, 2021). Carn et al. (2017) report ~ 23 ± 2 Tg yr^{-1} for global passive volcanic SO_2 emissions derived from satellite measurements for 2005–2015, with global eruptive volcanic emissions being more variable and ranging from ~ 0.2 to 10 Tg yr^{-1}. Bani et al. (2022) estimate the total volcanic SO_2 flux for Indonesian volcanoes as 1.27–1.69 Tg yr^{-1}. Therefore, the La Palma 2021 eruption emitted the equivalent to slightly more than one of the most volcanically active regions of the world.

These values demonstrate the substantial SO_2 emissions of the La Palma 2021 volcanic eruption and their importance on a global scale. Furthermore, the estimate of 1.84 Tg of SO_2 emitted during the nearly 3 months of the La Palma 2021 volcanic eruption exceeds the 1.66 Tg of anthropogenic SO_2 emitted in the 27 countries of the European Union throughout the whole of 2019 (EEA, 2021). The La Palma 2021 volcanic SO_2 emissions are >10 times the total anthropogenic SO_2 emissions reported for 2019 in Spain (0.167 Tg) (MITECO, 2022) and > 100 times the Canary Islands anthropogenic SO_2 emissions (0.016 Tg), evidencing the huge release of SO_2 into the atmosphere from the 2021 eruption and demonstrating how the volcanic emissions dominate the SO_2 anthropogenic emissions in this region.

3.3.2. Meteorological conditions

To explore the factors influencing the surface concentrations, measurements of the Trade Wind Inversion layer base height derived from radiosonde observations (following Carrillo et al., 2016), were utilised to characterise the vertical structure of the atmosphere during the La Palma 2021 volcanic eruption. The TWI height in the Canary Islands region shows a clear seasonal variation, with a minimum value in the summer months and reaching its maximum value in the winter months (Carrillo et al., 2016). The median TWI base height in the Canary Islands region in the summer months is 949 ± 18 m a.s.l. and in the winter months is 1522 ± 21 m a.s.l., with the inversion being thicker and stronger in summer (thickness = 308 ± 36 m a.s.l., ΔT = 4.6 ± 0.3 °C) than in winter (thickness = 210 ± 42 m a.s.l., ΔT = 2.7 ± 0.4 °C) (Carrillo et al., 2016).

During Phase I of the volcanic eruption, the TWI base height was mostly <2000 m (Fig. 5c), with a median value of 1221 m a.s.l., values typical for this season if we compare with the values given in Carrillo et al. (2016). Estimations of the eruptive column top height were obtained from the daily reports issued from the Scientific Committee of the Plan Especial de Protección Civil y Atención de Emergencias por Riesgo Volcánico en la Comunidad Autónoma de Canarias (PEVOLCA) (e.g. PEVOLCA, 2021), indicating that at the start of the eruption, the eruptive column top height reached 6000 m. Therefore, during this initial period of the eruption,

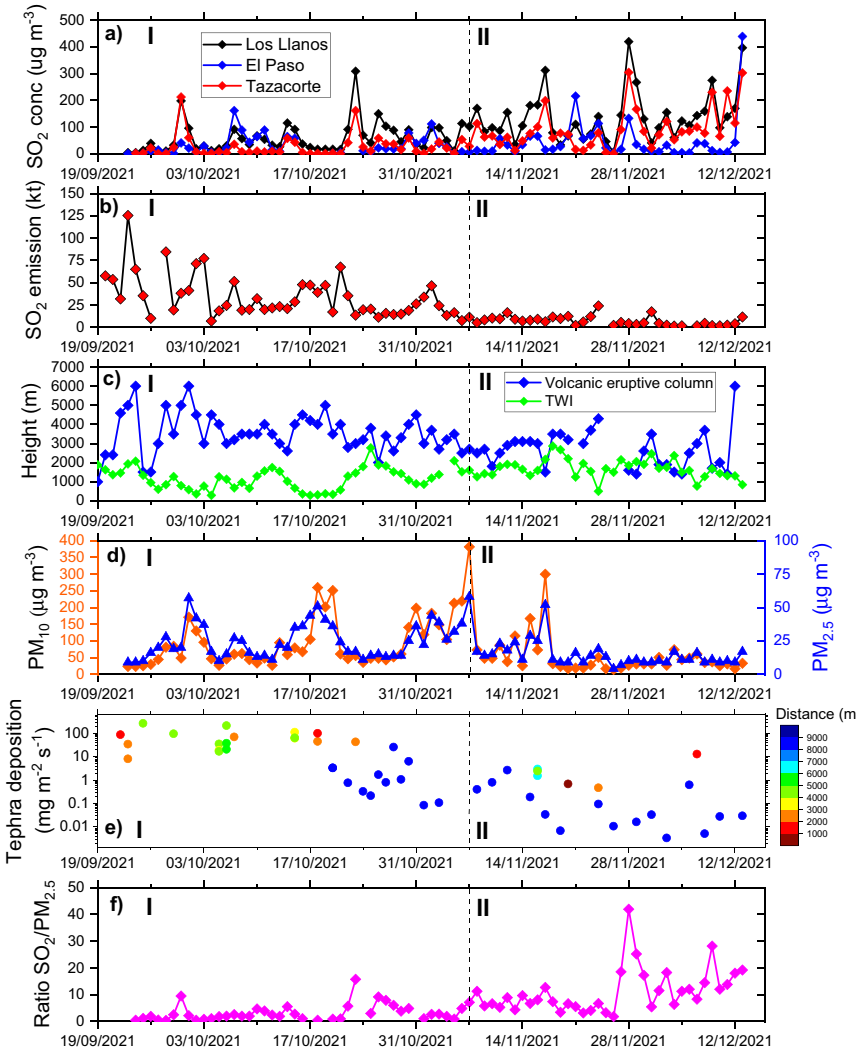

Fig. 5. Temporal evolution during La Palma volcanic eruption (19/9/2021–13/12/2021) of a) Daily (24-h mean) SO_2 concentrations in La Palma, b) Daily SO_2 volcanic emissions (kt) estimate provided by TROPOMI (credit: ESA, MOUNTS), c) Volcanic eruptive column top height and Trade Wind Inversion (TWI) base height (m a.s.l.). d) Daily (24-h mean) PM_{10} and $PM_{2.5}$ concentrations in Los Llanos. e) Tephra deposition (mg m^{-2} s^{-1}) recorded sporadically in various sites in the Aridane Valley and continuously in Tazacorte measurement site (24/10/2021–14/12/2021) (note logarithmic scale on the Y axis) and f) $SO_2/PM_{2.5}$ mass ratio. The vertical dashed line marks the date 7/11/2021 and separates the volcanic eruption into two phases (I and II).

the volcanic SO_2 emissions were likely injected largely into the free troposphere, above the MBL.

As the volcanic eruption progressed into the months of November and December 2021, the TWI base height increased, to values generally ranging from 1300 to 3000 m (Fig. 5c), with a median value of 1652 m a.s.l., values typical for this season. The volcanic eruptive column height also decreased notably as the eruption progressed and in the latter period of the eruption (Phase II), the eruptive column top height and the TWI base height were often at similar altitudes (Fig. 5c). Consequently, a larger proportion of the SO_2 emitted by the volcano in the latter stage of the eruption was now likely confined in the MBL rather than being emitted above the MBL into the free troposphere, as in the earlier stage of the eruption. This interplay of the volcanic emissions injection height and prevailing atmospheric conditions determines the impact of volcanic pollutants in the atmosphere, as also concluded by Thomas et al. (2017). These modulating factors lead to the SO_2 surface concentrations in La Palma increasing as the eruption processed (as seen in Fig. 5a) rather than decreasing in accordance with the decreasing emission fluxes (as seen in Fig. 5b). As a result of a larger proportion of the volcanic plume being confined to the MBL in the latter phase of the eruption (Phase II), it is in this phase that we observe the most adverse conditions (maximum hourly SO_2 concentrations experienced in La Palma reaching ~2600 µg m^{-3}, well above the typical long-term background values) and largest impacts for SO_2 surface air quality in La Palma. The majority of the SO_2 hourly and daily exceedances of air quality thresholds occurred during the MBL dominated Phase II, this is demonstrated in Fig. S3 which shows a time line of exceedances of air quality thresholds during the eruption.

3.3.3. Tephra deposition

The temporal evolution of PM concentrations is more complex than that of SO_2; there were multiple sources of PM emissions in La Palma during the eruption. The major source of volcanic PM is the ash plume. The tephra deposition measurements recorded sporadically and instantaneously in various sites in the Aridane Valley and continuously in Tazacorte measurement station (Fig. 5e) present a globally decreasing trend, similar to the trend of daily SO_2 emissions (Fig. 5b) and to that of the height of the plume (Fig. 5c), reflecting the progressively decreasing pressure in the system (Charco et al., 2022). The tephra deposition rate is generally several orders of magnitude higher during Phase I than during Phase II (Fig. 5e). This is clear from our record at the distal permanent Tazacorte station, although measurements only start on 24/10/2021. This is also apparent for the most proximal measurements, although they can include some intense shorter-term episodes

that are not necessarily maintained over the time scales of the permanent Tazacorte station experiments. The highest ash deposition rate of the deposition measurements presented in this study was 267 mg m^{-2} s^{-1}, recorded on 25/9/2021 in Phase I, whereas the largest ash deposition recorded in Phase II, in our measurements, was 13 mg m^{-2} s^{-1} on 16/11/2021. These tephra deposition rates compare with rates of 6–700 mg m^{-2} s^{-1} measured during the Eyjafjallajökull eruption (Bonadonna et al., 2011).

3.3.4. Volcanic plume transport

The injection height of volcanic gas and aerosol plumes is crucial for both successful forecasting of the plume transport for aviation purposes and for forecasting air quality impacts (Thomas et al., 2017). The subtropical eastern North Atlantic is characterised by a strong vertical stratification of the lower troposphere, which both impedes the vertical growth of the volcanic plume, and results in a typical change of wind direction with height, with a predominance of low-level north-easterly trade winds in the MBL and north-westerly winds above the inversion layer. For these reasons, the height at which the ash and SO_2 plumes are injected, and the atmospheric layer into which they were emitted, is crucial as this will determine the transport direction of the plumes. The constraint on the vertical growth of the volcanic plume, as well as its confinement in a relatively thin atmospheric layer, with a strong height dependent transport direction, are determining characteristics of the subtropical troposphere that differentiate it from mid latitude and tropical regions, which do not experience such a strong atmospheric vertical stratification.

Wind roses showing the frequency of wind direction during the entire volcanic eruption for both measurements at the surface (10 m a.g.l.) in El Paso meteorological station and for three increasing heights in the atmosphere, at the volcanic eruption location, from the HARMONIE-AROME model at 950 hPa (~ 0.6 km a.s.l.), 800 hPa (~ 2 km a.s.l.) and 700 hPa (~ 3 km a.s.l.) are presented in Fig. 6. Here we observe that measurements at the surface level in El Paso (Fig. 6a) show a dominance of easterly winds while modelled wind direction at 0.6 km a.s.l. (Fig. 6b) also show a E-NE dominance, rotating to a dominance of NE winds at 2 km a.s.l. (Fig. 6c). The modelled wind direction at 700 hPa, an altitude of ~ 3 km a.s.l., however, now shows a much wider set of wind directions, with both NE and SW directions as well as NW (Fig. 6d).

The wind rose diagrams demonstrate that in the lower levels of the troposphere during the La Palma 2021 eruption, the dominant wind direction was E-NE, this will have led to any ash and SO_2 plumes present at these altitudes being transported to the W-SW (that is towards the Aridane Valley and western slopes, see Fig. 1). Any plumes injected at higher levels of the troposphere, e.g. at or above 3 km, will have experienced a greater range of wind directions and been dispersed in various directions including to the NE and SE as well as to the SW.

Three example days illustrating these points are depicted in Fig. 7. The first example day is 12/10/2021, when the maximum SO_2 concentration was detected at Izaña Observatory. The image (Fig. 7a) clearly shows the plume being transported to the east of La Palma and FLEXTRA forward trajectories indicate SW transport at lower altitudes and easterly transport at higher altitudes (Fig. 7d). This is corroborated by satellite imagery of the SO_2 plume (Fig. 7g). The second example day is 13/10/2021 (Fig. 7b), where SW transport still dominates at lower altitudes but now the image clearly shows the volcanic plume being split and transported both eastwards and westwards and this is demonstrated by the westward transport of the forward trajectory at 700 hPa (~ 3 km) and eastward transport at 600 hPa (~ 4.3 km) and above. The third example day is 7/11/2021 (Fig. 7c), the day of maximum PM surface concentrations (see Fig. 5d). The image demonstrates that the volcanic plume height is now reduced and dispersion of the plume into the lower levels of the troposphere can be observed. The forward trajectories for 7/11/2021 show SW-W transport at all altitudes (Fig. 7f), the satellite imagery of the SO_2 plume corroborates this westerly transport (Fig. 7i).

Volume depolarization ratios from the micro-pulse lidar profiles in SCO and Tazacorte are presented in Fig. 7 for the same three example days, to corroborate the heights of the plumes in the different events and the type of aerosol. The profiles taken at SCO (Fig. 7j and k), show VDR values between 0.05 and 0.20 in a highly stratified volcanic plume, extending between 1.5 and 3 km. The lidar profile of 7/11/2021 in Tazacorte (Fig. 7l) shows slightly higher VDR values (up to 0.22) and also demonstrates that the height of the volcanic aerosol is now clearly lower, extending from the surface up to around 2 km height, and confined within the MBL.

The vertical stratification of the lower troposphere defined the transport of the La Palma 2021 eruption plume and it may well have been dispersed both within and above the MBL at times during the eruption. Any volcanic aerosol confined in the lower levels of the troposphere, according to the predominant W-SW transport at these altitudes, will have affected the Aridane Valley and western slopes of La Palma. These factors and the greater volcanic emission in the first phase of the volcanic eruption likely led to the higher PM concentrations observed in the earlier phase of the eruption. The three Saharan air mass episodes that were identified also occurred during Phase I of the volcanic eruption therefore this additional non-volcanic PM source increased the particulate load during this period.

The interplay of these influencing factors leads to the most adverse conditions (maximum hourly and daily PM_{10} and $PM_{2.5}$ concentrations)

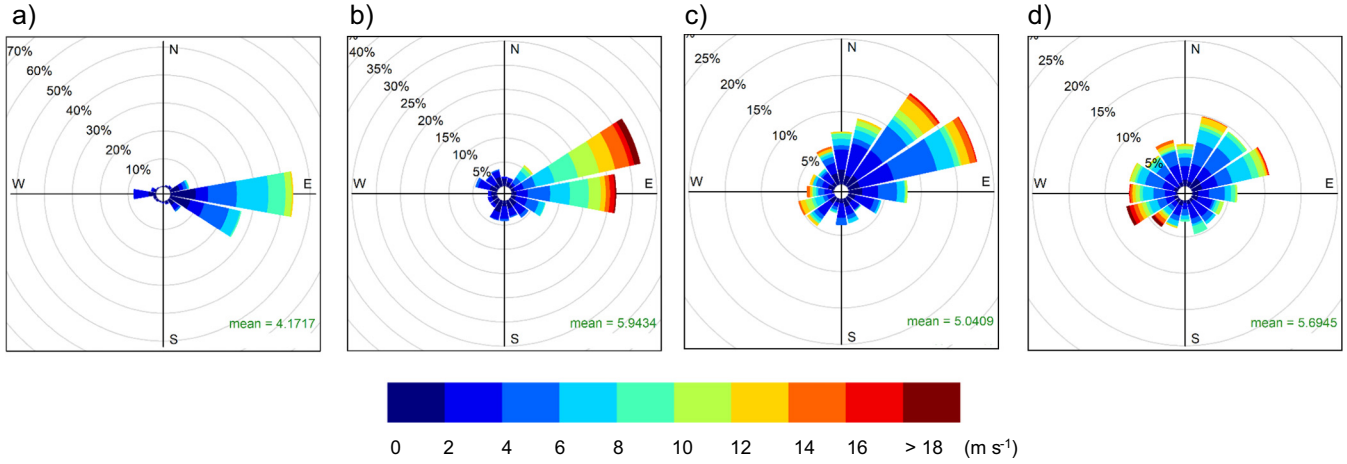

Fig. 6. Wind rose diagrams for a) surface measurements (10 m a.g.l.) El Paso meteorological station and HARMONIE-AROME simulated wind vectors for the volcanic eruption location at b) 950 hPa (~ 0.6 km a.s.l.), c) 800 hPa (~ 2 km a.s.l.) and d) 700 hPa (~ 3 km a.s.l.) for the entire eruption period (19/9/2021–13/12/2021). Graphics were generated with Openair software (Carslaw and Ropkins, 2012).

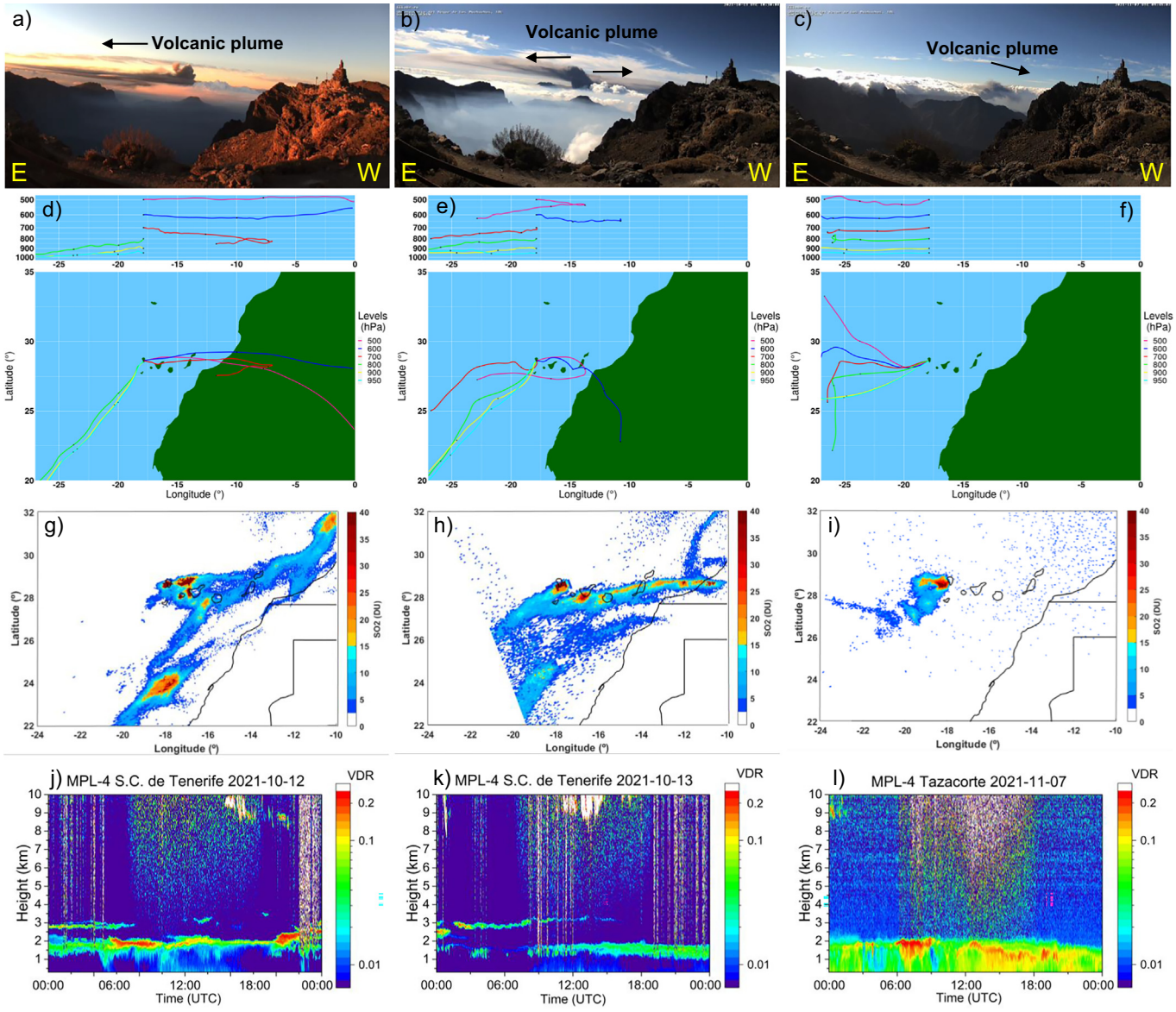

Fig. 7. Images taken from webcam at Roque de los Muchachos facing south for a) 12/10/2021 07:15 UTC, b) 13/10/2021 10:30 UTC and c) 7/11/2021 09:45 UTC. Credit: Proyecto EELabs, IAC (Instituto Astrofísico de Canarias). Note that the East direction is shown on the left of the image and West on the right. d) to f) Forward trajectories (72 h), starting point at the eruptive centre, from FLEXTRA using ERA5 reanalysis at six pressure levels (950, 900, 800, 700, 600 and 500 hPa). g) to i) Satellite column integrated SO_2 from TROPOMI. Credit: Copernicus Sentinel-5P TROPOMI. Micro-pulse lidar volume depolarisation ratio profiles extracted for Santa Cruz, j) (12/10/2021) and k) (13/10/2021) and Tazacorte l) (7/11/2021).

and largest impacts for PM_{10} and $PM_{2.5}$ surface air quality in La Palma occurring in the earlier part of the La Palma 2021 volcanic eruption (Phase I), in contrast to the largest SO_2 impacts observed in Phase II (see Fig. 5 and Fig. S3). Indeed, the data demonstrates that the majority of the PM_{10} and $PM_{2.5}$ exceedances of air quality thresholds occurred during Phase I (Fig. S3). This behaviour is also illustrated by the evolution of the SO_2/$PM_{2.5}$ concentration ratio during the volcanic eruption (Fig. 5f), which demonstrates how the $PM_{2.5}$ concentrations dominate in Phase I of the eruption while the SO_2 concentrations dominate in Phase II.

It is important for air quality managers or for future studies of the health impacts of the La Palma 2021 volcanic eruption to note that simultaneous exposure of both SO_2 gas and volcanic PM occurred during the eruption, and this should be further studied particularly in terms of public health implications. It is also important to highlight that the temporal evolution of SO_2 gas and volcanic PM showed opposite temporal trends throughout the eruption, leading to a dominance of PM concentrations in Phase I of the eruption and of SO_2 concentrations in Phase II, therefore the chemical composition of the plume originating from the volcanic eruption likely varied temporally during the La Palma 2021 eruption. Differences in the chemical composition of volcanic plumes and their subsequent health effects have been studied by Carlsen et al. (2021) and various studies have investigated the health effects of both SO_2 gas and sulphate aerosol exposure from volcanic eruptions (e.g. Longo, 2013). Epidemiological studies are currently being implemented to study the health effects of the La Palma 2021 eruption on various population groups (e.g. Ruano-Ravina et al., 2023). Further studies are needed to determine the chemical composition of the atmospheric aerosols and their temporal evolution during the La Palma 2021 volcanic eruption and their impact on health.

4. Conclusions

The La Palma 2021 eruption was the first subaerial eruption after a 50-year quiescence in the Canary Islands and the longest volcanic eruption in La Palma in the last five centuries, causing considerable destruction and

damage to homes, buildings, crops and other infrastructures. This study conducts a comprehensive evaluation of the impact of this eruption on air quality, covers the entire 85 days of the process (19/9–13/12/2021) and employs a multidisciplinary approach, including measurements from instrumentation already in place in La Palma and deployed as part of the emergency response. High concentrations of SO_2, PM_{10} and $PM_{2.5}$ were observed at ground level in La Palma (showing a strong volcanic enhancement compared to background concentrations) and also sporadically at ~ 140 km distance on the island of Tenerife, in the free troposphere at Izaña Observatory (~ 2.4 km a.s.l.). An unusual feature of the volcanic eruption was that volcanic aerosols and desert dust both impacted the lower troposphere in a similar height range (~ 0–6 km), therefore the air quality in La Palma during the volcanic eruption was impacted by both natural phenomena concurrently. This led to a greater increase in PM surface concentrations and this cocktail of SO_2 coupled with volcanic and dust aerosol should be further studied particularly in terms of public health implications. The impact of the La Palma 2021 volcanic eruption on surface SO_2 and PM concentrations was strongly influenced by the substantial magnitude of the volcanic emissions (~ 1.8 Tg), the injection height of emissions and the strong vertical stratification of the atmosphere and its seasonal dynamics in this region. As a consequence of the interplay of these modulating factors, SO_2 concentrations at ground-level showed an opposite temporal evolution to particulate matter and increased in the latter phase of the eruption, when a greater proportion of the plume was confined to lower levels of the troposphere, despite significantly reduced SO_2 volcanic emissions in this phase. The results of this study are relevant for emergency preparedness for future volcanic eruptions in the subtropical region and for all international areas at risk of volcanic eruptions; a multidisciplinary approach is key in understanding the processes by which volcanic eruptions affect air quality and to mitigate and minimise impacts on the population.

CRediT authorship contribution statement

All authors contributed to the writing of the manuscript and to data curation. C.M., C.T., O. G., N.P., T.B., N.T., S.R. and E.C contributed to conceptualization and formal analysis.

Data availability

Data will be made available on request.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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Glossary

AOD: Aerosol Optical Depth
AEMET: State Meteorological Agency of Spain
AQG: Air Quality Guideline
AQMN: Air Quality Monitoring Network
CSIC: Spanish National Research Council
EC: European Commission
ECMWF: European Centre for Medium-Range Weather Forecasts
FLEXTRA: FLEXible TRAjectories model
FT: Free Troposphere
HARMONIE-AROME: (Hirlam Aladin Regional/Meso-scale Operational NWP in Euromed)
IARC: Izaña Atmospheric Research Center
IZO: Izaña Observatory
MBL: Marine Boundary Layer
MODIS: Moderate Resolution Imaging Spectroradiometer
MOUNTS: Monitoring Unrest from Space
MPL: Micropulse Lidar
MPLNET: Micropulse Lidar Network
NRT: Near Real Time
NWP: Numerical Weather Prediction
OPS: Optical Particle Sizer
PDR: Particle Depolarization Ratio
PEVOLCA: Plan Especial de Protección Civil y Atención de Emergencias por Riesgo Volcánico en la Comunidad Autónoma de Canarias
PM_{10}: PM with aerodynamic diameter < 10 μm
$PM_{2.5}$: PM with aerodynamic diameter < 2.5 μm
S-5P: Sentinel-5 Precursor
SAL: Saharan Air Layer
SCO: Santa Cruz Observatory
TROPOMI: TROPOspheric Monitoring Instrument
TWI: Trade Wind Inversion
UVU: Ultraviolet
VDR: Volume Depolarization Ratio
WHO: World Health Organisation
WMO: World Meteorological Organization