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Key Points:

- Plumes from volcano Sabancaya can be transported to Arequipa through a series of advection and dispersion processes, thus exacerbating air pollution
- Sabancaya plumes can be captured by Mountain Chachani and transported to Arequipa by nighttime downslope winds linked to gravity waves
- Daytime convective boundary layer entrainment and dispersion further brings elevated plume over Arequipa to the surface

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Mountain-Facilitated Lee-Slope Transport and Daytime Boundary Layer Mixing of Volcano Plumes Exacerbates Air Pollution Over Arequipa, Peru

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Abstract Severe air pollution plagues Arequipa, Peru, due to anthropogenic and natural emissions. Persistent volcano emission in the vicinity of Arequipa makes it among the largest SO₂ sources in the world. Because volcano plumes mostly exist in the free troposphere and stratosphere where horizontal transport acts rather quickly, previous studies mostly focused on their global-scale impacts. Whether these plumes can affect near-surface air quality has not attracted much research attention. This study uses WRF-Chem simulations to reveal that in the presence of northerly/northwesterly winds and favorable mountain meteorology, the plume from volcano Sabancaya (elevation 5,960 m, ~80 km north of Arequipa) can be brought down to near the surface of Arequipa through two steps of transport and dispersion processes: (a) With northerly/northwesterly winds, the free troposphere plume from Sabancaya is transported southward and intercepted by Mountain Chachani located between Sabancaya and Arequipa and subsequently transported downward to Arequipa by nighttime downslope winds linked to large-amplitude lee-side mountain gravity waves. Often the plume reaches down to be close to the boundary layer over Arequipa. (b) In the following day, convective boundary layer growth brings the above boundary-layer plume to near the surface through vertical mixing processes, thus exacerbating ambient air pollution in Arequipa. A mechanism on how volcano plumes above 6-km height cause air pollution over the lower-lying Arequipa city is therefore revealed for the first time. The mountain dynamic effect in inducing the large-amplitude mountain lee waves is further illustrated by an idealized simulation excluding mountain's thermal effect.

Plain Language Summary Severe air pollution often plagues Arequipa, Peru. However, its air pollution formation mechanisms and contributing factors in the region are barely examined. Persistent gas emission from volcanoes in the surrounding region makes the region one among the largest SO₂ sources in the world. Given the high elevation of the volcano vent, it is not clear if the emitted gas plumes can reach the ground level to affect air quality there. Numerical model simulations performed in this study reveal how under favorable meteorological conditions the SO₂ plume released from volcano Sabancaya that is located 80 km north of Arequipa is brought down to the low-lying Arequipa city. When the prevailing winds blow from the north/northwest, the SO₂ plumes from Sabancaya are transported southward to encounter Mount Chachani. At night, the SO₂-carrying stable flow descends along the lee slope of Mount Chachani, in the form of downslope winds in response to the mountain-forced gravity waves. During daytime, the SO₂ plume transported to over the boundary layer of Arequipa are further mixed downward to the surface by boundary-layer eddies forced by surface heating, causing air pollution in Arequipa.

1. Introduction

Severe air pollution plagues several regions over South America, including the Arequipa city beside Andes in southern Peru due to anthropogenic emissions, volcanoes, wildfires, dust emissions, and poor air quality management (Carbo-Bustinta et al., 2022; Cazorla et al., 2022; Colla et al., 2021; Feron et al., 2023; Gómez Peláez et al., 2020; Jury & Gaviria Pabón, 2021; Larrea Valdivia et al., 2020; Li, Michalski, et al., 2021; Michalski et al., 2022; Nawaz & Henze, 2020; Olson et al., 2021; Silva et al., 2018; Yamasoe et al., 2015) with the

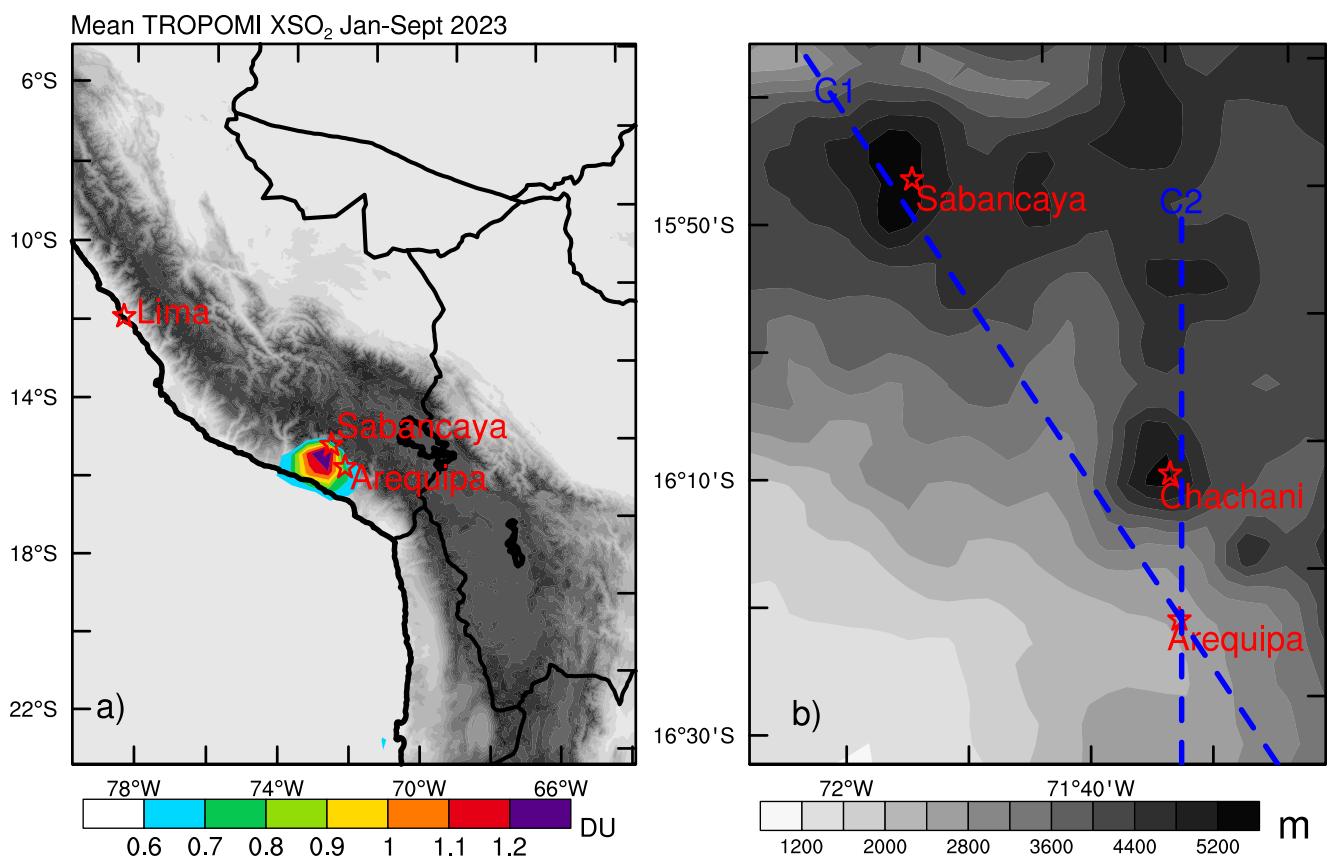


Figure 1. (a) Average column-integrated SO_2 (XSO₂) during January–September 2023 from TROPOMI overlaid on terrain height in domain 2 of WRF-Chem simulations and (b) detailed terrain height around Arequipa, Peru. Location of cross-sections (C1 and C2) are marked.

amplification effects of the meteorological processes in this mountainous region. Particulate matter (PM or aerosols), sulfur dioxide (SO_2), mercury, carbon monoxide (CO), nitrogen oxides (NO_x), and ozone (O_3) are the major air pollutants in the Arequipa region. PM with a diameter $\leq 2.5 \mu\text{m}$ (PM_{2.5}) poses severe health risks. High PM_{2.5} is also believed to exacerbate the transmission and geographic spread of virus-driven infectious diseases. Vasquez-Apestegui et al. (2021) attributed the high rates of COVID-19 cases in Peru to the increased PM_{2.5} exposure during the pandemic years, among other reasons. PM_{2.5} concentration in Arequipa frequently exceeds $200 \mu\text{g m}^{-3}$ (Larrea Valdivia et al., 2020), with sulfate (SO_4^{2-}), nitrate (NO_3^-), and carbonate aerosols being the major components (Olson et al., 2021). Fossil fuel combustion and dust are previously presumed to be the major sources of PM_{2.5} over Peru, and other contributing sources remain to be identified (Huamán De La Cruz et al., 2019; Olson et al., 2021; Pearce et al., 2009; Valdivia-Silva et al., 2009). Unique local mining activities and volcano emissions likely exacerbate air pollution over Arequipa (Moussallam et al., 2017; Nriagu, 1994; Torres & De-la-Torre, 2022).

SO_2 , a precursor of sulfate, shows elevated column-integrated concentration (referred to as XSO₂) over southern Peru (Figure 1a), which may partially contribute to the aerosol pollution in the region through oxidization and the subsequent gas-particle mass transfer process if the near-surface SO_2 is appreciable (Graf et al., 1997; Hu et al., 2008; von Glasow et al., 2009). The Arequipa region is ranked among the top sources of SO_2 pollution in the world because of the two active volcanoes emitting significant amount of gases in the vicinity (Fioletov et al., 2016), that is, volcanoes Ubinas (70 km to the east) and Sabancaya (80 km to the north, see Figure 1b for the location). Volcanoes Ubinas and Sabancaya are among the top volcanic emitters in the world, contributing nearly 1 M tons/year of SO_2 and CO₂ (Burton et al., 2013; Carn et al., 2017; Moussallam et al., 2017). The total volcanic SO_2 emission from Ubinas and Sabancaya is dominated by persistent daily degassing rather than the occasionally explosive eruptions (Moussallam et al., 2017; Shinohara, 2008, 2013). In recent years, Sabancaya is more active than Ubinas, emitting more prominent SO_2 plumes (Figure 1a).

Because the volcano plumes from explosive eruptions mostly exist in the free troposphere and stratosphere where horizontal transport acts rather quickly and influence continental to global scale, previous studies mostly focused on global-scale impacts of volcano plumes, for example, on climate (Dutton & Christy, 1992; Free & Robock, 1999; Gao et al., 2021; Gregori, 1995; Hegerl et al., 2003; Kirchner & Graf, 1995; Mann et al., 1998; Marshall et al., 2022; McCormick et al., 1995; Raible et al., 2016; Schurer et al., 2013; Tett et al., 1999). Previous research of the impacts of volcano plumes on near-surface air quality is limited to a few low elevation volcanoes, including Kilauea (elevation 1,250 m above sea level) in Hawaii (Crawford et al., 2021; Holland et al., 2020; Tang et al., 2020; Whitty et al., 2020), volcano LUSI (elevation 700 m) of Indonesia (Hidayati et al., 2018), Miyake (elevation 775 m) of Japan (An et al., 2003), Cumbre Vieja (elevation 1,949 m) of Spain (Filonchyk et al., 2022). A model trajectory study suggests that volcano plumes injected to the upper troposphere (>5 km above sea level) barely impact surface air quality for a few European volcanoes (Thomas et al., 2017).

Sabancaya (15.787°S , 71.857°W ; summit elevation 5,960 m) has been closely monitored by the Instituto Geológico Minero y Metalúrgico (INGEMMET), a Peruvian scientific and management agency. It is characterized by daily explosions or degassing with high SO_2 emission since 2016 (Global Volcanism Program, 2022). The volcano plumes rise 800–4,000 m above the summit and drift in the free troposphere. Daily volcano SO_2 emissions vary significantly, ranging from 400 to 5,600 tons/day (Global Volcanism Program, 2022), and Moussallam et al. (2017) estimated the Sabancaya emission rate to be $2,313 \pm 529$ tons/day. Some light ashfall is reported around the volcano during recent years, but rarely reaches Arequipa that is ~ 80 km to the south, likely due to the substantial deposition rate of particles that limited the particle transport distance (Global Volcanism Program, 2022). However, whether the volcano gas plumes with rich SO_2 can reach near the surface over Arequipa and exacerbate the ambient air quality remains unknown.

While there has been some observational studies of air pollution in Arequipa (e.g., Larrea Valdivia et al., 2020; Olson et al., 2021), the air pollution formation mechanisms and contributing factors (e.g., favorable meteorological conditions) have been little examined using modeling tools in the region. Arequipa is located within the Chili River valley in the Central Andes region with the Pacific Ocean to the west and the Andes Highland to the east and northeast (Figure 1a). Mountain Chachani (summit elevation of 6,057 m) and volcano Sabancaya are 20 and 80 km to the north (Figure 1b). In this mountainous region, topography plays an important role in modulating local meteorological processes and air quality through dynamic and thermal effects (Chow et al., 2012; Richner & Hächler, 2013; Sandu et al., 2019). Dynamic effects refer to topography's blocking effect to divert flows through its physical presence (Jackson et al., 2013), while thermal effects refer to the buoyancy effects associated with heating/cooling of topography and subsequently the lower atmospheric layers (Zardi & Whiteman, 2013). The related meteorological processes include thermally driven mountain-plain solenoids (Bao et al., 2011; Hu et al., 2014; Qian et al., 2015), mountain gravity waves (Durran, 1986; Xue & Thorpe, 1991), mountain wave breaking (Epifanio & Qian, 2008), hydraulic jump (Baines, 1998), barrier jets (Colle et al., 2002; Doyle, 1997; Emery et al., 2015; Harden et al., 2011; Holt, 1996; Li & Chen, 1998; Loescher et al., 2006; Olson & Colle, 2009; Olson et al., 2007; Overland & Bond, 1993, 1995; Parish, 1983; Schwerdtfeger, 1975), and upstream blocking (Epifanio & Rotunno, 2005). There has been increased concern about topographic effects on atmospheric pollutant transport and air quality in recent environmental and meteorological studies (Hu et al., 2014, 2016; Li, Miao, et al., 2021; Li et al., 2015; Lu et al., 2022; Zhang et al., 2018, 2019).

In this study, model simulations using the weather research and forecasting model with chemistry (WRF-Chem, Fast et al., 2006; Grell et al., 2005; Skamarock & Klemp, 2008) are conducted in the Arequipa region for the first time to reveal that in the presence of favorable meteorological conditions and with Mountain Chachani located 20 km north of Arequipa, the plumes from volcano Sabancaya can be brought to near the surface of Arequipa through a series of atmospheric processes, exacerbating urban air pollution. This study discovers an air pollution formation mechanism over Arequipa focusing on SO_2 as a first attempt.

The rest of this paper is organized as follows: In Section 2, satellite SO_2 and sounding data, WRF-Chem model configurations, and idealized numerical experiment design are described. In Section 3, the pathway of Sabancaya SO_2 plume transported to the surface of Arequipa, thus affecting ambient air quality, is elucidated using these observations and simulations. The importance of the dynamic effect of Mountain Chachani in such a transport pathway is emphasized. Finally, Section 4 contains a summary and discussion of the main findings.

2. Data Process, Model Configuration, and Numerical Experiment Design

2.1. TROPOMI SO₂ Data and Soundings

In Arequipa, in situ air pollutant measurements are deficient (de Moura & da Silva Júnior, 2023; Riojas-Rodríguez et al., 2016). To our best knowledge, currently no in situ SO₂ measurements are being made in Arequipa. Thus, satellite remote sensing data are first explored to understand the large-scale distributions of pollutants in the region. The TROPOMI instrument onboard the European Space Agency's Sentinel-5 Precursor (S5P) satellite has proven to be a valuable tool for studying SO₂ emissions and their distribution, particularly in the context of volcanic eruptions (Fioletov et al., 2020; Koukouli et al., 2022; Markus et al., 2023). TROPOMI is a hyperspectral spectrometer that images atmosphere with 13:30 local time (LT) overpasses, providing a nadir spatial resolution of 3.6 km × 5.6 km and covering a swath width of approximately 2,600 km (Theys et al., 2022). TROPOMI SO₂ product is highly sensitive to SO₂ at high altitudes, and the sensitivity to SO₂ in the lower boundary layer is somewhat limited due to contamination of aerosol, clouds, and ozone absorption.

Following the literature, we used TROPOMI SO₂ data to initially identify large-scale SO₂ patterns resulting from volcanic eruptions. In such cases, enhanced measurement sensitivity for SO₂ in the free troposphere is generally expected from TROPOMI. Specifically, we utilized the SO₂ vertical column densities (VCDs) from the Level 2 offline SO₂ product to quantify the concentration of SO₂ molecules within a given atmospheric column per unit area. These VCDs were then converted into Dobson units (DU) using a conversion factor. To ensure data quality, we applied a filtering process based on the retrieval quality assurance (QA) value, removing samples with a QA value less than 0.5. This step accounted for potential contamination in the retrievals stemming from bright surfaces, presence of clouds, and solar geometries. Finally, the QA-controlled SO₂ data were aggregated into 0.5-degree spatial bins for all available samples throughout the year 2023 (from January to September when we started drafting this paper), enabling us to generate the annual mean pattern of SO₂ distribution, as shown in Figure 1a.

Dispersion of air pollutants highly depends on atmospheric flows and stability. Daily sounding has been launched over Arequipa during early mornings (7–8 LT) since September 2022 on most days with certain days missing. Soundings on May 18 and 7 August 2023 are used for model evaluation and examining the stability conditions in these cases while sounding is not available for the case of 30 July 2023 and three cases before September 2022 examined in this study.

2.2. WRF-Chem Simulations

Daily 48 hr WRF-Chem (version 4.3.3) simulations are conducted to examine the transport/dispersion of SO₂ plumes from volcano Sabancaya for the period of October 2021–March 2022 when Sabancaya activities were recorded and reported by Global Volcanism Program (2022) and for the period of May–August 2023 when in situ soundings from Arequipa are available. Volcano plumes from Sabancaya on most days drifted to the southwest (Figure 1). Six cases in which the southeast-drifting plumes appear to have affected air quality over Arequipa are presented in this manuscript; they are those of 19 and 28 October 2021; 17 March 2022; and May 18, July 30, and 7 August 2023 (summarized in Table 1). Our analyses focus on 21–48 hr simulations to avoid the first 21 hr when the model goes through spin-up. Because the volcano SO₂ emissions dominate the SO₂ fields in the region, and background regional-scale SO₂ contributes insubstantially, we focus on the SO₂ plume from the emission of the Sabancaya volcano within the simulation domain in this study. Thus, a longer spin-up as in other chemistry simulations, where a more spun-up initial chemical composition fields are critical for the subsequent chemical reactions and fields, is unnecessary in this study. The initial and boundary conditions for meteorology are from hourly NCEP FNL (Final) operational global analysis and forecast (GFS) data on the 0.25 × 0.25° grid (National Centers for Environmental Prediction, 2015) and those for chemistry are from the Whole Atmosphere Community Climate Model (WACCM) outputs with a resolution of 0.9 × 1.25° (Gettelman et al., 2019). Two one-way nested domains are run simultaneously with 15- and 3-km horizontal grid spacings covering the entire South America and the Peruvian Central Andes region centered around Arequipa, respectively (see Figure 1 for domain 2 coverage and Figure 1a of Hu et al. (2023) for the exact domain configuration), following our previous convection-permitting regional climate simulations (Hu et al., 2023; Huang et al., 2023, 2024). Both domains use 48 vertical levels extending from the surface to 100 hPa. Physics parameterizations include the Yonsei University (YSU) PBL scheme (Hong et al., 2006), revised MM5 Monin-Obukhov surface layer scheme (Jiménez et al., 2012), and the rapid radiative transfer model for GCMs (RRTMG) longwave and shortwave radiation

Table 1*Summary of Characteristics of Meteorology and SO₂ Plumes During the Six Cases Examined in This Study*

Cases	Characteristics
19 October 2021	Prevailing northwesterly winds in the lower troposphere
28 October 2021	Prevailing northwesterly winds in the lower troposphere and vertical or overturning isentropes above ~6 km in early morning
17 March 2022	Prevailing northwesterly winds in the troposphere and two clear steps (wave + mixing) for the SO ₂ plume to be transported to the surface
18 May 2023	Prevailing northwesterly winds in the troposphere, prominent vertical or overturning isentropes above ~6 km in early morning, and sounding verification available
30 July 2023	Prevailing northwesterly winds in the troposphere, prominent vertical or overturning isentropes above ~6 km in early morning, and SO ₂ plume transported to near the surface over Arequipa in the early morning
7 August 2023	Prevailing northwesterly winds in the troposphere, sounding verification available, and two clear steps (wave + mixing) for the SO ₂ plume to be transported to the surface

scheme (Iacono et al., 2008). Because all the cases are free of precipitation over the Arequipa region, micro-physics and cumulus schemes are less relevant.

The gas-phase chemical reactions are simulated using the Regional Atmospheric Chemistry Mechanism (RACM, Stockwell et al., 1997), which is implemented within WRF-Chem using the Kinetic PreProcessor (KPP, Sandu et al., 2003). Hourly anthropogenic emissions of chemical species come from the REanalysis of the TROpospheric (RETRON) chemical composition inventories. The SO₂ emission rate from Sabancaya is set to 2,000 tons/day, and the emission is vertically evenly distributed in each model layer from the mountain summit to 10 km above sea level based on previous estimations (Global Volcanism Program, 2022; Moussallam et al., 2017). Note these previous estimations of the daily SO₂ emission rate from Sabancaya range from a few hundred to ~5,000 tons/day. Our simulations adopt a medium rate to represent a moderate daily outgassing. This study focuses on gas SO₂. Aerosol reactions/processes are excluded in these WRF-Chem simulations.

To diagnose the overall (dynamic plus thermal) effects of the nearby Mount Chachani on SO₂ plume over Arequipa, a sensitivity simulation removing the terrain of Chachani is conducted for the case of 30 July 2023.

2.3. Idealized Simulation Only Considering Mountain Dynamic Effects

The WRF-Chem simulations consider both dynamic and thermal effects associated with the complex terrains in the region. Additional idealized simulations only considering the dynamic effect of simplified mountain profiles are conducted to help isolate the dynamic effect of the mountain barrier on the lower level flows. Using an idealized approach can help understand the contributions of the most important effects/processes of primary interest (Braun et al., 1999; Hu et al., 2016; Pu & Dickinson, 2014; Saide et al., 2011). In our case, we are most interested in the flow pattern after the airstream passes over the tall Chachani Mountain located to the northwest of Arequipa.

The idealized experiments use the advanced regional prediction system (ARPS) model, a comprehensive regional- to storm-scale atmospheric modeling/prediction system (Xue et al., 2000, 2001, 2003), to simulate air flows around an isolated mountain that mimic the Chachani Mountain to some extent. The thermal effect of the mountain is excluded to isolate the dynamic effect.

An isolated asymmetric bell-shaped mountain with a peak height of 4 km is specified in the model that has an east-west half width of 30 km, and a 20- and 8-km half width on the north and the south side of the mountain peak, respectively. The horizontal grid spacing is 1 km, and vertical grid spacing increases from 100 m at the surface to 900 m at the model top located at 20 km height. The model domain is large enough to avoid any lateral boundary effect. The initial state is set close to the early morning sounding data over Arequipa and is characterized by a

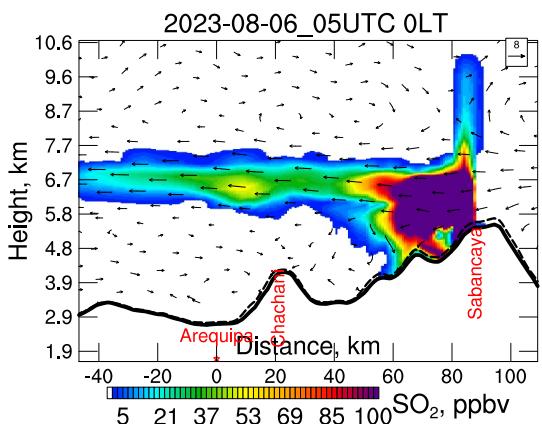


Figure 2. Simulated SO₂ plume from Sabancaya along cross-section C1 (see location in Figure 1b) in the free troposphere on the night of 5–6 August 2023.

et al., 2016, 2020). Given the Sabancaya summit elevation of 5,960 m and its plume rises to 800–4,000 m above the summit (Global Volcanism Program, 2022), the volcano plume is mainly found at 6–10 km above sea level, residing in the free troposphere having a stable stratification. The horizontal winds would normally transport the volcano plume over long distance in the stable free troposphere without much downward transport, often spreading the plume downstream across continent (Filonchyk et al., 2022). Such SO₂ plumes may not be able to affect the air quality near the surface in the Arequipa region that is located in the Chili River valley, if there is no other atmospheric process that brings the plumes down to the surface. Figure 2 shows a typical scenario with the simulated SO₂ plume in the night of August 5–6 mostly found at ~6.7 km above sea level or ~3–4 km above the ground of Arequipa.

3.2. Cases of SO₂ Plumes Using WRF-Chem Simulations

Different from the typical scenario, during periods of October 2021–March 2022 and May–August 2023, we notice some cases with northerly or northwesterly winds, in which the Sabancaya SO₂ plume is brought down to Arequipa's Chili River valley in the WRF-Chem simulations, exacerbating near-surface air quality over Arequipa. Six of these cases are presented in this paper; they are those of 19 and 28 October 2021; 17 March 2022; May 18, July 30, and 7 August 2023 (summarized in Table 1). In these cases, TROPOMI data indicate that XSO₂ plumes emitting from Sabancaya are transported southward and some of the XSO₂ plumes reach as high as 6 DU (Figure 3). Note that TROPOMI XSO₂ data on May 18 are missing while the XSO₂ plumes on May 17 and 19 are prominent. Thus, the southward transported XSO₂ plume on May 19 is shown in Figure 3 instead. Based on the prominent plumes observed on May 17 and 19, we can safely assume there was significant degassing from Sabancaya on May 18 also. The transport pathway of these cases and how they contribute to near-surface air pollution over Arequipa are examined using the simulation data.

For the Sabancaya SO₂ plumes to be transported to Arequipa, northerly or northwesterly large-scale winds are a prerequisite. Using the 7 August 2023 case as an example, in the evening of August 6, an anticyclonic circulation center was located at 64°W, 7°S or to the northeast of Arequipa at the 500-hPa level. The midtropospheric flows over the Arequipa region were northwesterly (Figure 4a). In the morning of August 7, the anticyclone center shifted to about 64°W, and the northwesterly winds on the south side of the anticyclone expanded further south (Figure 4b). Such northwesterly winds favor the southward transport of the volcanic plumes in the free troposphere toward the Arequipa region.

Note that there were clouds over Arequipa on August 6–7, as indicated by the MODIS images (not shown). However, TROPOMI was still able to retrieve the Sabancaya XSO₂ plumes, indicating at least part of the SO₂ plumes was above the clouds, confirming their free troposphere presence. The SO₂ at the high altitudes likely explains the nearly daily detection of prominent Sabancaya XSO₂ plumes by TROPOMI data even though clouds are often present in the region.

constant northerly flow of 7 m s⁻¹ and a base state with a constant static stability with a Brunt-Vaisala frequency of $N = 0.01 \text{ s}^{-1}$. The reference potential temperature (θ) near the surface is 308.5 K. The specified environmental profile and the height of the mountain result in a low Froude number flow. The model simulation is integrated up to 4 hr when the main features of the solution become established.

3. Results

3.1. Climatological SO₂ Plume From Volcano Sabancaya

Volcano Sabancaya (15.787°S, 71.857°W) was active in recent years with persistent daily degassing (Global Volcanism Program, 2022). Such daily degassing was captured by the TROPOMI XSO₂ data. In the TROPOMI average XSO₂ map during January–September 2023, the SO₂ plume with XSO₂ > 1 DU around volcano Sabancaya is the only significant source in this Central Andes region (Figure 1a). The average daily total SO₂ abundance in the region amounts to ~4,000 ton. This is consistent with previous reports that the Arequipa region is among the top SO₂ sources in the world (Fioletov

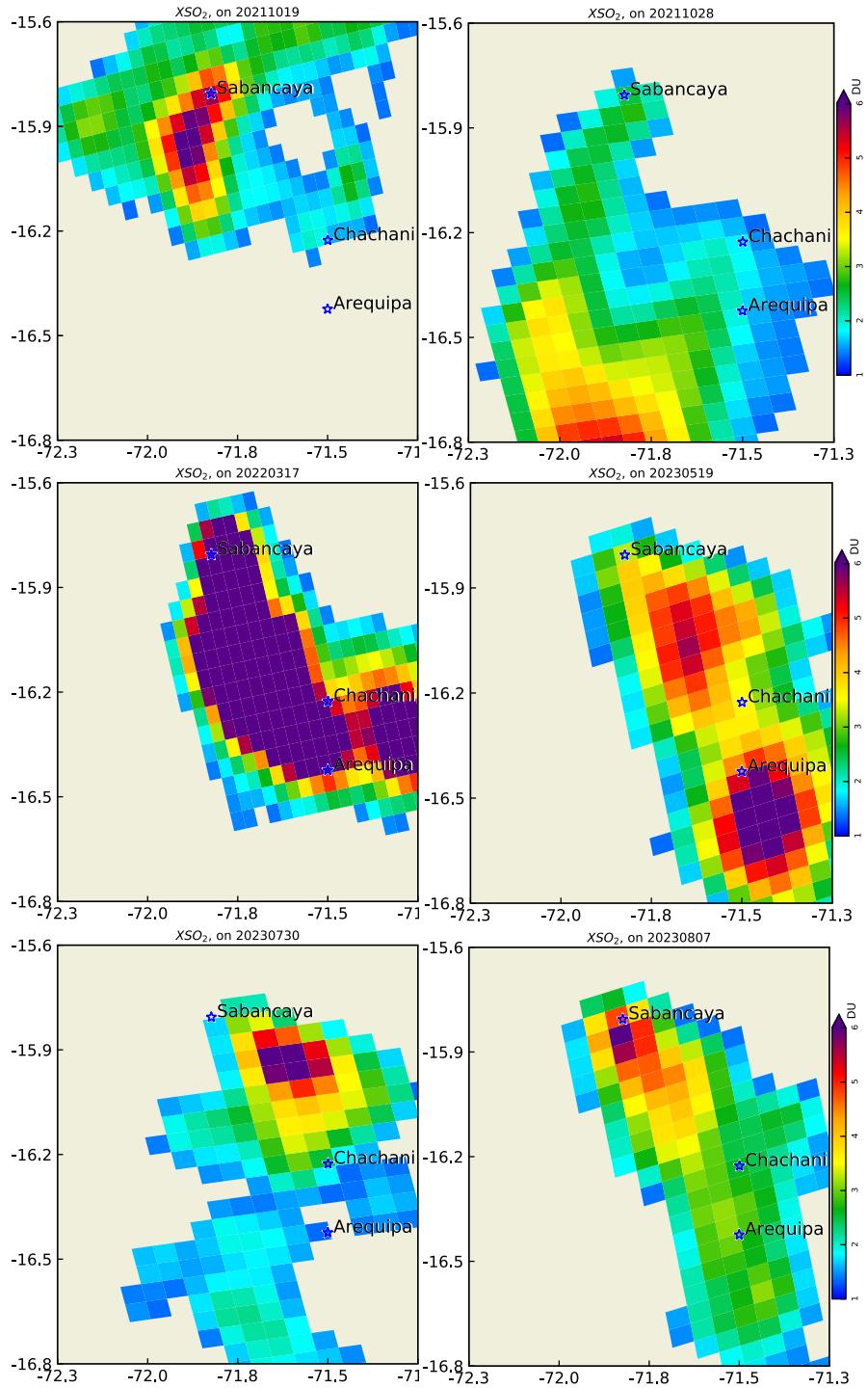


Figure 3. TROPOMI XSO₂ plumes for six cases (i.e., 19, 28 October 2021; 17 March 2022; and May 18, July 30, and 7 August 2023). Note that there is no TROPOMI XSO₂ available on May 18 while the TROPOMI XSO₂ on May 19 shows a prominent plume.

In the presence of northerly or northwesterly large-scale flows, southward-transported midtropospheric Sabancaya SO₂ plumes are intercepted by mountains around Sabancaya including Chachani with a summit elevation of 6,057 m (Figure 5). Note that the elevation of Chachani in the south-north cross-sections in Figure 5 is lower than its summit elevation because these cross-sections through Arequipa do not cut across the summit. In these

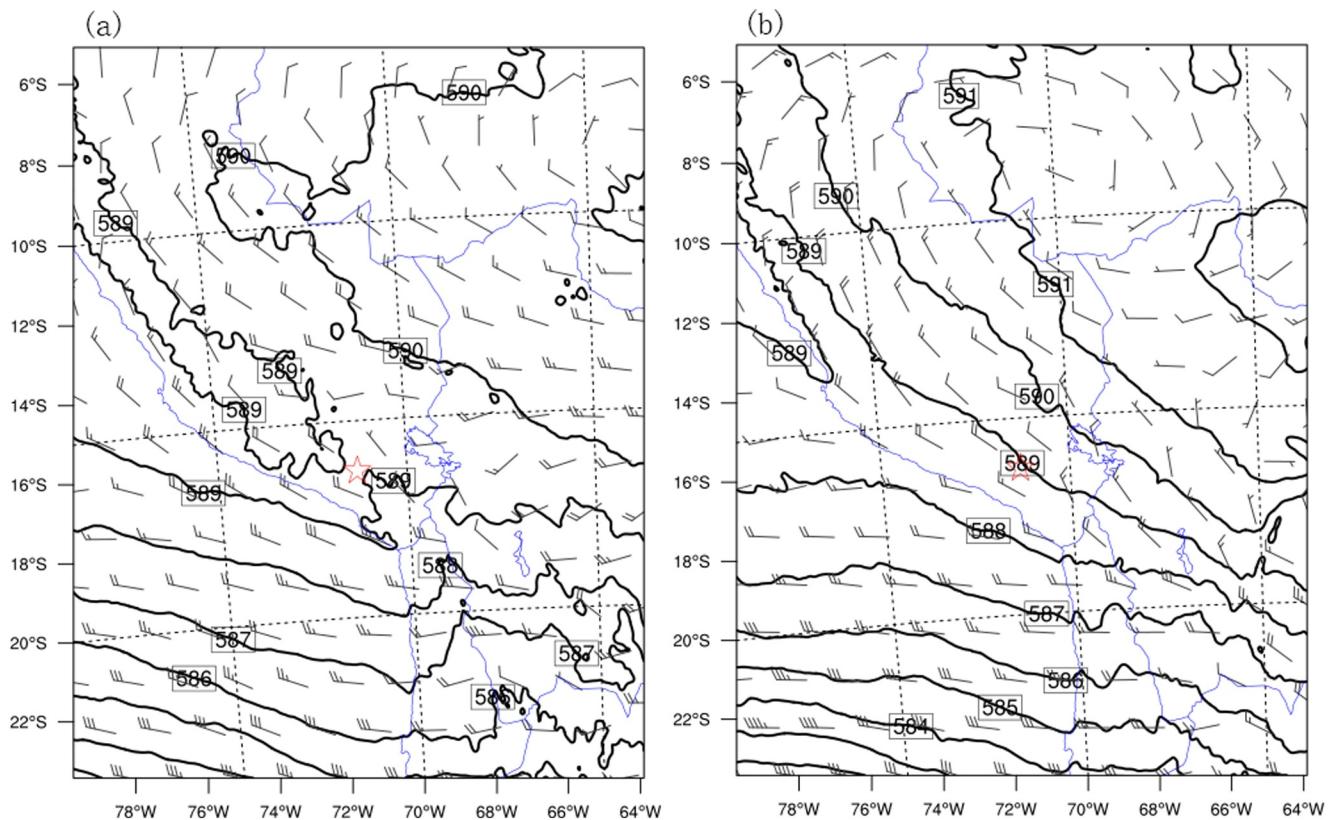


Figure 4. The geopotential height (black solid line, unit: geopotential meter [gpm]) and wind vectors at 500 hPa at (a) 18:00 LT on 2023-08-06 and (b) 04:00 LT on 2023-08-07. The blue thin lines are the country boundaries. The red pentagram is the location of Arequipa.

mountainous regions, daytime convective boundary layer can grow to 1–2 km above the ground, which roughly envelopes the mountain terrain. At some locations, the boundary layer top reaches the level of midtroposphere Sabancaya SO₂ plumes, thus capturing the volcano plumes. Mountain venting processes induced by thermally driven upvalley/upslope flows can further mix pollutants through the boundary layer top, making pollutants well mixed in a deeper layer over mountainous regions (De Wekker et al., 2004; Steyn et al., 2013). At 16 LT of all six cases, the SO₂ plumes are spread over the elevated mountainous region around Sabancaya and intercepted by Chachani (Figure 5). Thus, during the afternoon, the SO₂ concentration is elevated in those high mountainous regions. How does these SO₂-rich mountainous air mass reach low-lying Arequipa at later time is examined in detail below.

During the ensuing nighttime, as nighttime boundary layer develops, stability increases, and potential temperature shows a layered structure around mountains (Figure 6). As the air flows pass over the tall Chachani Mountain, they descend along the downwind slope of the mountain, more or less following the isentropic contours (Figure 7) as the flows are approximately adiabatic. The descending downslope flows act to transport some of the SO₂ plumes to lower levels, toward Arequipa's Chili River valley. The extents of downward transport vary on individual days (Figure 7). On 30 July 2023, the SO₂ plume penetrates to near the surface over Arequipa, while in other cases, the SO₂ plume does not reach the bottom of the valley. This is apparently strongly correlated with static stability of the layer or the potential temperature difference between the mountain top and the valley. On July 30, the difference is only about 6 K (Figures 6e and 7e) while on 17 March 2022, (Figures 6c and 7c), the difference is about 13 K. The much weaker stability on 30 July 2023 allows the downslope flow to reach much lower altitude, as the kinetic energy of the downward flow is converted to potential energy in the stable environment. Eventually, the downslope flow restores to a higher altitude, producing gravity wave oscillations whose signals propagate upward (Figure 7). For these early morning cross-sections, the downslope motion is very likely aided by nighttime drainage flows in lower valleys, enabling the SO₂ plumes to reach lower altitudes than the pure dynamically forced overmountain flow can. We can also see that within the valley near the surface, static stability

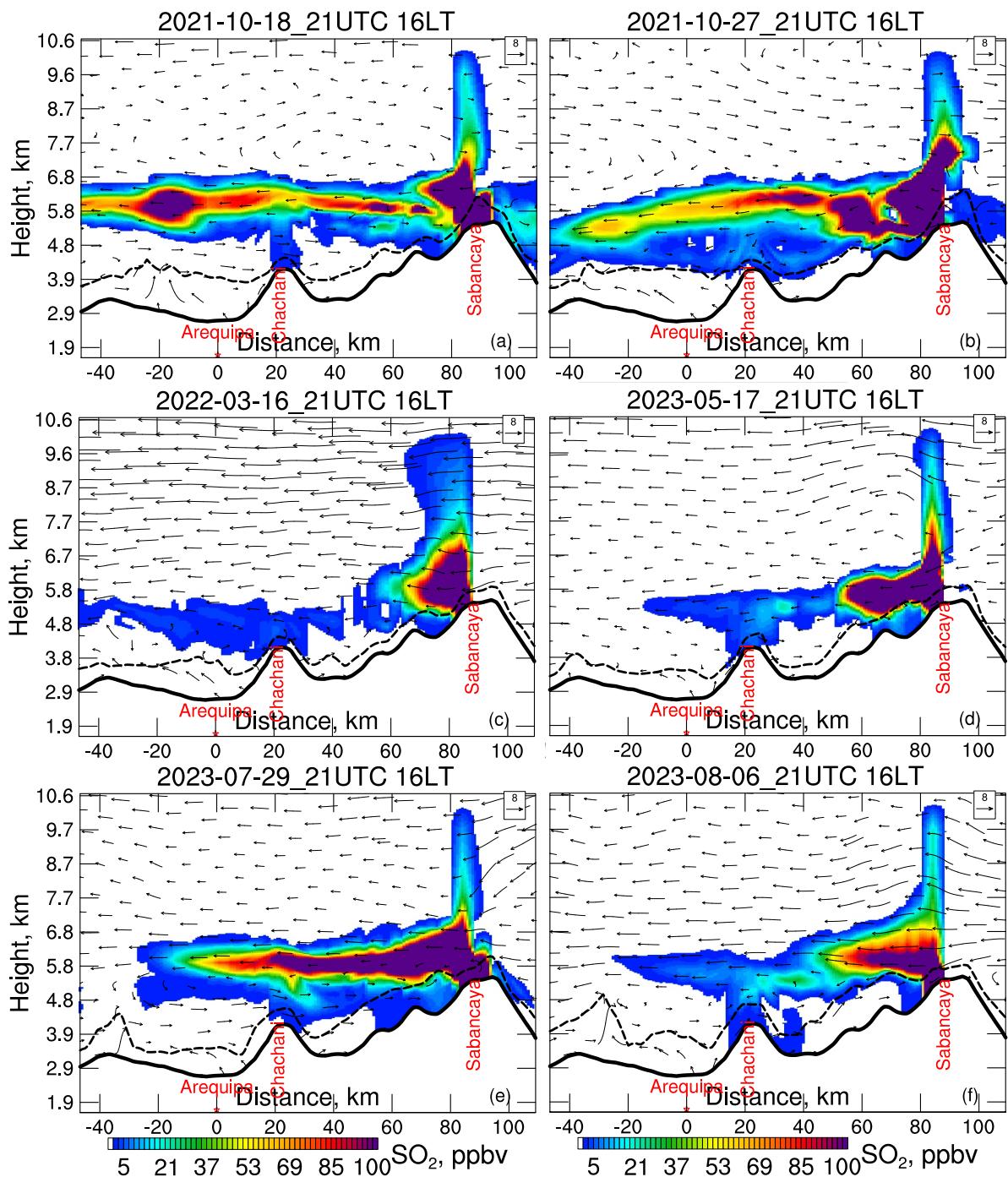


Figure 5. Cross-section of SO_2 , and winds through Arequipa and Sabancaya along C1 (see location in Figure 1b) during afternoon on (a) October 18 and (b) 27, 2021; (c) 16 March 2022; and (d) May 17, (e) July 29, and (f) 6 August 2023. The boundary layer top is marked using the black dash lines.

is strong (e.g., Figures 6d, 6f, and 7). Such a stable layer should be the result of nighttime cooling, and will make it difficult for the plumes to penetrate all the way to the ground surface. Figure 8 shows the early morning soundings observed and simulated on May 18 and 7 August 2023 at Arequipa. The simulations reproduce the general boundary layer structures reasonably well, including capturing the early morning near-surface stable layer (Figure 8).

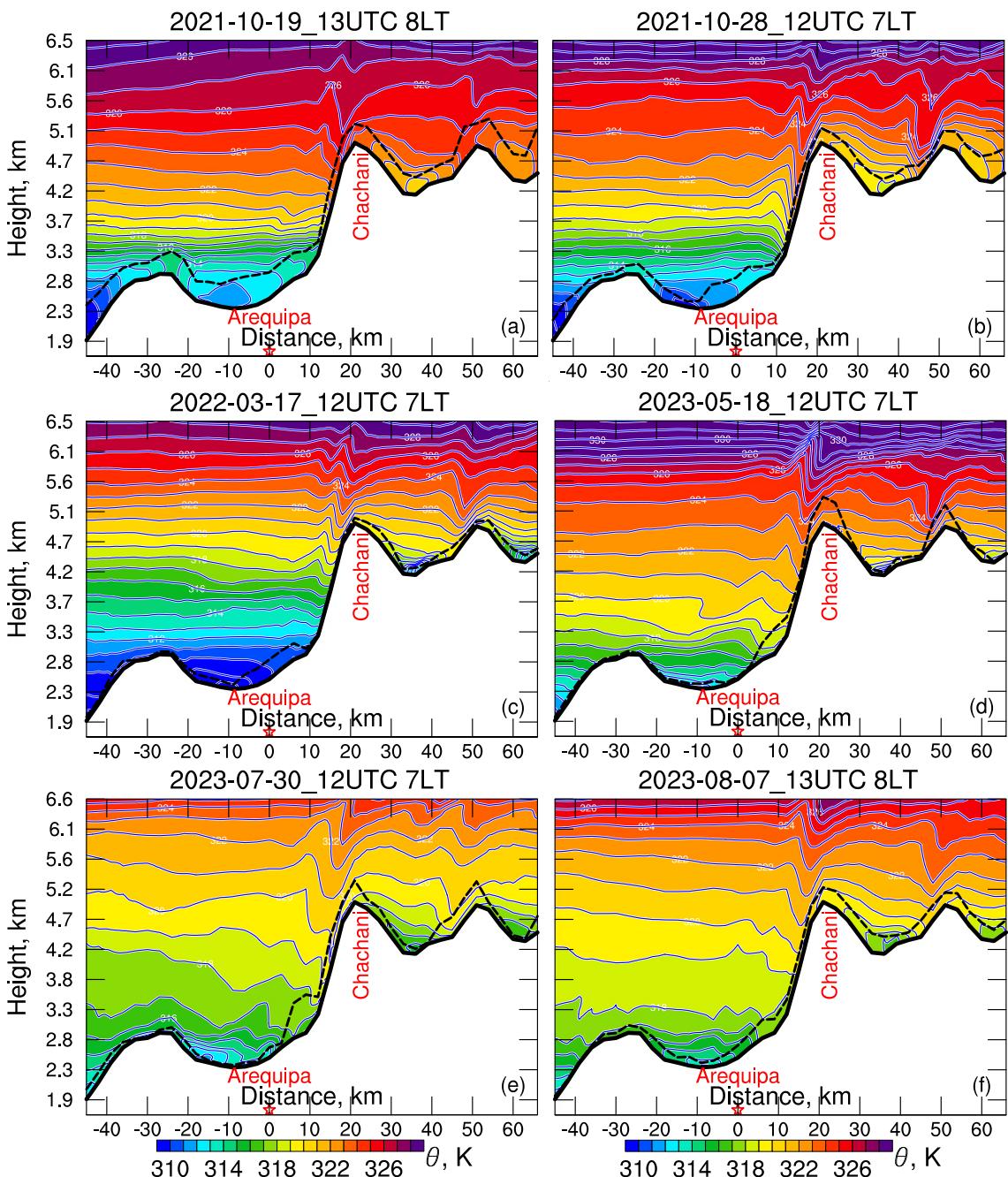


Figure 6. South-north cross-section of potential temperature through Arequipa and Mountain Chachani along C2 (see location in Figure 1b) during early morning on (a) October 19 and (b) 28, 2021; (c) 17 March 2022; and (d) May 18, (e) July 30, and (f) 7 August 2023. The boundary layer top is marked using the black dash lines.

The downward transport of SO_2 plumes on the lee side of Mountain Chachani is examined further in terms of its mechanism. Figure 6 shows the potential temperature contours or isentropes, which can be considered flow streamlines when diabatic heating and turbulent mixing are negligible (Bluestein, 1992; Bonin et al., 2020; Smith, 1979; Xue & Thorpe, 1991). Generally, the atmosphere is stably stratified on the synoptic scale in all six cases as illustrated by the vertical gradient of potential temperature (Figures 6 and 7). Flows over the Chachani Mountain are dominated by the mountain wave pattern above the lee slope during early morning on 19 and 28 October 2021; 17 March 2022; and July 30 and 7 August 2023. The upward propagating gravity waves are of significant amplitudes for these cases, and vertical or overturning isentropes are seen above ~ 6 km levels on May 18 and July 30 of 2023 (Figures 6d and 6e). When overturning occurs, the stratification becomes unstable and

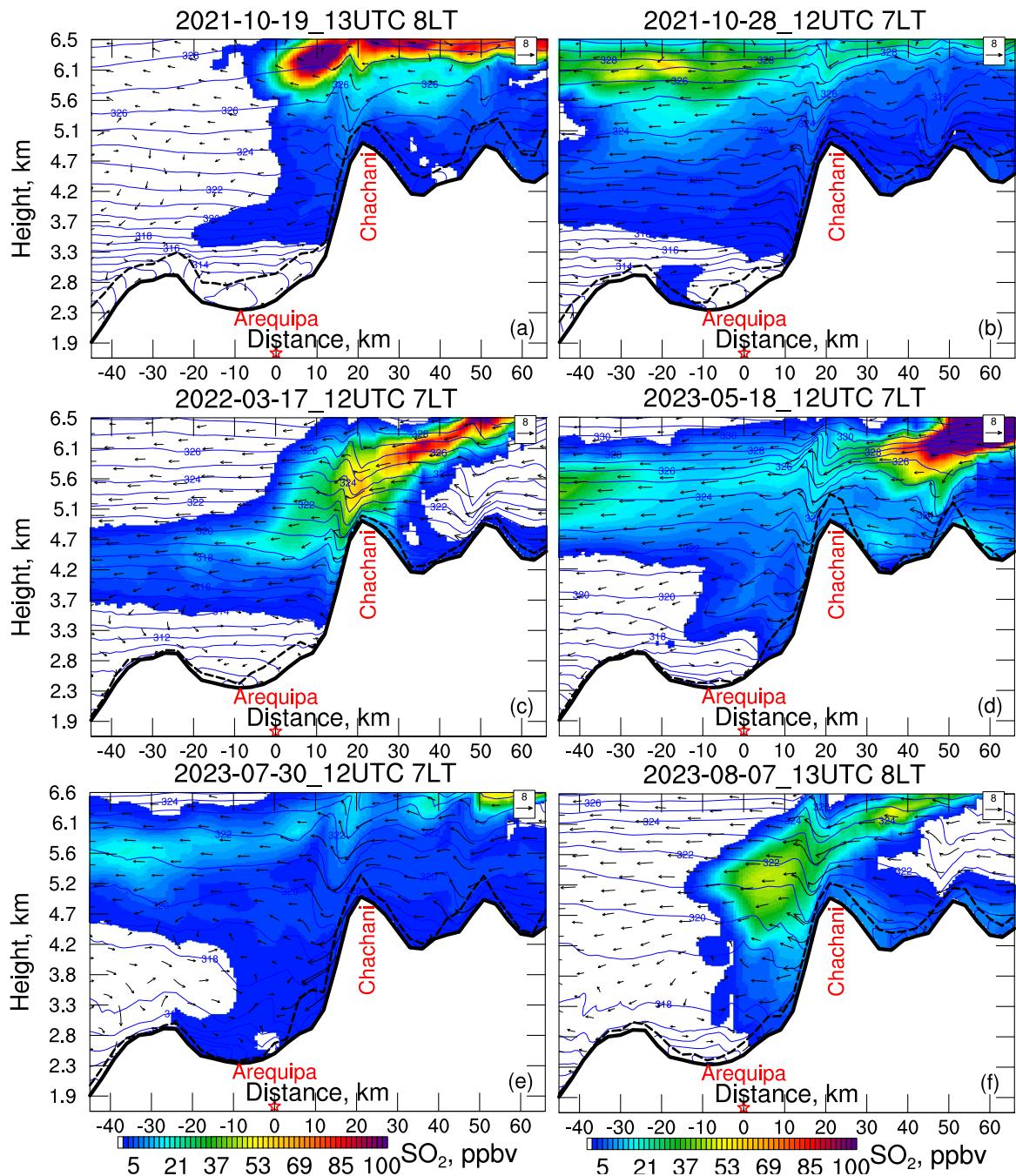


Figure 7. South-north cross-section of SO₂ (shaded), winds (vectors), and potential temperature (contours) through Arequipa and Mountain Chachani along C2 (see location in Figure 1b) during early morning on (a) October 19 and (b) 28, 2021; (c) 17 March 2022; and (d) May 18, (e) July 30, and (f) 7 August 2023. The boundary layer top is marked using the black dash lines.

gravity wave breaking would occur. Peltier and Clark (1979) showed that when wave breaking occurs, the well-mixed layer due to the breaking acts as a critical layer through which gravity waves cannot penetrate, and the downward reflected waves can interfere with upward propagating waves to create enhanced downslope winds on the lee side of the mountain. Such a process would increase the downslope transport of the SO₂ plumes in our case.

These wave features indicate that a mountain's dynamic effect dominates thermal effect under these circumstances. A mountain's thermal effect would lead to colder air near the slope during nighttime and early morning due to radiative cooling (Bao et al., 2011; He & Zhang, 2010; Hu et al., 2014; Qian et al., 2015), which is opposite

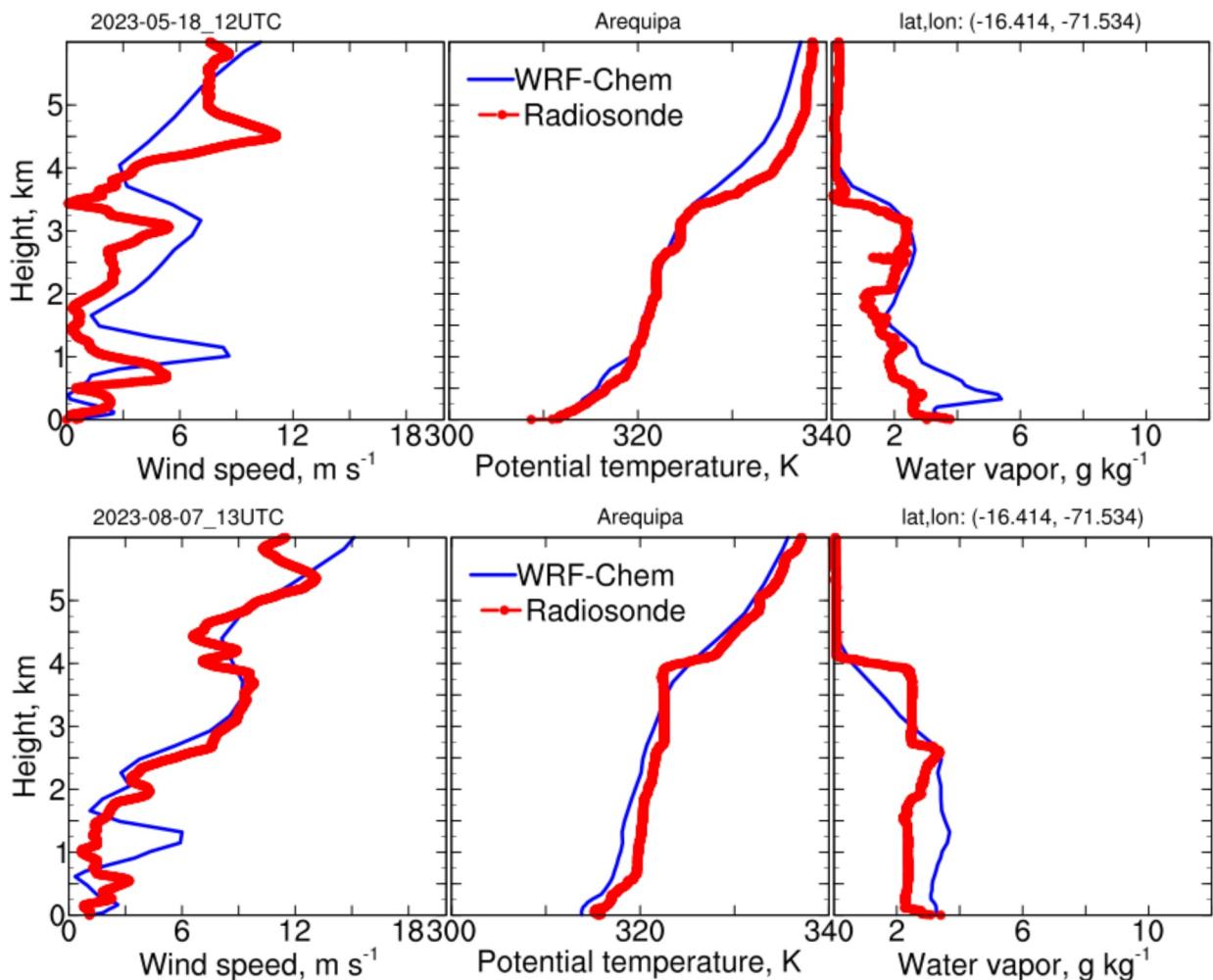


Figure 8. Observed and simulated profiles of (left–right) wind speed, potential temperature, and water vapor mixing ratios over Arequipa during early mornings (7–8 local time) on (top) May 18 and (bottom) 7 August 2023.

to the warm air zone along the upper slope seen in Figure 6. Thus, a mountain's thermal effect must play a minor role in these cases. Air behind the waves accelerates down the slope. The accelerated downslope flows bring the aloft SO₂ downward toward the valley, and the subsequent ascending flows in the wave cause the downstream spread of the SO₂ plumes across the valley (Figure 7). The wave breaking indicated by potential temperature overturns at 7 LT on May 18 and 30 July 2023 (Epifanio & Qian, 2008; Epifanio & Rotunno, 2005) is associated with the accelerated flow near the slope surface and a quick turbulent mixture region above the lee slope, which provides a favorable condition for enhanced SO₂ diffusion downstream for these cases (Figures 7d and 7e).

The vertical movement of air flow in the cross-sections can be illustrated more clearly using the cross-section-normal vorticity component, in this case the y-z plane-normal vorticity $\zeta = \partial w / \partial y - \partial v / \partial z$ (Figure 9). A narrow zone with negative vorticity along the upper part of lee slope is present in all six cases, indicating the downdrafts along the lee slope. The narrow negative vorticity zone, thus downdraft, extends above the Arequipa valley with a positive vorticity zone below, or positive and negative vorticity zones alternating below in some cases, that is, 17 March 2022, and 18 May 2023, indicating reversed circulations over Arequipa (Figure 9). Such reversal circulations and associated turbulence eddies contribute to further downward transport of the downstream SO₂ plumes over Arequipa. However, the altitudes of the reversal layer are different, likely dictated by atmospheric stratification. As a result, the extents of the downward penetration of the SO₂ plumes are different for each case, with the SO₂ plume on 30 July 2023, penetrating to near the surface over Arequipa, and the early

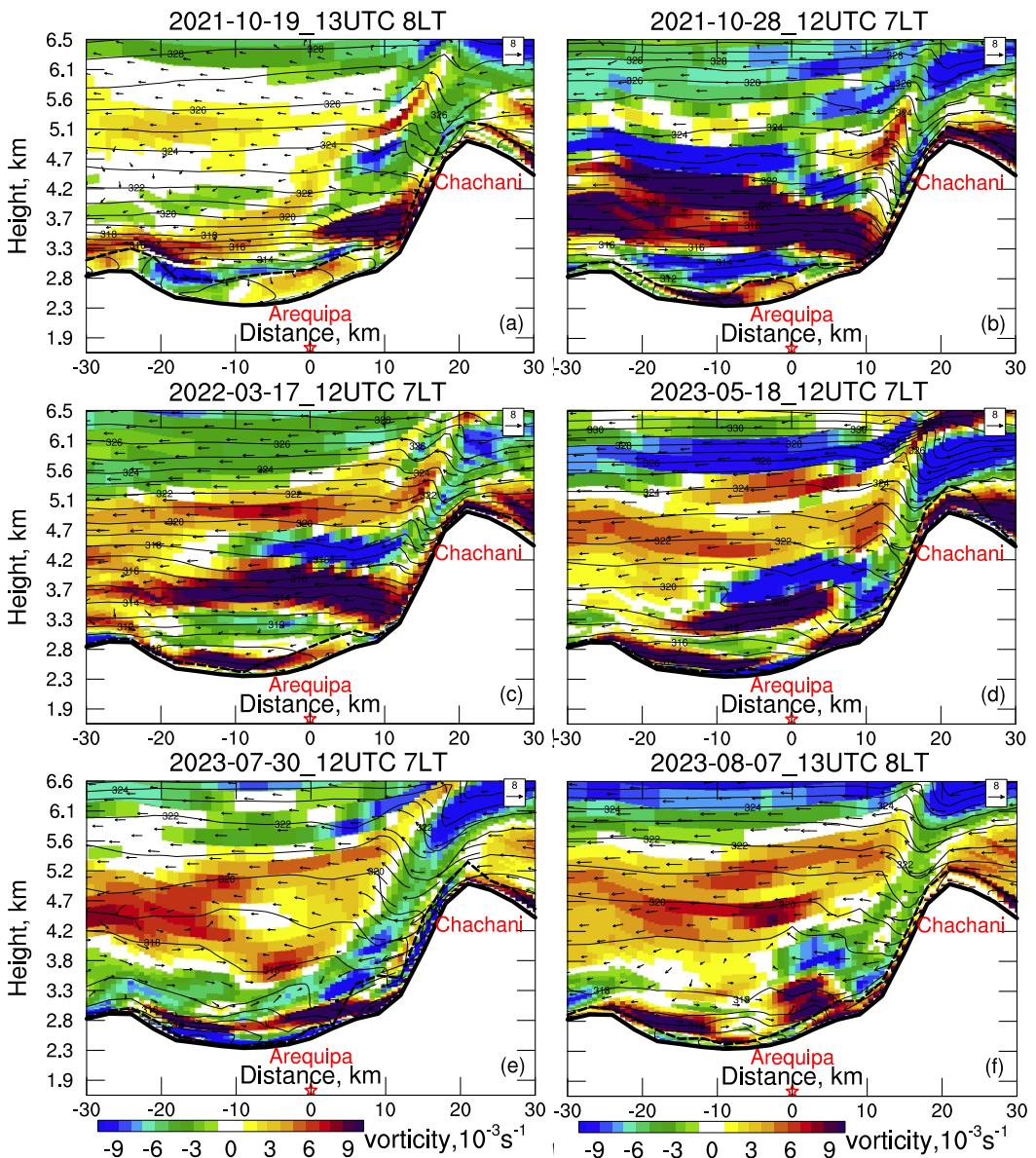


Figure 9. South-north cross-section of vorticity, winds, and potential temperature through Arequipa and Mountain Chachani during early morning on (a) October 19 and (b) 28, 2021; (c) 17 March 2022; and (d) May 18, (e) July 30, and (f) 7 August 2023. The boundary layer top is marked using the black dash lines.

morning plumes during all the other cases spread above the stable boundary layer, not penetrating through the top of the boundary layer (Figure 7).

Nighttime downward transport of SO_2 plumes facilitated by the gravity waves is further confirmed by a WRF-Chem sensitivity simulation removing Mountain Chachani for the case of 30 July 2023. This case has the widest early morning spread of SO_2 plumes into the Arequipa valley. With Chachani removed, the nighttime downslope transport of the SO_2 plume is less prominent and the SO_2 plume in the Arequipa valley is less widespread in the morning of 30 July 2023 (Figure 10).

Such mountain-facilitated downward transport of SO_2 plumes by the downslope winds linked to mountain gravity waves and by reversed local circulations occur mostly during late night and early mornings. In addition to the case of July 30 seen in Figure 10, time evolution of the waves and local circulations for the case of 28 October 2021 is illustrated in Figure 11 more clearly by focusing on/shading potential temperature rather than SO_2 . At 03 LST

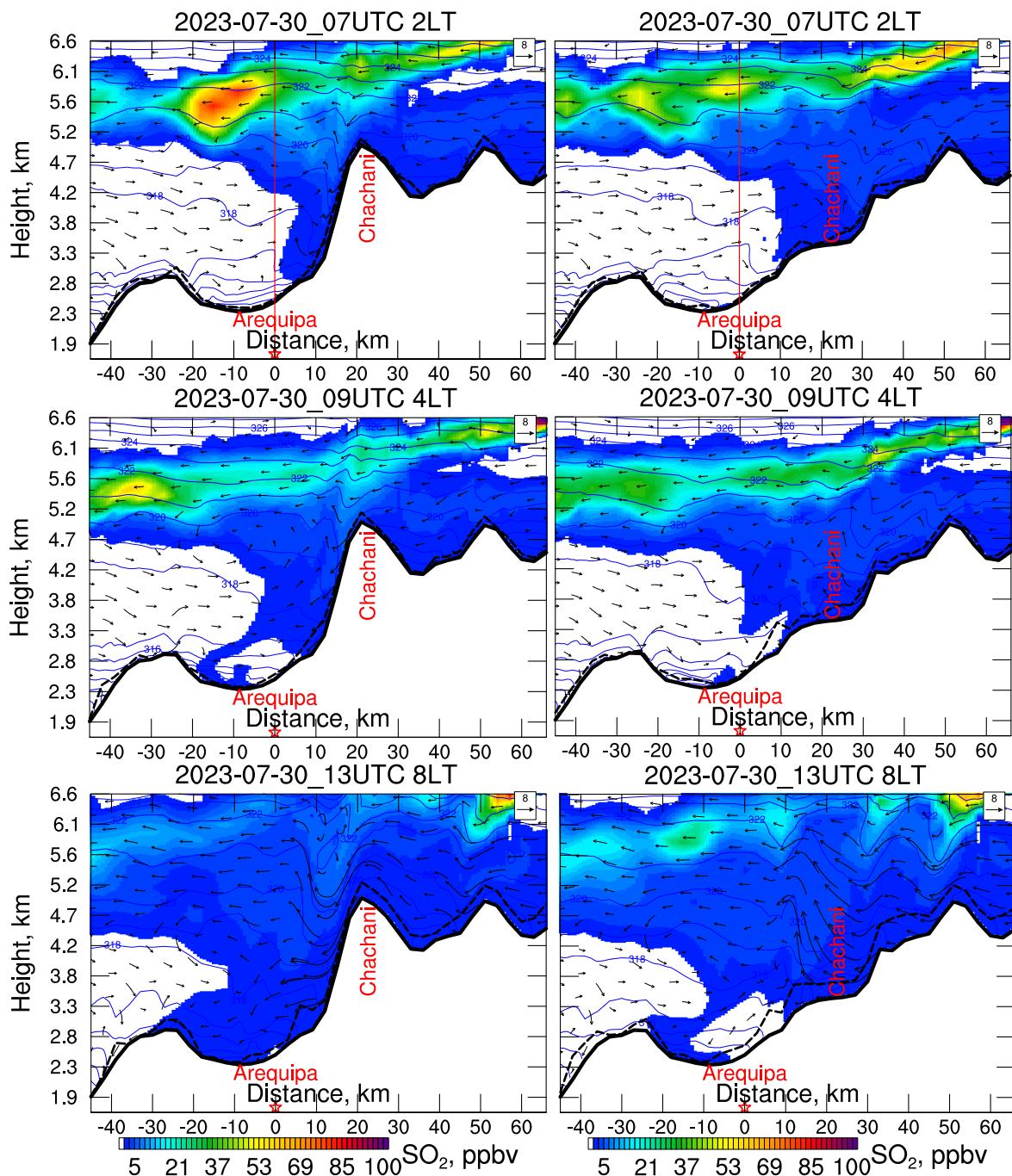


Figure 10. South-north cross-section of SO₂ (shaded), winds (vectors), and potential temperature (contours) through Arequipa and Mountain Chachani at (top–bottom) 2, 4, and 8 LT on 30 July 2023 (left) with Chachani and (right) without Chachani. The boundary layer top is marked using the black dash lines. Red vertical lines over Arequipa are drawn in the top panel to measure the distance of the SO₂ plume from Arequipa.

(Figure 11a), a near-surface stable layer is established on the mountain top north of the Chachani peak and within the valley while the gravity wave motion is still weaker. By 04 LST (Figure 11b), the wave amplitudes have increased significantly, with the downslope flow off the mountain peak on the lee side fully developed. The rapid amplification of the waves with rapid downslope descent is likely due to the occurrence of overturning at ~ 6 km level, as evidenced by the nearly vertical isentropes there. At 04 LST, there is a thin stable boundary layer above the terrain profile, due to nighttime cooling. This process would create a thin layer of drainage flow on the lee slope, which will enhance the downslope flow, bringing some higher-level air with SO₂ to lower levels, and the

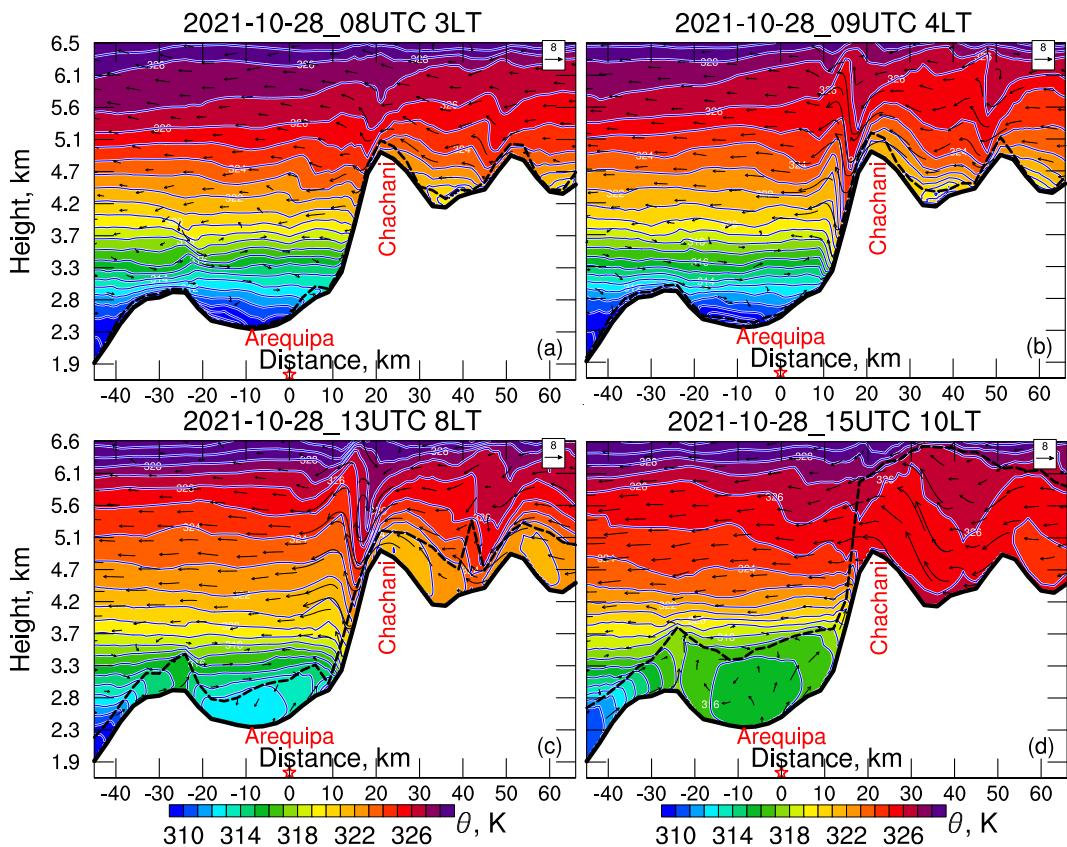


Figure 11. Time evolution of the south-north cross-section of potential temperature (shaded) and winds (vectors) through Arequipa and Mountain Chachani at (a) 3, (b) 4, (c) 8, and (d) 10 LT on 28 October 2021. The boundary layer top is marked using the black dash lines.

adiabatic cooling would allow the air parcels to cross the isentropes. By 08 LST (Figure 11c), the downslope flow has significantly weakened, likely related to the erosion of near-surface stable layer before the mountain peak due to near morning warming. The surface temperature in the valley has also increased by several degrees, the boundary layer depth has increased significantly, and the boundary layer becomes mostly neutrally stable. By this time, the aloft SO₂ plume has propagated halfway the lee slope (Figure 7). By 10 LST (Figure 11d), the boundary layer warming continues, and the temperature is increased by 3–4° upstream of the mountain top and in the valley. The weak stratification upstream of the mountain peak prevents the formation of strong mountain gravity waves on the lee side so that the downslope flow there has essentially disappeared and there is an indication of weak upslope flows on the lower to the middle lee slope by the upward bending/bulging of isentropes, due to upward advection of lower potential temperature air. Within the valley, the well-mixed boundary layer is more than 1 km deep, and such daytime mixing can easily mix SO₂-rich air above down to the surface as discussed next.

Even though the gravity waves and the associated downslope winds subside in early morning, some of the SO₂ plumes spreading above the stable boundary layer may be further transported to near the surface during the day when the convective boundary layer keeps growing, for example, 17 March 2022, and 7 August 2023 (two cases when daytime downward transport of SO₂ through vertical mixing is mostly clearly identified) as illustrated in Figure 12. At 9 LT, the SO₂ plume stays above the boundary layer on 17 March 2022, while the SO₂ plume starts to be transported/mixed to the Chili River valley to the south of Arequipa on 7 August 2023. When the convective boundary layer grows high enough (>1 km above the ground at 10 LT for the two cases) due to turbulent eddy mixing (Stull, 1988) to approach the SO₂ plumes above, the SO₂-rich air is entrained into the convective boundary layer and mixed down to the surface (Figure 12), affecting the air quality in the Arequipa city. As a result, the near-surface air quality is directly affected by the main volcano SO₂ plumes higher up in the free troposphere by 11 LT, and such a mixing process continues into afternoon, bringing down more SO₂ to the surface (Figure 12). The enhancement to near-surface air pollution through boundary layer top entrainment and downward mixing of

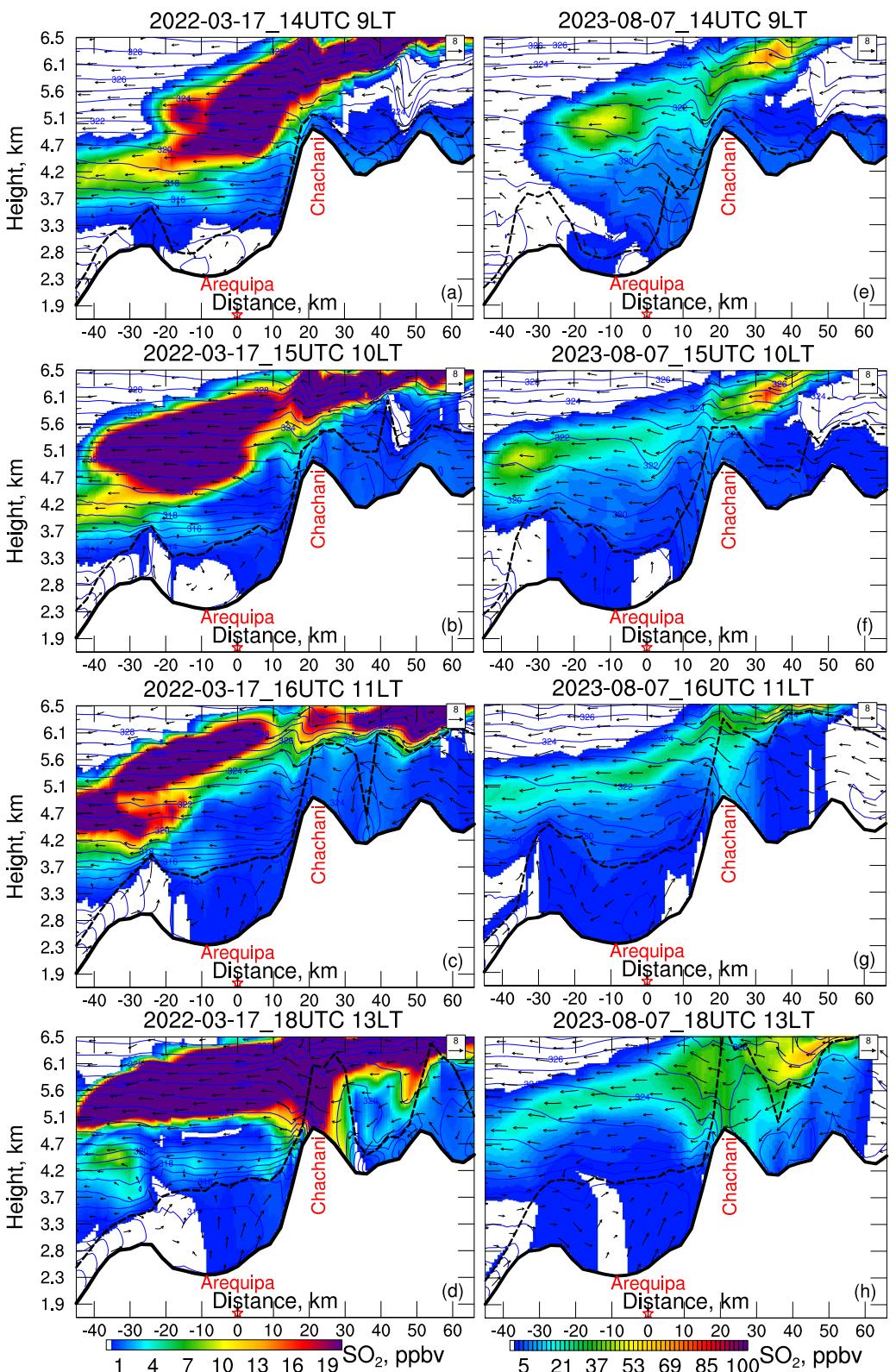


Figure 12. Downward transport of SO_2 to Arequipa when the convective boundary layer grows at (a and e) 9, (b and f) 10, and (c and g) 11 LT, (d and h) 13 LT on (left) 17 March 2022 and (right) 7 August 2023. The boundary layer top is marked using the black dash lines.

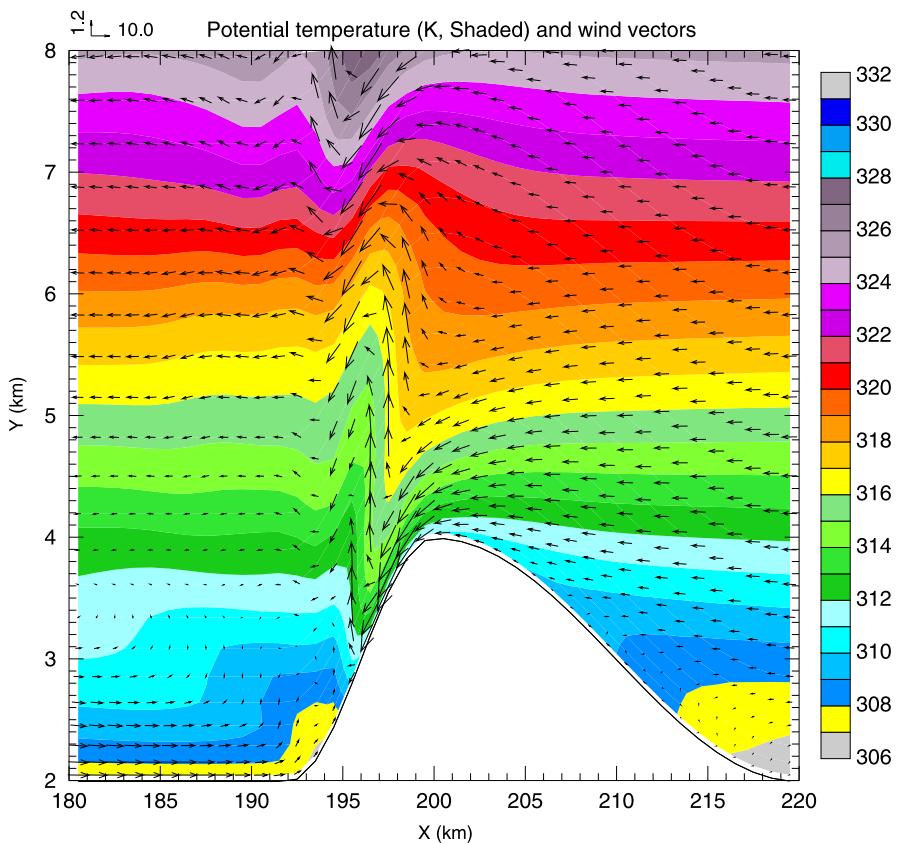


Figure 13. South-north vertical cross-section of potential temperature and wind fields through the mountain peak simulated by ARPS after 4 hr of integration.

polluted air higher up during morning hours when the boundary layer deepens has been documented in many earlier studies (Aneja et al., 2000; Hu et al., 2013, 2018; Morris et al., 2010; Neu et al., 1994; Tong et al., 2011; Yorks et al., 2009; Zhang et al., 1998; Zhang & Rao, 1999). Such a mechanism was illustrated in a single-column model study by Hu et al. (2013).

Another feature worth noting is the general downward slope of the main midtroposphere SO₂ plumes on the downwind side of the Chachani Mountain (Figure 12). The weak descent of the main SO₂ plumes appears to have played some role. More important is likely the mountain-valley circulation that develops during the day that is counterclockwise in the plots—the thermally driven upslope circulation toward the Chachani Mountain top seen in Figure 12 would induce downward motion at the middle levels above the valley. This downward motion will also help lower the SO₂ plumes, for them to be more easily reached and entrained into the boundary layer during daytime. These are named as the mountain venting processes (De Wekker et al., 2004; Steyn et al., 2013).

3.3. Air Flows in an Idealized Simulation Considering Mountain Dynamic Effect Only

The WRF-Chem simulations analyzed above use real complex terrain and involve both dynamic and thermal forcing of the mountain surface. To illustrate the effect of pure dynamic forcing by the mountain peak like that of Mount Chachani on a stable flow over the mountain, we perform an idealized simulation using the ARPS model (Xue et al., 2000, 2001, 2003) that had been shown to work well for this type of problem (Doyle et al., 2000). The 2-km asymmetric bell-shaped mountain sits on a plateau of 2 km above sea level (see Figure 13), mimicking Mount Chachani rising above the lower-lying Arequipa valley (Figure 12). Figure 13 shows the simulated wind vectors in the vertical cross-section through the mountain peak, and the potential temperature contours that approximately represent the flow streamlines. The simulated flow has roughly reached a steady state by this time.

Flows over the idealized mountain are dominated by the large-amplitude mountain wave feature above the lee slope in the presence of a stable stratification. The flow over the upper part of the slope features flow overturning

with nearly vertical isentropes, as a response of stable flow passing over a tall mountain (Figure 13). Such flow structures are similar to that over Mount Chachani during nighttime and early morning. The 312-K isentrope descends by about 1 km over the lee slope in the simulation (Figure 13), similar to the real case simulations (Figure 11b) discussed earlier. Thus, this idealized simulation further confirms that the dynamic forcing of Mount Chachani plays an important role to induce the downslope flow on the upper part of the lee slope as part of the high-amplitude mountain gravity wave response. The downslope winds bring the SO₂ plumes from the mid-troposphere to the lower levels, where the SO₂ plumes can be further transported by a thin-layer nighttime drainage flow in the real case and mixed downward by daytime boundary mixing, leading to SO₂ pollution within the Arequipa valley on some days.

4. Conclusions and Discussion

TROPOMI data suggest volcano Sabancaya (elevation 5,960 m, ~80 km north to Arequipa) emits significant amount of SO₂ with ~4,000 ton of SO₂ drifting in the air over the Arequipa region on a daily base. This study aims to examine whether such SO₂ plumes can be brought down to the lower-lying Arequipa city, thus exacerbating ambient air quality in this mountainous region. For the first time, we use WRF-Chem simulations to reveal that in the presence of favorable meteorological conditions, the plumes from volcano Sabancaya can be brought to Arequipa through a series of transport and dispersion processes. (a) In presence of northerly/northwesterly winds the free troposphere plume from Sabancaya is intercepted by Mount Chachani (summit elevation of 6,057 m, ~20 km north to Arequipa) and is widely spread over elevated mountainous regions during daytime through convective boundary layer growth and the mountain venting process related to upvalley/upslope flows and the corresponding compensation circulations. These SO₂-rich mountainous air mass is further transported downward to Arequipa by downslope winds behind the gravity wave over the upper slope induced by the mountain's dynamic effect during nighttime. The downslope motion is likely aided by nighttime drainage flows in a thin layer near the surface over the lower part of the slope due to nighttime cooling. Often the plume is downward transported to above the boundary layer over Arequipa until the strong stratification of stable boundary layer prevents the downward transport. (b) On the following day, convective boundary layer growth and mountain venting process further transport the plume above the boundary layer to near the surface through entrainment and vertical mixing processes, thus exacerbating ambient air pollution over Arequipa. Idealized flow simulation over a bell-shaped mountain using the ARPS model excluding the mountain's thermal effect but considering dynamic effect only further confirms that the nighttime wave motion is indeed induced by mountain's dynamic effect.

The air pollution formation mechanism revealed in this study has implications for air quality management and public health in Arequipa. Convective boundary layer growth in the morning plays an important role to transport the SO₂ plumes at the boundary layer top downward to exacerbate surface air pollution of SO₂ and secondary aerosols. Such a boundary layer process was also reported to exacerbate surface O₃ pollution rapidly in the morning in many regions around the world (Aneja et al., 2000; Athanassiadis et al., 2002; Hu et al., 2013; Morris et al., 2010; Zhang & Rao, 1999). Thus, air quality management during the early morning boundary layer growth period is critical to mitigate these pollutants to protect public health. Mountain effects also play important roles for the air pollution in the Arequipa region; thus, the discovered air pollution mechanism may also help understand the air pollution in other mountainous regions (Hu et al., 2014; Li, Miao, et al., 2021; Steyn et al., 2013).

Explicit comparisons with the air pollution impact of other volcanoes are also worthwhile for future studies. Because most volcano eruption plumes exist in the free troposphere and stratosphere, previous studies mostly focused on climate impact of volcano plumes, with limited research on impact of volcano plumes on air quality near the surface discussing a few low elevation volcanoes. Volcano plumes injected to the upper troposphere (>5 km above sea level) were presumed unlikely to impact surface air quality (Thomas et al., 2017). This modeling study discovers a mechanism of how the daily volcano Sabancaya's degassing plumes between ~6 and 10 km above sea level exacerbates air pollution over Arequipa, a mountainous region, focusing on SO₂ as a first attempt. A mountain's dynamic effect is shown to play an important role in the downward transport of the plumes. Further research regarding mountain effects including both dynamic and thermal effects on other environments such as meteorology and local climate in addition to air quality is needed in this mountainous region. More quantitative studies estimating contribution of volcano plumes to ambient aerosol pollution are warranted using model simulations including aerosol processes and more accurate volcano emissions in terms of total amount and vertical distribution (e.g., Tang et al., 2020). SO₂ partially contributes to the aerosol pollution through gas-phase oxidization and the subsequent gas-particle mass transfer process as well as aqueous-phase oxidization (Graf

et al., 1997; Hu et al., 2008; von Glasow et al., 2009). Future research could examine the impact of volcano emissions on both gas- and aerosol-phase pollutants through aerosol-including simulations.

Data Availability Statement

The Sentinel-5P TROPOMI Sulphur Dioxide data (Fioletov et al., 2020) are available at https://disc.gsfc.nasa.gov/datasets/S5P_L2_SO2_HIR_2/summary?keywords=S5P%20SO2. The sounding data are provided by Servicio Nacional de Meteorología e Hidrología del Perú (SENAMHI), Arequipa, Perú (Jara, 2024). Model data produced from this study (Hu, 2025) have been archived at CAPS website <https://caps.ou.edu/micronet/Peru.html> and the Luster NSF projects data server at the San Diego Supercomputer Center, /expanse/luster/projects/uok114/xhu2.

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