~~Assessing the influence of long-range~~ Long-range transport of ~~aerosols on the~~ air pollutants increases hazardous components of PM$_{2.5}$ ~~chemical composition and concentration~~ in ~~the Aburrá Valley~~northern South America

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Abstract. ~~Assessing long-range~~ Long-range transport (LRT) of ~~pollutants recognizes that multiple sources of varying scale and location can impact air quality~~air pollutants, from a range of sources, can substantially enhance background pollution levels, especially in urbanized region, which can exacerbate high pollution episodes. In the Aburrá Valley (AV), Colombia, and other cities in Northern South America, biomass burning (BB), dust, and volcanic degassing have been identified as sources of LRT of aerosols. However, the impact of these sources on air quality and their characterization have yet to be thoroughly studied. This work investigates the influence of these sources on the chemical composition of PM$_{2.5}$ during annual and intra-annual high-load aerosol events in the AV. We identified, tracked, and meteorologically characterized LRT events and evaluated their influence on PM$_{2.5}$ ~~concentration and~~ concentrations and its chemical composition. ~~Annually, we found that~~ We found that the LRT of aerosols from BB, dust and volcanic degassing ~~influence~~ influenced approximately 13%, 8% and 13% of days through the year, respectively. We ~~ran a~~ applied the Positive Matrix Factorization (PMF) ~~for each kind of event , identifying high contribution in~~ statistical model for the different LRT event types (e.g. BB) to quantify the corresponding PM$_{2.5}$ concentration and chemical composition. For BB events, we identified high contributions from organic carbon (OC1, OC2), F$^-$ and secondary ~~aerosols trace~~ aerosol tracers, SO_4^{2-} and NO_3^- ~~for the BB~~. For dust LRT events, crustal mineral components, along with Ti and Ca~~contribution for dust events and~~, were the primary contributors to the aerosol composition, while SO_4^{2-}, Na, Al and Ca ~~for~~ were the primary constituents during volcanic events. The ~~increasing~~ concentration of some ions and toxic heavy metals (Cr, Mn, Cd, and Ni) were also ~~related to~~ elevated during BB and volcanic degassing ~~influence~~events. BB exhibited the highest contribution of PM$_{2.5}$ within the LRT events ($\sim 11\ \mu g/m^3$), while aerosols from dust and volcanic events were also ~~significant~~ substantial ($< 7\ \mu g/m^3$).~~The study highlighted hotspot zones such as~~ Our study identifies the Orinoco and Middle Magdalena Valley ~~for BB aerosols , the Caribbean for dust,~~ as sizeable sources of BB aerosols and the Nevado del Ruíz volcano for volcanic aerosols. ~~This study gives insights~~ Additionally, we found that African dust approached the Andean region via the Caribbean route. As a result, we have identified the need for future chemical transport modeling studies in the region and ~~supports~~ new

support strategies to manage internal and external pollution sources and effects for that degrade air quality in the AV and the surrounding region.

1 Introduction

Long-range transport (LRT) of aerosols influences the chemical composition of air over thousands of kilometers (Kaneyasu et al., 2014; Wang et al., 2015; Rincón-Riveros et al., 2020) and plays a crucial role in the biogeochemical cycle of some components, such as dust and biomass aerosols, which distribute iron and phosphorus across the oceans and continents (Okin et al., 2004; Boyd and Ellwood, 2010). Furthermore, aerosols interact with solar radiation, influencing cloud formation and light scattering (absorbance) with a cooling (warming) effect on the planet (Choobari et al., 2014). In particular, dust and black carbon can contribute to albedo reduction and accelerate melting in a reduction in the albedo over snow-covered zones, regions, which can accelerate its melting, significantly affecting climate worldwide (Kaspari et al., 2014).

LRT of aerosols also increases human health risks, particularly in urban areas with high local emissions. Above allOut of the different air pollutants, fine aerosols, represented by $PM_{2.5}$ (particulate matter with a diameter less than 2.5 μm), penetrate deeply deep into the human body and trigger cerebrovascular and heart diseases, lung cancer and obstruction, and respiratory infections (Xie et al., 2021; Lippmann et al., 2013). Indeed, LRT of the LRT of aerosols can substantially increase the total $PM_{2.5}$ enlarges overall concentration to more dangerous hazardous levels and can increase specific components with particular effects on the toxicity of the aerosols in respect to human health. For instance, carbonaceous dominant particles are assumed known to be more toxic than crustal components (Tuomisto et al., 2008). Increased OC (organic carbon) and EC (elemental carbon) associated with biomass burning episodes drive (BB) episodes have been linked to a greater risk of cardiovascular diseases (Hwang et al., 2017).

The accountability of $PM_{2.5}$ concentrations from LRT has urged cooperation between cities and countries to identify and control pollution increases and population vulnerability emissions of key precursor aerosol emission to reduce population vulnerability to hazardous air pollution events. For decades, in Europe and Asia, LRT of the LRT of air pollutants has been recognized as a significant factor for of air pollution (Kulshrestha et al., 2014), and accounted into policy planshas informed the development and implementation of key policy strategies. For instance, the Task Force on Hemispheric Transport of Air Pollution (TF HTAP) is a collaboration led by the European Union and the United States that targets intercontinental and north-hemisphere transport of PM northern-hemisphere transport of particular matter (PM) and O_3, acknowledging their influence in meeting air quality goals the importance of these pollutants if air quality targets were to be met (UN, 2010). The collaboration has contributed to identifying regional sources, tracking, and basing more effective emission reduction strategies (Liang et al., 2018; Zhao et al., 2021; Dong et al., 2018). On a smaller scale, Hong Kong/Guangdong Cooperation addresses transboundary Kong-Guangdong cooperation has addressed the transboundary issue of air pollution in the Pearl River Delta Region (Zhong et al., 2013). The collaboration task consists of monitoring pollution level region (Zhong et al., 2013). This collaboration consisted of targeted of monitoring pollution levels and changes, evaluating the effectiveness of control measures, and providing feeback/training to stakeholders.

In Northern South America (NSA), ~~LRT of~~ the LRT of air pollutants is a more recently recognized public problem, and regional cooperation is gaining importance and momentum. ~~Three primary kinds of LRT of aerosols have been identified in the region:~~ Particularly in the Colombian Andes, biomass burning, desert dust, and volcanic emissions have been identified as three main sources of aerosols that can impact air quality from distant regions. Although for South America, biomass burning around the Amazon basin is one of the primary sources of aerosols (Ballesteros-González et al., 2020), atmospheric circulation patterns and substantial precipitation (i.e. aerosol wet deposition) ~~means there is limited~~ limit the LTR of aerosols towards Colombia (Hamburger et al., 2013). Therefore, transboundary emissions from open fires in the Orinoco basin and the Caribbean are ~~significant~~ more important drivers of intra-annual periods of hazardous air quality for Colombian cities such as Bogotá, Medellín, Arauca, Yopal, Bucaramanga and Villavicencio (Mendez-Espinosa et al., 2019; Rincón-Riveros et al., 2020; Henao et al., 2021; Rodríguez-Gómez et al., 2022) and on Pico Espejo in Venezuela (Hamburger et al., 2013).

Likewise, the LRT of dust has also been reported in NSA, ~~mainly produced from~~ primarily emitted from the Sahara and Sahel deserts (Prospero et al., 2020). These ~~primarily~~ particles predominately affect the Caribbean region, as reported in islands as Barbados, Guadalupe, Virginia, and Tobago Island, and ~~the continental land with the Cayenne (French Guiana)~~ parts of the continent, e.g., French Guiana (Prospero et al., 2014; Kumar et al., 2014). African dust effects have also been reported in Colombia, with the analysis of high particle loads ~~on~~ in June 2014 and 2020 (Bolaño-Ortiz et al., 2023a; Mendez Espinosa et al., 2018; Bedoya et al., 2016). Notably, the LRT of African dust in June 2014 covered 95% of the country (dust concentrations $> 90kgkm^{-2}$) (Mendez Espinosa et al., 2018). ~~Few~~ However, a few long-term assessments have also been done in the Caribbean and Andean regions (Bolaño-Ortiz et al., 2023b; Arregocés et al., 2023).

Volcanic ~~activities (~~eruption and degassing ~~)~~ also play a crucial role in the LRT of air pollutants in NSA and are part of the natural ecosystem in the region since it belongs to the Andean volcanic belt. Few studies have been conducted assessing the influence of volcanic aerosols on air quality ~~, which has mainly focussed~~ in the AV, but those which have mainly focused on the Sangay volcano in Ecuador and Nevado del Ruíz in Colombia (Casallas et al., 2024; Moran-Zuloaga et al., 2023). These studies ~~track~~ identified elevated $PM_{2.5}$ and ~~Sulfur~~ sulfur dioxide (SO_2) concentrations in the air ~~and road particles from the~~ from these volcanoes (Casallas et al., 2024; Cuesta-Mosquera et al., 2020; Trejos et al., 2021). The degassing activities ~~for~~ from the Nevado del Ruíz ~~have~~ has been especially highlighted for their magnitude and frequency (Carn et al., 2016).

In the last few years, ~~different~~ multiple studies in Colombia have ~~made substantial contributions~~ undertaken substantial efforts in monitoring and ~~identifying impacts caused by open fire emissions on~~ assessing the impacts of open fire burning emissions on local air quality (see e.g., Hernandez et al., 2019; Mendez-Espinosa et al., 2019; Ballesteros-González et al., 2020; Rincón-Riveros et al., 2020; Henao et al., 2021). Nonetheless, dust and volcanic aerosols studies have been limited to episodic high-pollution events (Mendez Espinosa et al., 2018) and do not provide ~~comprehensive information on the sources' influence and the aerosols transport (Liu et al., 2022). Even though, for the three kinds of emissions sources,~~ a comprehensive long-term assessment of LRT of aerosols from these sources. (Liu et al., 2022). Overall, there are still significant gaps in ~~understanding the representative influence on~~ our understanding of how LRT of aerosols from these three sources influences regional air quality throughout the year~~and their impact on aerosol composition, particularly~~ . This relates directly to the absolute concentrations and the aerosol chemical composition. This is especially true for dust and volcanic aerosols, which are frequently overlooked due to their uncontrolled nature (Woo et al., 2020; Pouliot et al., 2012). Obtaining the annual representation of the LRT of aerosols would contribute to the characterization of concentrations natural source type (Woo et al., 2020; Pouliot et al., 2012) . Therefore, quantifying the properties (i.e. absolute concentration and chemical composition) of aerosols from LRT events would allow for a detailed assessment of the surface aerosol mixture (and air quality impacts) at local and national scales. It enables would enable decision-makers to develop cooperative and effective projects to manage the risk of air pollution involving natural sources (Gómez Peláez et al., 2020; Jiao et al., 2021). Furthermore, advances in the characterization of the chemical composition of air give rise to can yield the opportunity to derive strategies to reduce the exposure of the population to high concentrations of certain species such as toxic heavy metals and carbonaceous matter (Gómez Peláez et al., 2020; Briffa et al., 2020; Allajbeu et al., 2017).

This study aims to analyze the impact of inter-annual LRT of biomass burning (BB-LRT), dust (Dust-LRT), and volcanic aerosols (Volcanic-LRT) on $PM_{2.5}$ concentrations and chemical composition through year-frequent high aerosol load events in the Aburrá Valley (AV), which is one of the most populated metropolitan areas in Colombia, situated over the Andean Mountains. We characterize meteorologically favorable conditions for transport to the valley the LRT events and involve information from one of the largest registered $PM_{2.5}$ chemical characterization campaigns conducted in the territory region (April 2019 to October 2022), as well as in-situ $PM_{2.5}$ measurements and satellite-based products. We identify representative events, analyze atmospheric transport, and highlight potential sources. Finally, we evaluate the impact of these sources on ground-level $PM_{2.5}$ concentrations and chemical composition in the AV.

2 Data and Methods

2.1 Study region

The AV, illustrated in Fig. 1b, is a 1.152 km^2 natural river basin located in the northeast of Colombia (see Fig. 1a). The territory contains ten cities, with Medellín as the largest city. The AV is in the central mountain range of the Colombian Andes, and its height ranges from 1300 masl (meters above sea level) in the valley to 2800 masl at the western mountaintop. By 2018, the population in the AV reached 3.73 million inhabitants, with a dense conurbation. Due to the accelerated urban expansion (Echeverri and Orsini, 2011; Salazar Hernandez et al., 2022), the increasing vehicular fleet (Corrales Espinosa et al., 2016), and the limited air pollutant dispersion, the average daily concentrations of $PM_{2.5}$, frequently exceed frequently exceeds national and international standards (WHO, 2021, 15 $\mu g/m^3$) in the valley, with . Across the AV, more than 60% of days in the year exceeding this limit these standards at most stations located in the urban areas of the AV (see Supplementary Table S1). Hence, the addition of external pollution sources has caused severe air quality episodes in the valley (SIATA, 2021).

The AV has two rainy periods responding to the latitudinal migration of the Intertropical Convergence Zone (ITCZ), with maximum precipitation during April and November and minimum during January and July. In the The transition period, i.e., February and March are , is characterized by persistent atmospheric stability and a thinner atmospheric boundary layer enhancing the accumulation of air pollutants (Herrera-Mejía and Hoyos, 2019). During this period of dry conditions, fires are more likely to spread in NSA, which affects air quality in the AV and neighboring regions (Mendez-Espinosa et al., 2019;

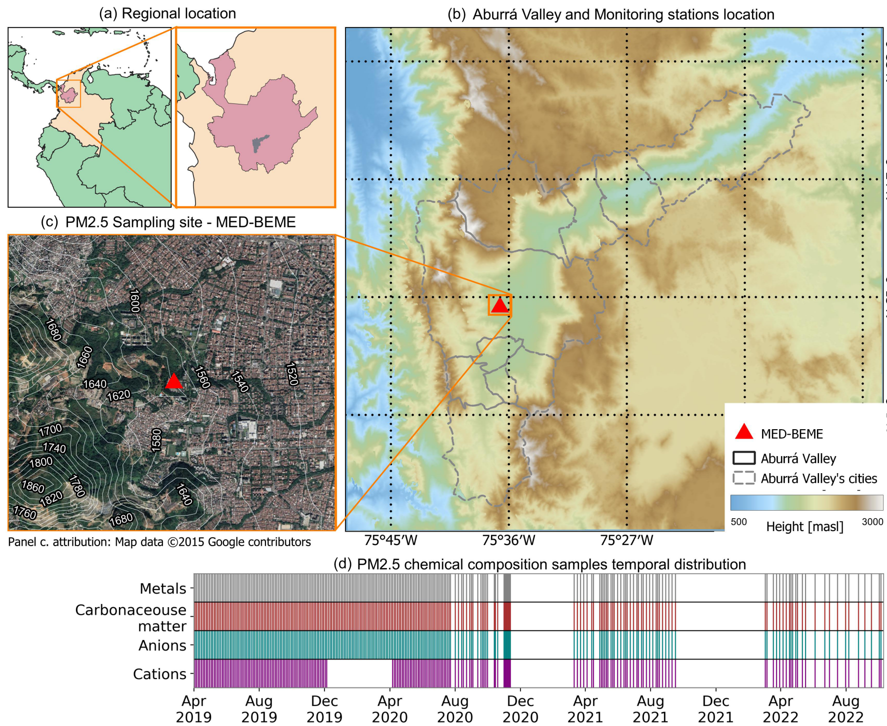

Figure 1. (a.) Regional location of the AV, (b.) Geomorphology of the AV and the distribution of the ten municipalities in black lines; (c.) Location of the $PM_{2.5}$ chemical sampling campaign station (MED-BEME), represented by the red triangle on (b) and (c.), and d. Temporal coverage of the $PM_{2.5}$ sampling campaign with white patches denoting no-sampling days.

Henao et al., 2021). These conditions modulate the intra-annual variability of $PM_{2.5}$ and PM_{10} (Mendez-Espinosa et al., 2019) . In this period, the transport and environmental local authority (Área Metropolitana del Valle de Aburrá) implemented special control on mobile sources implementes special mobile sources control, limiting daily vehicle use.

2.2 Data sources

2.2.1 $PM_{2.5}$ chemical campaign

A sampling campaign was conducted at an urban background station from April 3, 2019, to October 5, 2022, to characterize chemicals the aerosol composition on $PM_{2.5}$ filters. A total of 247 daily (noon-to-noon) $PM_{2.5}$ samples were analyzed and describedrecorded. While April 2019 to July 2020 represented an intense sampling campaign with samples every three days, the frequency of the surface site observations became less intense after July 23, 2020 (i.e. up to a maximum of 2 weeks during periods of routine sampling). However, there were two extended gaps in the campaign from November 2020 to mid-March 2021 and mid-September 2021 to March 2022. Despite the decrease in sampling frequency, the measurements still provide sufficient temporal coverage to get robust seasonal and annual information on aerosol concentration level and compositionin this studymass concentration and composition.

The samples were taken in Belén, Medellín, on the western slope of the valley by the laboratory GYGHAM , close to (Grupo de Higiene y Gestión Ambiental) from the Politécnico Jaime Isaza Cadavid. The sampling campaign was by MED-BEME, a station of the official air quality monitoring network (see Fig. 1c). Although some brick factories exist, the zone's The sector has a brick factory, but in general, the local economy is based on service and small businessbusinesses. The residential and closer brick factory areas areas and brick factory are around 250 m and 620 m from the station, respectively. According to Gómez-Marín et al. (2021), the primary sources of $PM_{2.5}$ in MED-BEME are the ceramic industry (22.2%), emissions from BB (21.9%), diesel (13.6%), gasoline combustion (12.8%), incineration (1.7%), and coal-fired boilers (16.3%).

The campaign considered minerals (Be, Na, Mg, Al, Si, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Mo, Ag, Cd, Sb, Ba, Hg, Pb), anions (F^-, SO_4^{2-}, NO_3^-, Cl^-), cations (K^+, Mg^{2+}, Ca^{2+}, Na^+), and carbonaceous matter species (OC1, OC2, OC3, OC4, OC5, Pyrogenic Carbon- PyC, EC1, EC2, EC3, EC4, EC5, EC6, OC, EC, C). In particular, cations were measured only in 207 of the 247 sampling days. Fig. 1d shows the temporal distribution of the samples throughout the study period. An exception for minerals is Note, that measurements of some minerals (e.g. Na, Mg, K, Se, Ca, and Hg, whose measurements were) finished in March 2021. The elements were analyzed from an 8"x10" quartz filter in a High-volume $PM_{2.5}$ ambient air sampler (Reference: TE-6070, TISH), following the Australian/New Zealand Standard 3580.9.14:2013 Method 9.14 (AS/NZS, 2013). An inductively coupled plasma mass spectrometry and thermal/optical transmission (TOT by NIOSH 5040) were used to measure mineralsand carbonaceous matter , and Ionic Distinct analytical methodologies were applied to determine the concentration of minerals, carbonaceous matter, and ions in the filters. An Inductively Coupled Plasma Mass Spectrometry (ICP-MS) methodology was used for minerals, a thermo-optical transmission (TOT) methodology for carbonaceous matter and an ionic chromatography (IC) was used for both anions for both anion and cations. The PM25 PM_{25} was additionally sampled by a Low-volume sampler (Reference: Wilbur TE-WILBUR - Tish) . Since these measurements followed following the reference method described by in the CFR 40 Appendix L Part 50 reference method suggested by the US-EPA (2011) and adopted by Colombian regulations (MinAmbiente-Colombia, 2010), the regulators (MinAmbiente-Colombia, 2010). The measured 24-hour $PM_{2.5}$ concentrations from the Low-volume are used for the positive matrix factorization Positive Matrix Factorization (PMF) statistical model. Further details on the techniques and the support of laboratory protocols are described in Gómez-Marín et al. (2021).

To complement the characterization of carbonaceous matter, the secondary organic carbon (SOC) was calculated using the elemental carbon trace methodology (Huntzicker et al., 1986). This method assumes that the organic carbon measured comes from the background, primary, and secondary sources and hence follows the relation described by Equations 1 and 2.

$$POC = OC_{back} + EC \times (OC/EC)_{pri} \quad (1)$$

$$SOC = OC_{Total} - POC \quad (2)$$

Where POC represents the primary organic carbon, OC_{back} is the background OC, OC_{total} is the consolidated OC measured in the campaign, and $(OC/EC)_{pri}$ is the primary ration between OC and EC.

Equation 1 describes a linear relation of primary carbon matter ~~compositions~~composition. The linear model was calculated using the 20^{th} percentile OC and EC concentrations, aligned with the approach of Yao et al. (2020); Lin et al. (2009). The process was repeated for each ~~hydrologic trimester~~ season (DJF, MAM, JJA, and SON) due to the influence of weather conditions in the region. The resulting slope is interpreted as $(OC/EC)_{pri}$, and the intercept is the OC_{back}. After calculating ~~the~~ $(OC/EC)_{pri}$ and OC_{back}, Equation 1 was used for calculating POC every sampling day ~~POC using the sampled~~ using the measured EC. Then, the SOC was obtained with Equation 2.

This study involved creating four different linear regression models (Equation 1) for the two ~~yearly~~ dry (June-August and December-February) and two rainy (March-May and September-November) periods. The square of correlation coefficient (R^2) showed high performance of the models with values ≥ 0.95.

2.2.2 $PM_{2.5}$ concentrations

In addition to the campaign data, for the station MED-BEME, we downloaded hourly $PM_{2.5}$ concentrations from the official air quality monitoring network operated by the local early-warning system – SIATA (Sistema de Alerta Temprana de Medellín y el Valle de Aburrá, https://siata.gov.co/). From this, the $PM_{2.5}$ concentrations from April 2019 to October 2022 at the MED-BEME station coincided with the period and site of the chemical sampling campaign (illustrated in Fig. 1b). Daily average concentrations were calculated matching the sampling schedule (noot-to-noon). Only days with at least 75% of the hourly data were processed. Within our study period, we derived valid daily average concentrations for 96.7% of the days investigated.

We found a good agreement between the campaign (low-vol sampler) instrument and the official MED-BEME automatic station. For the study period, $PM_{2.5}$ concentrations from the automatic instrument had a minor overestimation against the reference method with a mean bias error of -0.76 $\mu g/m^3$. The corresponding mean absolute error (MAE) was 21.5%. For $PM_{2.5}$ measurements, the low-volume as a reference method provides better precision and accuracy than the MED-BEME sensor (Tasić et al., 2012), which follows ~~equivalent methods~~an equivalent method. Despite this, the official sensor provides continuous measurements that are used in this study for more robust comparisons. Regarding temporal variability, the Pearson correlation coefficient was 0.84, highlighting good consistency between them.

2.2.3 CAMS Reanalysis dataset

The European Centre for Medium-Range Weather Forecasts (ECMWF) provides atmospheric composition datasets through the Copernicus Atmospheric Monitoring Service (CAMS; Inness et al., 2019). For this research, we utilized the ECMWF Atmospheric Composition Reanalysis-4 (EAC4) dataset. It is a global atmospheric chemistry model, simulating a range of key tracers of composition, which uses a 4-dimensional variational data assimilation to assimilate satellite retrievals of, e.g., aerosol optical depth (AOD), carbon monoxide (CO), nitrogen dioxide (NO_2) and ozone (O_3) (Inness et al., 2019).

CAMS simulates different aerosols, which ~~complete~~ can be summed to determine the total AOD at 550 nm~~and different gases~~. To identify considerable loads from ~~biomass burning (BB)~~BB, dust, and volcanic aerosols, respectively, this study considers two AOD ~~fractions~~types: dust AOD (Du-AOD) and organic matter (OM-AOD), in addition to ~~the~~ total column sulfur dioxide ($TCSO_2$). The products were downloaded daily at the original 3-hourly resolution and then resampled for local noon-to-noon periods aligned with the sampling campaign schedule. Only ~~pixel information over the sampling site~~ reanalysis information for the sampling location was used.

2.2.4 Meteorological data

Meteorological data come from the ECMWF Reanalysis v5 (ERA5; Hersbach et al., 2020). Fields of zonal and meridional winds, temperature, and moisture were obtained at temporal and spatial resolutions of 3 hours and 0.25~~°~~ $^{\circ}$ (approximately 27 km), respectively. ERA5 data were up-sampled to a daily frequency to guarantee consistency with the chemical sampling. Due to poor performance ~~in representing~~ of reanalyses to simulate precipitation in the region ~~by reanalysis~~ (Posada-Marín et al., 2019), satellite-based fields were retrieved from the Global Precipitation Measurement (GPM) daily final precipitation product (Huffman et al., 2019) with a spatial resolution of 0.1~~°~~$^{\circ}$.

Further, ~~a back trajectories dataset was built from~~ back trajectories were exploited to estimate pollutants arriving at the chemical sampling point ~~.~~ in the AV (see Fig. 1c). 8-day trajectories starting at heights of 800 hPa, 750 hPa, and 700 hPa were run every 3 hours with a time step of 3 hours. The ~~model~~ basic model was created with SIATA and has been used in other investigations like Hoyos et al. (2019) and Pérez-Carrasquilla et al. (2023). This model only uses wind data from the ~~products~~ product's 4h-daily pressure levels, U-wind, V-wind, and omega (with a spatial resolution of approximately 2.5^{o}) from NCEP-NCAR Reanalysis 1 (https://psl.noaa.gov). The model follows Equation 3. for estimating every back position.

$$X(t-\Delta t) = X(t) - V(X,t)\Delta t \tag{3}$$

Where X is the location with vertical and horizontal coordinators, V is the wind vector, and t is the time.

2.3 Identification and tracking of aerosol events

2.3.1 Identification of LRT events

Despite the selected CAMS products covering the whole atmospheric columns and not only the surface, CAMS has a high predictive capability for $PM_{2.5}$ in the AV (Pérez-Carrasquilla et al., 2023). Furthermore, ~~its products relatively reasonably reproduce~~ the CAMS' products reasonably capture $PM_{2.5}$ tendencies and extreme events in the territory (Casallas et al., 2022). Therefore, the magnitude of CAMS products OM-AOD, Du-AOD, and $TCSO_2$ in the AV were utilized to identify possible BB-LRT, Dust-LRT, and Volcanic-LRT events, respectively.

To help identify $PM_{2.5}$ days that were subject to ~~long-range transport~~LRT events, the time series of the CAMS data sets were standardized. Here, for each day, the time-series average (mean) was subtracted from the daily value and then normalized by the time-series standard deviation (i.e., 1 σ). This then allowed for the identification of LRT events using a range of subjective thresholds and the quantification of the corresponding $PM_{2.5}$ concentrations and composition. The thresholds investigated ranged from 0.8 to 2.0 σ, but in this study, we present the results for thresholds of 0.8, 1.5, and 2.0 σ. A 7-day rolling window was used to accurately identify prolonged and intense periods of LRT events. Within this window, at least 4 days had to have values above the respective thresholds to be classified as a LRT event. We subjectively chose ~~4 days~~ 4-days of elevated values due to the sampling frequency of the campaign. Here, campaign temporal sampling was $\geq$ 3 days, so these criteria were required to get representative samples of the aerosol ~~characterization for the PMF analysis~~composition for the chemical characterization of the sources.

~~Observe that continuous days marked as peaks are possible for longer-lasting events. Finally, for each peak event (DtE), the surrounding days, relative to the peak event centered in the~~ After identifying potential LRT events, the center day of the 7-day ~~window (i.e., DtE_0), were labeled. For instance, the~~ window was marked as the peak of the LRT event. Then, every day was labelled according to their distance to the peak of the LRT event as n days to the event (DtE_n). For this, the peak of the event is labled as DtE_0 and the LRT events ranged from ~~three days~~ 3-days before (DtE_{-3}) to ~~three days~~ 3-days after (DtE_3) the peak event. The days outside of ~~three-day~~ the 3-day windows around a DtE_0 are ~~generally~~ typically identified as no-event days ~~, and no significant/lasting event affectation is supposed.~~ and assumed to be independent of LRT events. Note that this methodology allows continuous days to be marked as peaks for longer-lasting events.

Although the process described above was run for every event and threshold, ~~further assessments~~ the results in this study are based on 0.8 σ for BB-LRT and 1.5 σ for Dust-LRT and Volcanic-LRT, as is described in Section 3.1.

2.3.2 Regional meteorological analysis

A regional analysis of meteorological fields was performed during ~~pollution~~ the LRT events to identify ~~conditions that favor~~ atmospheric conditions that favored aerosol transport from the sources of interest. ~~We computed anomalies for each pixel of the meteorological fields by~~ Here, we derived the anomalies for multiple meteorological variables by taking the values of each pixel and subtracting the average value of the corresponding month~~from the original values in the study period. Subsequently~~. Then, composites during days with ~~events (~~different LRT events (i.e. days in the range of DtE_{-3} to DtE_3) were ~~performed with~~ calculated for both meteorological fields and anomalies~~for each source~~. This approach ~~helps reveal particular~~ helped to reveal particular meteorological conditions during days with aerosol LRT events. The results are also supported by analyzing air masses arriving at the AV using back-trajectories during the LRT events. The number of times trajectories pass through a ~~pixel was counted to estimate a percentage indicating the influence area~~grid box was counted and the percentage of occurrences calculated yielding an estimate of the probability that a particular grid box will experience a LRT event for each type.

2.3.3 Influence on local $PM_{2.5}$ concentrations

The local measurements of $PM_{2.5}$ were used to assess the influence of CAMS-based identified LRT events on air quality in the AV. The daily $PM_{2.5}$ datasets were labeled with the days to the closest event peak (DtE) to compare concentrations before and after the events. The days before the event constitute from 15 to 4 days before the event peak (DtE_{-15} to DtE_{-4}), while the days after the event peak consisted of 4 to 15 days after the event peak (DtE_4 to DtE_{15}). These ranges were determined to ~~have~~ ensure enough data from the chemical campaign ~~on~~ from similar weather conditions for comparison between the days affected by the LRT of aerosols and the days before and after. We used the Mann-Whitney U test to compare these periods. The null hypothesis is denied with the confidence of 90% (p-value ≤ 0.1) and 95% (p-value ≤ 0.05).

The COVID-19 lockdown period (April 1st to June 1st, 2020) was filtered from our record to avoid perturbation out of standard emission patterns in the region.

2.3.4 PMF and $PM_{2.5}$ chemical composition

The ~~Positive Matrix Factorization (PMF)~~ PMF is a receptor model developed by the EPA (Environmental Protection Agency) to analyze water and air samples. The model expresses observations of $PM_{2.5}$ as a sum of the contributions from a number of source profiles (non-time dependent). In this case, we are using the PMF model and our campaign measurement data of $PM_{2.5}$ chemical composition and $PM_{2.5}$ concentrations to identify the dominant sources (e.g., coal combustion, vehicular emissions, secondary pollution) during ~~different LRT classed events~~ each LRT event (i.e., BB, dust or volcanic LRT events). For the AV, 5 to 7 factors (i.e., sources) have been suggested (see Gómez-Marín et al. (2021)).

The PMF is mathematically expressed as (Paatero and Tapper, 1994):

$$X_{ij} = \sum_{k=1}^{N} g_{ik} f_{kj} + e_{ij} \tag{4}$$

The objective of PMF is to determine the values of g_{ik}, f_{kj}, and N (i.e. the number of factors) that best reproduce X_{ij}. Here, X represents the measurement data for sample i (daily temporal resolution) and chemical component j. An inversion approach is used for this matrix problem, exploiting an iterative scheme to converge on the solutions for g_{ik} and f_{kj}. In the factorization problem, f_{kj} contains the concentration of each chemical component (j) in the unit profile for the factor k (i.e. $PM_{2.5}$ source). The matrix $g_{i,k}$, the contribution factor, defines how much of the profile is counted in the total concentration in day i. $e_{i,j}$ contains the residual for each compound/sampling day. Outputs from the PMF include statistical metrics that help to evaluate the model performance, such as the correlation coefficient (R^2), the recuperated mass (% RM), and the objective function value (Q). The PMF adjusts g_{ik} and f_{kj} to minimize the function Q in Equation 5 (Paatero, 1997).

$$Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \left[\frac{X_{ij} - \sum_{k=1}^{N} g_{ik} f_{kj}}{\mu_{ij}} \right]^2 \tag{5}$$

Where μ_{ij} contains the uncertainty compound/sampling day.

The model uses two input uncertainties. The first is the species uncertainty (μ_{ij}), which does not change in the process and is according to the ~~method unique~~ unique method used for every species ~~sample~~sampled. In this study, μ_{ij} considers the measurement and analytical errors, comprising uncertainties of the volume of air processed, filter area, and species mass, in addition to the uncertainty of the detection limit of the ~~measurer~~ measurement method. The last one represents the analytical uncertainty and was calculated following Noris and Duvall (2014) by using the ~~methods~~ corresponding method's detection limit suggested by Eugene Kim and Qin (2005). However, for $PM_{2.5}$, we followed the approach of Eugene Kim and Edgerton (2003) who set μ_{ij} (where j represents $PM_{2.5}$ for day i) greater than the daily $PM_{2.5}$ concentration (i.e., three times the concentration in this study). The second uncertainty in μ_{ij} is recognized as the "Extra modeling uncertainty". It is added to the model as a percentage to cover other inherent errors, such as variations of source profiles and chemical transformations in the atmosphere. A range from 10% to 16% was tested for the models of every kind of LRT event; this aligns with PMF for each type of LRT event, which aligns with the approach of other studies (Callén et al., 2009; Shin et al., 2022; Salim et al., 2019).

During the modeling process, species can be classified as "strong", "weak," or "poor" based on individual statistics. The "weak" category triples the uncertainty μ_{ij}, and the "bad" category excludes the species from the model. Two statistics are key for categorizing the species. First, the signal-to-noise ratio (S/N) represents overall uncertainty per concentration. S/N<0.5 is generally classified as "bad" because the uncertainty surpasses two times the concentrations concentration (Noris and Duvall, 2014). Second, the regression diagnostic where R^2 informs provides information about the linear relation of every species' observed and modeled concentrationrelationship between the observation and modeled concentrations for each species.

The model convergence evaluation statistics considers this classification with the $Q/Q_{expected}$ ratio. $Q_{expected}$ is the theoretical value of Q (Equation 5), expressed as Equation 6.

$$Q_{expected} = (n \times m_s) - ((N \times n) + (m_s \times N)) \quad (6)$$

where n is the number of sampling days, and m_s is the number of "strong" species used for modeling (Noris and Duvall, 2014).

For the PMF we anticipate small datasets since the target events may not significantly impact the entire We expected relatively small temporal samples (i.e. number of days) of the measured pollutants for each LRT type in the PMF model since the targeted LRT events would not cover the majority of the days during the $PM_{2.5}$ chemical campaign. Nonetheless, multiple studies with PMF samples ranging from 14 to 30 have reported useful and meaningful results (Yu et al., 2015; Haghnazar et al., 2022; Via et al., 2022). As the sample dataset decreases, rotational ambiguity caused by infinite valid solutions strongly affects the results and increases overall uncertainty (Manousakas et al., 2017). To mitigate the error, the software EPA PMF v 5.0 allows for estimating the effect of random errors and rotational ambiguity in the dataset using bootstrapping (BS) and Displacement (DISP) tools. While BS evaluates random errors by performing 100 runs with randomly relocated blocks of observation of the original dataset, DISP focuses on indicating rotational ambiguity by adjusting up and down all values in the factors profilerestricted by . This is restricted to 4 allowed changes in the calculated permitted changes when calculating Q (dQmax) and monitoring major factors factor swaps (Noris and Duvall, 2014).

Additionally, constraining the base run can improve the solution when data is limited by reducing the rotational space (Dai et al., 2020). The PMF software has the functions to "pull down maximally," "pull up maximally," "set to zero," and manually set the profile concentrations. While the first two options are soft constraints, the third and fourth are hard constraints and require a high level of confidence in the magnitude of the profile contributions. For this study, only soft contains are contemplated. The constraint increases in the final Q value, which should be less than 5% (i.e., %dQ < 5%), the recommended maximum change (Noris and Duvall, 2014). For this study, the %dQ was set by default to <0.5%.

Table 1. Statistics of OM-AOD, Du-AOD, and $TCSO_2$ representing BB-LRT, Dust-LRT, and Volcanic-LRT

Threshold	Events stats.	OM-AOD	Du-AOD	$TCSO_2$
0.8 σ	Samples	31	31	59
1.5 σ	Samples	18	19	32
2.0 σ	Samples	10	4	21
0.8 σ	Limit	0.22	0.01	0.70mg/m^2
1.5 σ	Limit	0.28	0.02	0.87mg/m^2
2.0 σ	Limit	0.32	0.02	0.99mg/m^2
0.8 σ	Average	0.25	0.02	0.78mg/m^2
1.5 σ	Average	0.29	0.02	0.88mg/m^2
2.0 σ	Average	0.32	0.03	0.97mg/m^2

"Samples" represent the number of sampling days available for the PMF characterization. Each threshold limit defines the calculated absolute magnitude. Note, AOD is a dimensionless quantity.

Due to missing data, all species except cations are included in the model. Only total OC and EC from carbonaceous matter are included for Duts-LRT and Volcanic-LRT events since comprising all carbon species could particularly weigh those and overshadow ~~trace~~ tracer elements for the sources. For those LRT events, the potential run period is reduced until April 2021 since the tracer minerals Na, K, and Mg were not measured afterwards.

The factor interpretation of this study was primarily supported by the source characterization made by Gómez-Marín et al. (2021) for the studied station.

A final ~~element comparison is made between the days of events with a positive relative contribution~~ comparison of the characterized compounds was conducted, focusing on the days when a positive contribution occurred for the LRT events ($g_{i,k}$>0) and the days categorized as before (DtE_{-15} to DtE_{-4}) and after events (DtE_4 to DtE_{15}).

3 Results

This section is divided into four subsections. The first subsection ~~shows possible~~ identifies potential aerosol events from BB-LRT, Dust-LRT, and Volcanic-LRT. The second is focused on the temporal and regional characterization of ~~the~~ these events, including a description of the associated meteorological patterns and possible ~~sources of aerosols~~ aerosols sources. Finally, the third and fourth subsections locally assess the identified events using ground-level $PM_{2.5}$ ~~concentration~~ concentrations and chemical compositions, respectively.

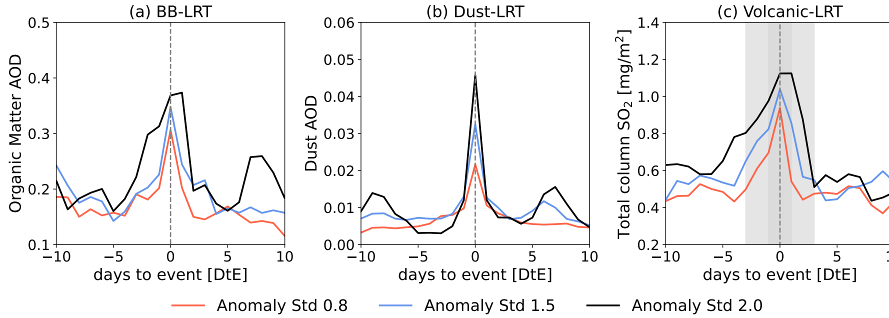

Figure 2. Average magnitudes of OM-AOD (a~~.~~), Du-AOD (b~~.~~), and $TCSO_2$ (c~~.~~) concentration around the events. Different ~~colors~~ colours represent different anomalies. The shadowed region delimits the event. Du-AOD is on a scale that is ten times lower than OM-AOD.

3.1 Identification of LRT events

The products OM-AOD, Du-AOD, and $TCSO_2$ from the CAMS ~~reanalysis~~ reanalyses were used to identify aerosol from BB-LRT, Dust-LRT, and Volcanic-LRT, respectively. ~~Tab.~~ Table 1 shows different statistics about the identification for each ~~kind~~ type of event using the threshold of 0.8 σ, 1.5 σ, and 2.0 σ. The ~~intensest~~ most intense events reached maximum magnitudes of 0.66 for OM-AOD (March 30, 2020), 0.11 for Du-AOD (June 24, 2020), and 1.96 mg/m^2 for $TCSO_2$ (September 10, 2022). ~~These specific days~~On these days, the products exceeded the average ~~values for identified~~ magnitudes for the target events more than twice for OM-AOD and $TCSO_2$ and more than five times for Du-AOD. Notably, OM-AOD reached 77% of the total AOD in the ~~intensest~~ most intense event. Regarding the variability, all identified events showed high variability with a standard deviation around 42%, 90%, and 35% of the average for OM-AOD, Du-AOD, and $TCSO_2$, respectively.

~~Fig.~~ Figure 2 shows the average behavior of each variable centered around the event peak for the threshold of 0.8 σ, 1.5 σ, and 2.0 σ. The events exhibit a clear peak centered on DtE_0 for most of the considered ~~threshold~~thresholds. However, events identified with 2.0 σ exhibit ~~noisy behavior~~ noiser behaviour around the peak for BB-LT and volcanic-LRT. In particular, OM-AOD exhibits a second peak after the event peak. This could be caused by either the smaller sampling size or ~~high~~ higher aerosol loads that do not meet the duration criteria for the threshold. Besides, the strictest thresholds (2.0 σ) result in a considerable reduction of studied events (see ~~Tab.~~ Table 1), which is critical for the assessment of the $PM_{2.5}$ chemical composition.

~~Based on the study's objectives, only one identification threshold was selected for each event, allowing the investigation of more accurate identification indexes~~For each type of LRT transport event, we selected a single σ threshold value. Based on the results and literature review about the expected frequency of each type of event for the city (Mendez-Espinosa et al., 2019; SIATA, 2021), we selected events that exceed 0.8 σ for BB-LRT and 1.5 σ for Dust-LRT and Volcanic-LRT. Anomalies over 2.0 σ significantly limited the number of days for the analysis, whereas 0.8 σ probably ~~oversamples~~ over sampled volcanic and dust aerosol load events ~~, whose affectation is recorded as infrequent~~since these are recorded as extremely infrequent events. Moreover, the selection ensures ~~yearly representativity~~ annual represenation for each event (see supplementary ~~Figure~~ Fig. S2). BB-LRT, Dust-LRT, and Volcanic-LRT ~~were represented~~ events were all detected during the complete study period, ~~at least~~

~~for three years~~ occurring in at least 3-years of the record each. These events ~~cover~~ typically occurred for 13%, 8%, and 13% of days in the year, respectively. With this selection, the sampling campaign can represent BB-LRT, Dust-LRT, and Volcanic-LRT events in the PMF with 31, 19, and 32 samples.

3.2 Regional analysis of events

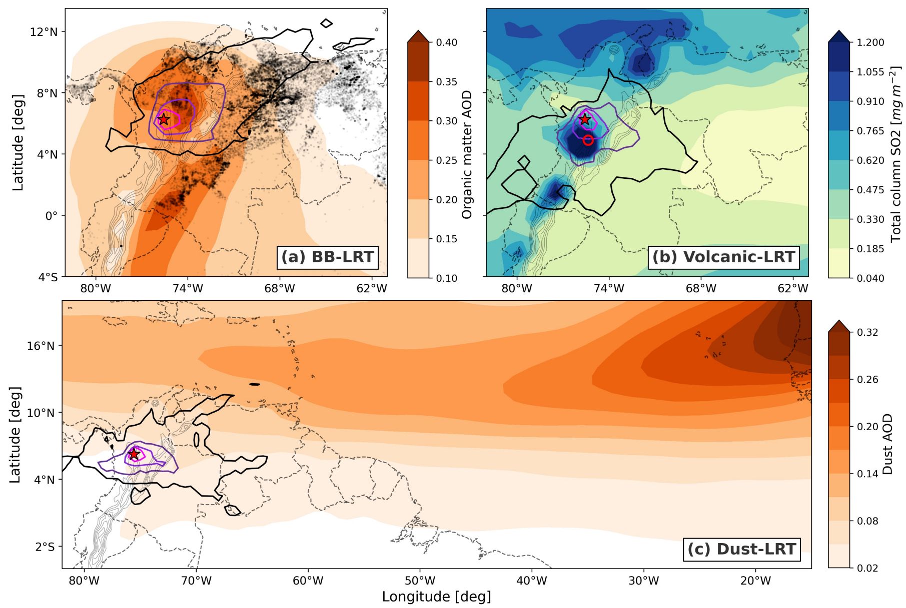

Figure 3. Mean spatial distribution of OM-AOD (a~~.~~), $TCSO_2$ (b~~.~~), and Du-AOD (c~~.~~) for days with each type of LRT event. Pink-to-black contours enclose the regions from which 20, 10, 5, and 1% of air masses arrive at the AV according to the back-trajectories. Gray dots in a. show MODIS-retrieved hotspots associated with fires with a 70% of confidence. The red star marks the location of the AV, while the red circle in the upper right panel is the location of the Nevado del Ruíz Volcano. The black contours show the terrain elevation from 1500 to 5000 masl.

Regional analysis was conducted to establish the meteorological conditions that may ~~favor~~ favour air pollution LRT events and identify potential aerosol sources. The average spatial distribution of the identified AOD-OM, Du-AOD, and the $TCSO_2$ during the aerosol LRT events is shown in Fig. 3, with the integrated back trajectories of air masses arriving at the AV. OM-AOD values above 0.2 occur over most of Colombia during BB aerosol events (Fig. 3 a), consistent with a high number of fire hotspots in northern South America (Venezuela and Colombia). In particular, the back trajectories during these days suggest that a large part of the air masses arriving at the AV (10 to 20 %) come from the northeast, where OM-AOD exhibits its ~~maximums~~ peak values (exceeding magnitudes of 0.35).

Volcanic-LRT (Fig. 3 b) shows high SO_2 values (over 1.2 $mg\,m^{-2}$) in the atmospheric column above the Nevado del Ruiz volcano (red hollow circle). Additionally, another two spots are evident: the first one, ~~toward~~ towards southwestern Colombia, could be related to another degassing volcano called the Galeras, whereas the other one is on Maracaibo Lake, with relatively high values are possibly associated with oil extraction (see Fioletov et al. (2016)). According to the back-trajectories, during Volcanic-LRT events, around 10% of the air masses arriving in the AV come from the southeast, where the Nevado del Ruiz hotspot exists. A smaller fraction of air masses (1%) arrive from the hotspot in the country's southwest (i.e., Galeras volcano) in the southwest of Colombia. Similarly, a clear dust propagation pattern is evident there is clear propagation of dust aerosol from the Sahara desert towards the Caribbean and, to a lesser extent, NSA (see Fig. 3 c). While air masses for BB-LRT and Volcanic-LRT have different directions, most air masses that reach the AV during Dust-LRT events come from the east, showing consistency in the analysis.

Despite the short study period, the identified events suggest a marked annual cycle (Fig. 4). During March, 50.0% of studied days represent the BB-LRT. April and February also showed high proportions of this eventshow high proportions BB-LRT events, with 43.3% and 24.8%, respectively. Concerning the dust events, two maximums seasonal peaks are observed, April and July, with a proportion of 23.3% and 23.4%. The distribution for volcanic-LRT ranges from June to September, with a clear peak in August with a frequency of 49.2%. Additionally, as shown in Fig. 4, external events overlap in some cases. The higher intersections observed were for largest occurrence of combined LRT events was observed in April comprising BB-LRT and Dust-LRT events, while BB-LRT and Dust-LRT in April and for BB-LRT and volcanic-LRT events showed substantial overalp in September. Here, where days were defined as both, e.g., BB-LRT and Dust-LRT, they were added to the joint classification (i.e., BB-LRT & Dust-LRT) and were not included in the respective singular classifications. Overall, the annual cycle of these frequencies LRT event occurrences and the local $PM_{2.5}$ concentration show the event's possible influence on shows that the influence of LRT on local on measured $PM_{2.5}$ is non-linear.

Although LRT events display a marked seasonality, a significant percentage of days in each month have a negligible impact experience negligible impacts from LRT events (see Fig. 4), suggesting that intraseasonal . This suggests that intra-seasonal variations are also relevant in explaining the occurrence of these events. To elucidate isolate specific meteorological patterns during LRT events days, Figs. 5 and 6 present composites of meteorological variables for the low (800 to 700 hPa) and mid (600 to 400 hPa) troposphere for the different kinds of events. The Andes mountain range generally causes a large spatial variability in the low-level wind field. The winds blowing from the east of the Andes cross the mountain range through two zones with relatively low altitudes (hereafter mountain passes), which are demarcated with white boxes (see Figs. 5 a, b, c).

Anomalous dry winds of around 1 m/s originate from the north during BB-LRT events (Fig. 5 d), connecting the AV with the high OM-AOD region. Additionally, winds blowing from Venezuela, where multiple BB hotspots were identified, reach northern Colombia through the mountain pass located north of the Andes, at the border between Colombia and Venezuela (see Fig. 5 a). Low-level winds during LRT events are in agreement with the back trajectories of Fig. 3 a. A reduction of 2 to 3 m/s in the intensity of mid-level winds (between 5°N and 8°N; see Figure Fig. 6 a, d) was also observed along with less rainy conditions (anomalies of around -2 mm/d) in northern Colombia.

For Dust-LRT events, low-level winds flow across the two mountain passes in the Andes (Fig. 5 b) with increased easterly winds north of Colombia and northeasterly winds east of the Andes (Fig. 5 e), representing a dry flow from the Atlantic Ocean where Du-AOD is higher. Figs. 6 b and e, show that mid-level winds have notable eastward direction with anomalous winds coming from the Caribbean and less rainfall a rainfall reduction between 3 to 4 mm/d in NSA.

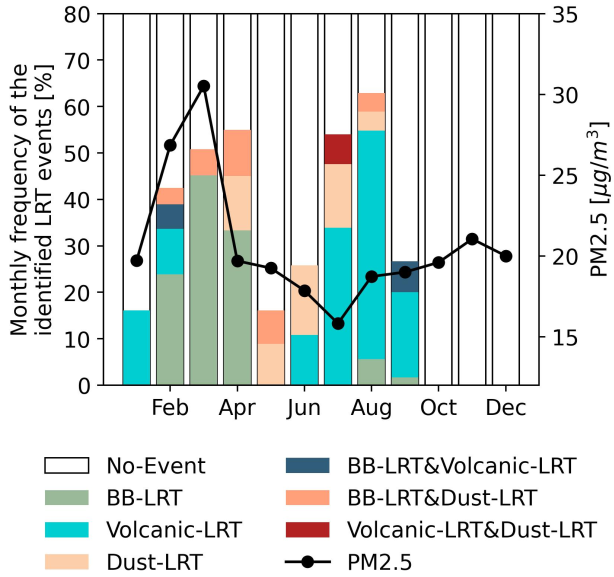

Figure 4. Monthly frequency of days with BB-LRT (green bars), Dust-LRT (wheat bars), and Volcanic-LRT (blue bars) events as identified from the CAMS reanalysis. Overlapped events are depicted in dark blue (BB and Volcanic), orange (BB and Dust), and red (Volcanic and Dust) bars since different LRT events can happen at the same time. White bars represent the frequency of days without LRT events, while the black line shows the monthly average $PM_{2.5}$ concentration ($\mu g/m^3$) for the MED-BEME station.

Finally, days with volcanic-LRT events are characterized by southeasterly winds ~~blowing~~ traveling toward the AV (see Fig. 5 c) from the Nevado del Ruíz volcano region. ~~These winds may be favored by anomalous southeasterly winds of more than~~ Here, the southeasterly wind anomalies are 3 m/s ~~crossing~~ yielding stronger transport across the southern mountain pass, as shown in Fig. 5 f, along with drier air reaching the AV. Less rainy conditions are also present in the Colombian Andes (Fig. 6 f), particularly between the AV and the Nevado del Ruíz volcano where a reduction in rainfall of around 3 mm/d is observed. ~~Noteworthy~~Overall, the three types of events are accompanied by less rainy and drier atmospheric conditions in northern Colombia, Venezuela, and the Andes, respectively~~, with rainfall anomalies greater~~ . Here, rainfall and specific humidity anomalies are typically larger (in absolute terms) than -4 mm/d and ~~specific humidity anomalies up to~~ -1 gkg^{-1}, respectively. These results ~~allow us to~~ highlight the importance of precipitation ~~to enhance or even trigger the aerosol LRT events~~ control on aerosol LRT to the AV (i.e., lack of precipitation aids the LRT of aerosols).

3.3 $PM_{2.5}$ concentrations change

After identifying ~~possible~~ LRT events and the meteorological conditions ~~favoring them, a local analysis was performed to connect and validate the different events' influence on~~ favouring them, an analysis of local surface $PM_{2.5}$ was undertaken to understand the influence of LRT on the AV's ~~local~~ air quality. ~~The daily concentration of $PM_{2.5}$ reasonably responds to the~~

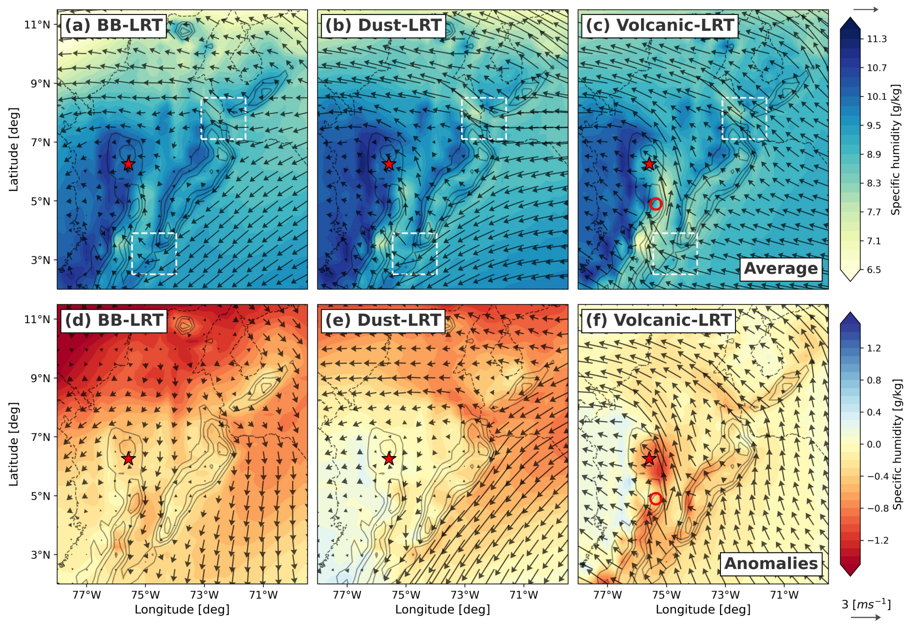

Figure 5. Meteorological composites for low-level (800 to 700 hPa) winds and specific humidity during days with (a~~.~~) BB-LRT, (b~~.~~) Dust-LRT, and (c~~.~~) volcanic-LRT events. d, e, f: as upper panels but for anomalies. A red star marks the Aburrá Valley, and the red circle in the upper right panel is the Nevado del Ruíz volcano. The white rectangles are two mountain passes in the Colombian Andes. The black contours show the terrain elevation from 1500 to 5000 masl.

~~BB-LRT identified by OM-AOD. Fig.~~ Figure 7 compares the $PM_{2.5}$ concentrations before (DtE_{-15} to DtE_{-4}), during (DtE_{-3} to DtE_3) and after (DtE_4 to DtE_{15}) each event. BB-LRT had the highest $PM_{2.5}$ increments, especially at DtE_0. 86.7% of values during the peak day exceed the average $PM_{2.5}$ for the study period (20.25 $\mu g/m^3$). ~~Besides, over~~ Over half of $PM_{2.5}$ concentration during DtE_0 exceeds twice the WHO (2021) guidelines for daily concentrations (15 $\mu g/m^3$). ~~On~~ At DtE_0, the $PM_{2.5}$ average concentrations (31.48 $\mu g/m^3$) were significantly higher than the surrounding days (p-value $\leq$ 0.05), according to the Mann-Whitney U test. Similarly, the event days before the peak (DtE_{-3} to DtE_{-1}) had significantly higher concentration compared to days before and after the identified BB-LRT events (p-value $\leq$ 0.05). In contrast, concentrations following the event peak are similar to the average magnitude, indicating a sudden decrease.

~~As for~~ While there is a substantial BB-LRT ~~, the peak of the events~~ signature in the local $PM_{2.5}$ measurements, the response is more subtle for Dust-LRT ~~showed higher concentrations than days before~~ and Volcanic-LRT events. For Dust-LRT and Volcanic-LRT events, the median $PM_{2.5}$ concentrations during the event (Dte_{-3} to Dte_3) are actually lower than the campaign average (20.25 $\mu g/m^3$). However, for Dust-LRT events, the $PM_{2.5}$ concentrations are significantly (p-value $\leq$ 0.1) ~~. On the other hand, the increase of PM~~higher than before the event. This is likely due to the large variability in the Dust-LRT event data with the Q_3 quartile peaking over 35 $\mu g/m^3$, which was lower before the event (especially Dte_{-7} to Dte_{-4} reaching approximately 28 $\mu g/m^3$). For Volcanic-LRT events, there is a small $PM_{2.5}$ ~~due to~~ gradient between days before and during

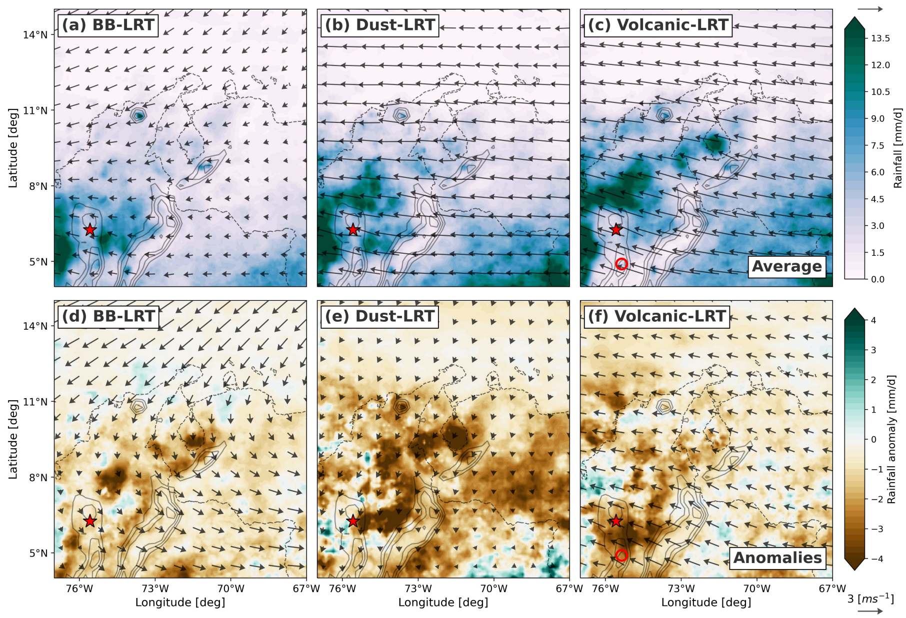

Figure 6. Meteorological composites for mid-level winds (600 to 400 hPa) and rainfall during days with (a) BB-LRT, (b) Dust-LRT, and (c) Volcanic-LRT events. d, e, f: as upper panels but for anomalies. The red star marks the Aburrá Valley, and the red circle in the upper right panel is the Nevado del Ruíz volcano. The black contours show the terrain elevation from 1500 to 5000 masl.

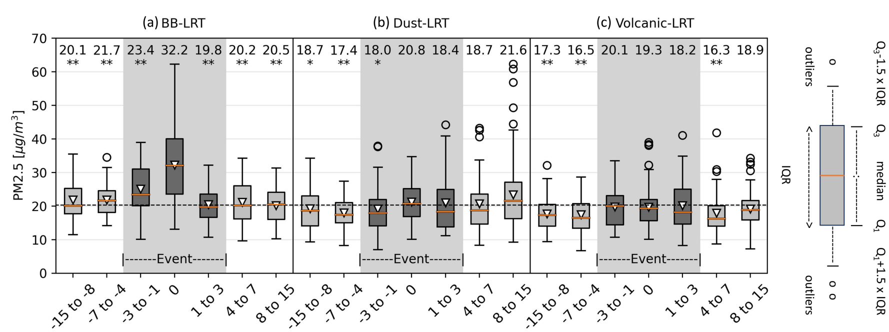

Figure 7. $PM_{2.5}$ concentrations before, during and after (a~~.~~) BB, (b~~.~~) dust and (c~~.~~) volcanic aerosols events. The chart's top presents the median concentration of each dataset, along with the significance level compared with the peak of the data, ≤ 0.1 (*) or ≤ 0.05 (**). The shadowed darker boxes represent the events. The down triangles represent the average concentration for each day range, and the grey dashed line represents the total study period average. The boxes are limited by the first and third quartile, Q_1 and Q_2

the LRT event, which is not dissimilar to that of the Dust-LRT events. Although, the tighter range in the variability (i.e. lower Q_3 values) yields a non-significant difference despite the larger Volcanic-LRT ~~was consistent at the beginning of the events, not just during the peak~~event median values (e.g. by approximately 2-3 $\mu g/m^3$). After the ~~events, the~~ peak Dust-LRT event, $PM_{2.5}$ ~~concentration for Dust-LRT remains at similar~~ in the Dte_4 to Dte_7 window decreases to pre-event levels. However, ~~outliers~~

are observed, particularly the Dte_8 to Dte_{15} shows a similar median value to the Dust-LRT peak (DtE_0) at approximately 20 $\mu g/m^3$ and has a larger data range (i.e. Q_3 peaks at approximately 43 $\mu g/m^3$). These larger than expected values occurred in March 2020, which was a period strongly affected by BB-LRT . On the other hand, the concentration of events. For days (Dte_4 to Dte_7) after Volcanic-LRT events, we find that $PM_{2.5}$ right after the peak of Volcanic-LRT presented significantly lower concentrations concentrations are significantly lower (p-value ≤ 0.1), contrasting with the subsequent days (Dte_8 to Dte_{15}). Overall, it is clear that the BB-LRT event has a higher impact events have a larger influence on $PM_{2.5}$ concentration to concentrations in the AV than Dust-LRT and Volcanic-LRT.

3.4 PMF and chemical composition change

The PMF models of the BB-LRT and Volcanic-LRT presented a good fit, and all residuals are normally distributed; however, for the Dust-LRT, some components' residuals exceed the recommended range of -3 to 3 (Noris and Duvall, 2014), so three 3 samples were excluded due to outlier data. In general, the three final selected PMF models showed a anomalous data. Overall, the 3 final PMF models selected for the analysis highlighted good performance, meeting the acceptance criteria with a reconstructed mass (RM) an RM in the range of 80 to 120 and an R^2 coefficient greater than 0.8 (Noris and Duvall, 2014). The results show a good convergence, where the $Q/Q_{expected}$ ratio for the selected base models were was close to 1, a determinant parameter for the crucial parameter for determining the correct number of factorsand the component categorization. A summary of the models' statistics is given in Table 2.

In evaluating errors of the PMF, none of the three 3 simulated scenarios showed significant rotational ambiguity, nor were there substantial random errors in the dataset after running the DISP and BS methods. Results for the DISP method showed no factor swaps for all dQmax values. In the BS analysis, outputs were considered stable, yet not all base factors were mapped to the boot factors. On average, the percentage of factors correctly mapped was 84%, which is in line with (Noris and Duvall, 2014), where a minimum of 80% mapped factors are suggested for interpretability robust results and to support the number of factors selected. The target BB-LRT, Dust-LRT, and Volcanic-LRT profiles for the models were mapped in 88%, 80%, and 98% of the runs, respectively. The 25^{th}, 50^{th} and 75^{th} percentile percentiles of the $PM_{2.5}$ contribution rates calculated for the target profiles in the BS runs are presented in Table 2. The contribution rate variability variability of the contribution factor (g) represented by the BS runs is particularly important for small database models, where contribution rate results might be unstable compared to the more stable profile the PMF model when using small dataset inputs (i.e., sample size < 100). Here, it will determine if the results from the model are stable or unstable (Feng et al., 2023).

Final base models were constrained to improve the correspondence between the chemical profiles found by the PMF and the profiles expected based on the identified emission sources. Specific constraints were defined in the different modeling scenarios to refine the factor profiles. The model's factors are presented in Figure Fig. S3. For the BB-LRT model, three soft constraints were applied to the "coal boiler" factor to pull up the EC1, Se, and As concentrations. Conversely, the Ag, Se, and EC1 concentrations were pulled down in Factors the "ceramic industry", "gasoline" and "Dieseldiesel" factors. For the Dust-LRT model, four soft constraints were established: for . For "vehicular emissions", the concentrations of EC and Ni were maximally adjusted downward and upward, respectively; for . For "resuspended materialre-suspended materials," Ni was

Table 2. Statistics performance of PMF for models during days affected by BB-LRT, Dust-LRT, and Volcanic-LRT

Properties	BB-LRT	Dust-LRT	Volcanic-LRT
		Model statistics	
Samples	31	16	32
N. factors	6	6	6
Non-weak species	32	27	27
Bad species	OC5,EC6	Be	—
$Q/Q_{expected}$	0.99	1.00	0.93
Add. uncertainty[%]	16	11	12
BS mapped factor [%]	82	82	88
DISP swaps	0	0	0
		Model statistics for $PM_{2.5}$	
$PM_{2.5}$ error [$\mu g/m^3$]	2.50	1.84	2.60
R^2	0.92	0.90	0.73
RM [%]	99.31	98.24	97.50
		LRT factor statistics for $PM_{2.5}$	
$PM_{2.5}$[$\mu g/m^3$]	11.14	6.77	6.46
$PM_{2.5}$[%]	37.61	33.93	30.85
$PM_{2.5}$ BS 25th [$\mu g/m^3$]	9.17	4.00	5.66
$PM_{2.5}$ BS 50th [$\mu g/m^3$]	10.53	5.63	6.53
$PM_{2.5}$ BS 75th [$\mu g/m^3$]	11.48	6.95	8.47

reduced to improve the fit; and for "biomass burning," OC was maximally increased. In the volcanic-LRT event scenario, two soft constraints were implemented. For volcanic-LRT, we pulled down the concentration of Cu and Mg, while Se was pulled up for "secondaries" and Cu was pulled down for "vehicular". For the models, the %dQ (i.e. the Q change because of the constraint) ~~is~~ was $< 1\%$.

The results from the PMF for BB-LRT ascribed concentration to six emissions sources (or factors): BB (37.6%), coal boiler (20.1%), ceramic industry (16.7%), gasoline (12.8%), ~~Diesel~~ diesel (11.4%) and ~~Incineration~~ incineration (1.4%). The factor of contribution for BB-LRT in the ~~sampling days varies~~ days sampled ranged from 0.0 to 4.5 (maximum ~~identified on~~ value identified on the 25 March 2020), which means that not all identified event days are backed up by the PMF (~~The~~ the factor of contribution ~~for the models ran~~ from the PMF model runs are in Supplementary ~~Figure~~ Fig. S4). Of the 31 samples identified, 27 days (equivalent to 87.0%) have positive contributions from ~~BB abscrited~~BB-LRT events. The PMF profile ~~of~~ for the BB factor, represented in Fig. 8a, is identified by the dominant contribution ($\geq 30\%$) of OC species, PyC and some anions. The identified contribution of this factor in OC, but especially in OC1 (50.9%) and OC2 (46.1%), agrees with observations made by Chow et al. (2004) for vegetation burning. Similarly, the high contribution of CyP (43.0%) ~~back~~ supports the profile since

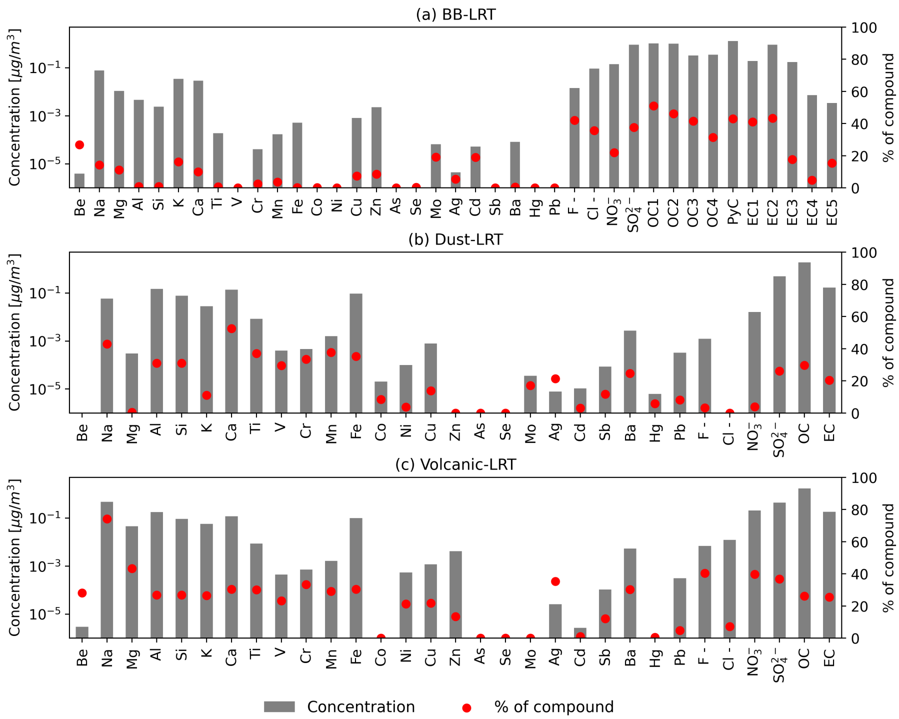

Figure 8. PMF output profiles for (a~~.~~) BB-LRT, (b~~.~~) Dust-LRT, and (c~~.~~) volcanic-LRT events. The bars represent the average concentration of the compounds, while the red points denote the compound's average percentage contribution to the whole element concentrations. Refer to Supplementary ~~Figure~~ Fig. S3 to see the contribution of all factors.

BB produces ∼50% of global CyP emissions (Santín et al., 2016). Regarding anions, the high contribution to F^- (42.1%) ~~head contribution and support the identification and hypothesis of LRT as a trace element with high~~ supports the identification of BB-LRT since this is a tracer of BB with a long lifetime (Jayarathne et al., 2014). The contribution to SO_4^{2-} (37.7%) and NO^{3-} (22.0%) ~~evidence secondary pollutants~~ suggests secondary pollutant formation from the BB emissions during the daytime and nighttime, respectively (Rastogi et al., 2014). Moreover, the influence ~~over~~ of Cl^- ~~concentration~~ concentrations (35.7%) may ~~evidence~~ be suggestive of semiarid vegetation burning (Andreae et al., 1998). Other species such as K (16.2%) ~~traces~~ are indicators of BB emissions (Yu et al., 2018; Rastogi et al., 2014).

For the Dust-LRT event days, the model identified ~~the dust contribution~~ $PM_{2.5}$ contributions from dust (33.9%) and ~~others from~~ BB (27.8%), vehicular (12.4%), resuspended material (6.5%), incineration (7.7%), and ceramic (11.7%). Due to only total OC and EC being considered, splitting the vehicular emissions between diesel and gasoline is difficult. The contribution factor for dust varies from 0.0 to 4.1 (maximum identified on 27 February 2020). For this event, 81.2% (13/16 samples) of the identified days exhibited a positive contribution. The dust profile ~~identification~~ classification is supported by a high contribution (>31.0%) of Al, Ca, Fe, and Si ($\geq 0.1\ \mu g/m^3$), as illustrated in Fig. 8b~~illustrates~~. Similarly, Malaguti et al. (2015) highlights Fe, Al, Ca, Ti, and Mg in the fine ~~Sahara's~~ Saharan dust aerosols. Ca and Ti are specially weighted for LRT from the Sahara desert since the other elements are more generic crustal aerosol tracers (Martinez-Verduzco et al., 2023; Nicolás et al., 2008). The dust contributes to 52.6% and 37.0% of Ca and Ti concentrationconcentrations, respectively. The profile also diverges from the general crustal material in the contribution of secondary inorganic aerosol as SO_4^{2-} probably from $(NH_4)_2SO_4$ (Varrica et al., 2019; Malaguti et al., 2015), with 33.9% of average concentration in the samples. Likewise, a particular mix between dust minerals and OC (29.7%) might also be a characteristic to identify the dust profile (Aymoz et al., 2004; Malaguti et al., 2015).

For the days of Volcanic-LRT events, the model identified 30.8% of volcanic aerosol contributionthe $PM_{2.5}$ profile from volcanic aerosol contributions. Other sources were BB (29.1%), vehicular emissions (15.7%), resuspended dust and ceramic industry (11.0%), secondary aerosol formation (9.1%), and incineration (4.3%). The volcanic factor presented had contributions varying from 0.0 to 3.9 (maximum identified on 6 February 2020). According to the model, the volcanic factor influences only 78.1% (25/32 samples) of identified days. As expectedSatsumabayashi et al. (2004), the volcanoes profile in Fig. 8c, shows a high contribution of SO_4^{2-} (36.9%). The contribution but not the concentration of SO_4^{2-} was surpassed by However, the days affected by this LRT event did not exhibit significantly higher concentrations than the surrounding days. The contribution of the LRT event was higher for F^- and NO^{3-} than for SO_4^{2-}. F^- from volcanic emissions frequently concentrated onto the ash surface concentrates onto the surface of ash as CaF2, AlF3 or Na_2SiF_6 (Bia et al., 2020; Delmelle et al., 2021); while NO^{3-} may relate with the oxidized trace to the oxidized tracer species HNO_3 (Martin et al., 2012). Minerals with relatively large concentrations in the identified profile include Na, Al, Ca, Fe, Si, K, and Mg in descendent descending order presented concentration >0.04 $\mu g/m^3$ (contribution of >26.5%). The finding of Trejos et al. (2021) and Vanegas et al. (2021) supported supports the volcanic profile, with Si, Al, Fe, Ca, K, Mg, and Na as the main minerals for the Nevado del Ruiz Volcano ashes. Although the composition might vary between volcanoes, the results for Sangay volcanic ashes in South America also back backs up the profile with similar main elements (Moran-Zuloaga et al., 2023). From these, Fe, Al, and Na are the most representative for the ashes ash aerosols with a diameter < 2.5 μm (Mason et al., 2021);, while Ca, Na, Si, and K might be particularly persistent after LRT (Ruggieri et al., 2012), due to the atmospheric lifetime. Cu and Zn are other tracers observed here and which have been identified before for the Colima Volcano in the southeast of the ring of fire in Mexico (Miranda et al., 2004).

The total daily composition of $PM_{2.5}$ is also evidence of the impact of the LRT event. Fig. Figure 9 shows the average daily concentration $PM_{2.5}$ concentrations of the campaign-measured data for each kind of type of LRT event and when no event is identified. This only considers days with a positive contribution factor ($g_{i,k}$>0) from PMF results. Although the non-event average concentration is presented, the statistical comparison is made with the days before and after events to maintain similar meteorological conditions (i.e. the combined time-series of before (DtEbetween the LRT event period (DtE$_{-3}$ to DtE$_{+3}$) and the combined before and after event time series (i.e. the DtE$_{-15}$ to DtE$_{-4}$) and after event ($_{-4}$ period is combined with the DtE$_{+4}$ to DtE$_{+15}$) is statistically compared to the event time-series (DtE$_{-3}$ to DtE$_{+3}$period) using the Mann Whitney U test. Unlike the PMF model, the comparison in Fig. 9 contains an analysis of the cations, the carbon matter species and the OC/EC and SOC/OC ratios for every type of event. Here, the major elements generally have a more significant increment in LRT events. Some elements supported the model's fingerprint (Fig. 8), e.g., OC, OC1, OC2, SO_2^4 for BB; Fe, Al, and Ti for dust; and Si, Al, Fe, Ca, Mg, and Na for volcanic aerosols (Fig. 9). The OC was significantly higher for BB, presenting a median OC/EC

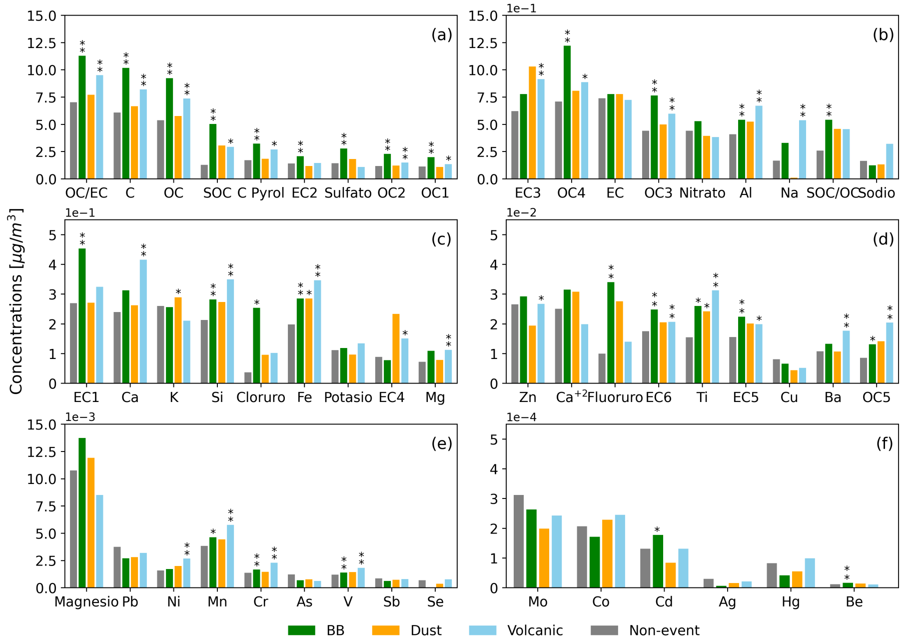

Figure 9. Campaign average concentration for non-event and positive-contribution days for BB-LRT, Dust-LRT, and Volcanic-LRT. The bars represent the concentration of the compounds along with the significance level compared with the days before and after the events, ≤ 0.1 (*) or ≤ 0.05 (**). The y-axes change for every figure to better identify concentrations. Supplementary Table S2 presents these values and the mean concentrations of the compounds for days around (i.e., before and after) each event.

ratio of 11.3 that surpasses common urban combustion ratios like from fossil fuel ($\sim$4), combustion, and ~~Diesel exhausted~~ diesel exhausts (<1) (Pani et al., 2019). Although OC/EC is more commonly used to identify sources of urban combustion and BB, some studies have shown its potential for determining the influence of volcanic activity (Pongpiachan et al., 2019). This is supported by evidence of carbon enrichment resulting from volcanic ~~influence (Martinsson et al., 2009), which~~ activity (Martinsson et al., 2009). This is also observed in this study, with consistently ~~high~~ higher concentrations in all OC species and almost all EC species (EC3, EC4, EC5, EC6). For volcanic aerosols, OC/EC had a median ratio of 9.5. Likewise, the formation of SOC significantly increased for both events, with ~~magnitudes~~ ratios of 5.0 and 2.9 for BB and volcanic aerosols, respectively. Heavy metals Cr (2.30 ngm^{-3}), Ni (2.67 ngm^{-3}), and Mn (5.76 ngm^{-3}) concentrations notably increased in event days for volcanic aerosols. Only the maximum daily concentration for Ni (25.05 ngm^{-3}) ~~exceeds the concentration limit suggested~~ exceeded the air quality standards suggested by the European Commission (2019) for the annual average ~~of the compound~~ concentration (20 ngm^{-3})~~by European Commission (2019)~~. Increments for Mn (4.64 ngm^{-3}) and Cr (1.67 ngm^{-3}), in addition to Cd (0.18 ngm^{-3}), were observed for BB. The ~~(European Commission, 2019) set an annual limit~~ European Commission (2019) set an air quality standard of 5 ngm^{-3} for the Cd ~~average~~annual average concentration.

The elevated concentrations of ions in the days of events (Fig. 9) also support the profiles (Fig. 8) and align with the literature. In addition to the ions observed in the PMF profile for the BB, the cations K^+ are representative of ions (Rastogi et al., 2014; Moreno et al., 2023) that present significant increments for this type of event. Regarding the volcanic aerosol compositions (Fig. 9), the observed increment in Na^+ and K^+ also ~~align~~ aligns with previous reports (Moreno et al., 2023; Mather et al., 2003; Roberts et al., 2018). On the other hand, although the PMF's fingerprint presented a high contribution of $SO2^{4-}$ and F^-, this was not enough for a significant rise in daily concentrations shown in Fig. 9.

4 Discussion

Our analysis focused on ~~events with high aerosol load and frequency, supported by a relatively long~~ the LRT of aerosols from a range of sources (BB, dust and volcanic) to the AV. Long-term (i.e. several years) field measurements of $PM_{2.5}$ ~~chemical composition record. This approach~~ and its chemical composition helped to identify the impact of these events on local air quality. This long-term approach was important as it avoided restricting the study to short periods (i.e., sporadic singular large-scale ~~events) and made it possible to quantify longer-term and more frequent events (i.e., the long-term LRT transport of pollutants to the AV and not just the infrequent large-impact LRT events)~~high impact events). We used model/satellite-derived reanalysis data to identify regional-scale transboundary events and ~~evaluate~~ evaluated the effects on $PM_{2.5}$ ~~concentration increments and composition~~using local aerosol field measurements.

Similar to other studies, thresholds were set for identifying ~~events (Ridley et al., 2012; Carn et al., 2008); however~~LRT events (Ridley et al., 2012; Carn et al., 2008). However, their selection in this study was representative of local events rather than high-polluted sporadic events. The OM-AOD threshold for BB-LRT events (0.2) agrees with previous results (0.2-0.3) (Kaiser et al., 2012; Misra et al., 2020). The Du-AOD (0.02) threshold was lower (i.e., 0.05) than suggested by Ridley et al. (2012) and that used by Achilleos et al. (2020) to track outstanding episodes but is of a comparable order of magnitude. In contrast, the $TCSO_2$ threshold in this study for volcanic degassing events differed from that discussed in the literature. The threshold used (0.87 mg/m^2) was ~~lower more than ten~~ more than 10 times than the used for another degassing volcano (17.15 mg/m^2) (Carn et al., 2008).

The lower $TCSO_2$ threshold derived in this study is likely linked to the CAMS product we used. While we use the CAMS reanalyses for OM-AOD, Dust-AOD, and $TCSO_2$ for consistency (i.e., tracers from the same model), the representation of the actual magnitude of $TCSO_2$ in the reanalysis product is lower than the operational product. The reanalysis uses an older climatological (2005-2010) emissions dataset (CAMS-GLOB-VOLC) for volcanic degassing (Granier et al., 2019) and only assimilates satellite AOD. While satellite AOD is a useful product to help represent volcanic properties (e.g., volcanic plumes, ~~SO2~~ SO_2 concentrations), the operational product also assimilates satellite $TCSO_2$ (CAMS, 2023), providing more constraint on the absolute magnitude of $TCSO_2$. However, regarding this study, the identification process is more important based on the data variability rather than the overall magnitude.

The identified BB-LRT reproduced the seasonality described in the literature (Mendez-Espinosa et al., 2019; Hernandez et al., 2019; Rodríguez-Gómez et al., 2022). Peak $PM_{2.5}$ concentrations occurred in February and March for BB-LRT events.

In ~~concordance~~ line with Mendez-Espinosa et al. (2019) and Henao et al. (2021), our findings show that the average conditions in this period are characterized by wind transport from the northeast of the ~~valley~~AV, covering part of the Orinoco and Caribbean regions. BB aerosols from fires in the Orinoco can reach the ~~valley~~ AV through two passes in the Andes mountain range. The limited transport throughout this complex topography ~~might promote the longer stance and~~ potentially promotes the accumulation of aerosols, increasing their concentration in the AV (Ballesteros González, 2021). Furthermore, the back-trajectories ~~warn about the~~ highlight the the Middle Magdalena Valley, which suggests high BB emissions ~~according to the average and the hotspots of~~ based on the reported OM-AOD hotspots. This area has been previously considered critical for open fires in Colombia (Bolaño-Díaz et al., 2022; Ballesteros González, 2021). According to Ballesteros González (2021), fires from this ~~zone~~ region are more likely to affect the Andean cities than fires from the Orinoco due to the mountain ~~barriers~~terrain acting as a natural barrier to LRT.

The meteorology characterization for BB-LRT showed ~~some conditions that might enhance the longer influence of the identified events. The assessment highlights the dry conditions in NSA as promoters of~~ conditions favourable in enhancing LRT of aerosols to the AV. Here, dry conditions (i.e. low precipitation and humidity) in NSA promotes both the occurrence of fires and the LRT of pollutants. In contrast, despite the high emissions of Amazon fire smoke in the southern hemisphere, these do not considerably increase aerosols in the city due to the scavenging of particles ~~in~~ during their transport (Hamburger et al., 2013). ~~The~~ However, the BB-LRT around August might be related to fires ~~in~~ from that region (SIATA, 2021; Hamburger et al., 2013).

The estimated contribution to $PM_{2.5}$ concentrations during the BB-LRT showed the highest magnitude for the studied events, with 11.14 $\mu g/m^3$ (37.6%), ~~backed up~~ supported by the comparison of $PM_{2.5}$ concentration. According to Ballesteros-González et al. (2020), the contribution from BB was calculated to be 4.7 $\mu g/m^3$ for ~~the monitored city~~ Medellín in February 2010 and 2018. This ~~finding~~ result is consistent with an average (maximum) estimation of 6.0 (12.5) $\mu g/m^3$ for samples taken in the same month in ~~2020. Furthermore,~~ 2020 from our campaign, which is also supported by the PMF results~~support the record of historical events, such as~~. In the case of March 2020, at the beginning of the COVID-19 pandemic~~. For this event~~, the PMF ~~estimates~~ estimated a maximum contribution of 50.0 $\mu g/m^3$ out of 60.9 $\mu g/m^3$. ~~The~~ This particular event was characterized by ~~local emissions suddenly decreasing and climbing~~ a sudden reduction in local emissions but a sharp rise in $PM_{2.5}$ ~~concentration~~ concentrations due to LRT from BB (Henao et al., 2021; Mendez-Espinosa et al., 2020).

Like BB-LRT, the monthly frequency of Dust-LRT events coincides with the annual cycle described by other studies. South America experiences a dust reception from the Sahara desert mainly in two ~~trimesters~~seasons, from March to May and June to August (Prospero et al., 2020). The seasonality of dust events partially coincides with the months of ~~occurrence of~~ BB-LRT events, causing ~~high~~ substantial overlapping between the identified days for these two types of events. Consequently, the identified weak impact on $PM_{2.5}$ by dust events controlled by Andes Mountain (Prospero et al., 2020) might be overshadowed by the most critical effect of BB-LRT. The only widely recognized event recorded for the study period occurred on June 24 and 25, 2020 (Mendez-Espinosa et al., 2020; SIATA, 2021). According to the PMF results, this exhibited ~~almost two~~ an almost 2 times higher contribution to $PM_{2.5}$ than the average event considered in this study. However, the PMF identified equivalent contributions in February 2020. The overlapping between these ~~events~~ LRT event types, and the small database size for the

Dust-LRT model~~might provoke impurities in~~ , might result in less robust information from the PMF factor related to ~~Sahara~~ Saharan dust. However, the dust profile aligns with the literature; the organic carbon contribution ~~evidenced a likely mix with a combustion source~~likely contained contributions from combustion sources.

In contrast to BB-LRT and Dust-LRT events, the impact of Volcanic-LRT is expected to be primarily determined by the direction of the wind rather than an actual seasonality in the emissions. Volcanic-LRT events occurred more frequently from July to September, when low to mid-level winds ~~blew~~ flow from the Nevado del Ruíz volcano region in the south-east, facilitating the transport of emitted aerosols to the AV. According to the ~~sulfur dioxide (~~SO_2 ~~)~~ V2 catalog, available on https://so2.gsfc.nasa.gov/measures.html (Fioletov et al., 2023), the AV ~~belongs to the influence area~~ is within the influence of the Nevado del Ruíz volcano. Indeed, this study identified a significant increment in $PM_{2.5}$ concentrations. According to the PMF results, on average, the contribution for $PM_{2.5}$ was 6.46 $\mu g m^{-3}$. Likewise, Casallas et al. (2024) linked the increase in $PM_{2.5}$ concentration during the JJA ~~trimester~~ season with volcano activity in Cali, a relatively equidistant city located south of the volcano.

Although the Volcanic-LRT profile is predominantly made up by SO_4^{2-}, the total concentration of the elements ~~does~~ do not show a significant increment compared with surrounding days (i.e. non-event days). This lack of significance in SO_4^{2-} has been observed in other cities where urban sources of SO_2 ~~dominated~~ dominate (Miyakawa et al., 2007). However, some alternative hypotheses ~~highlight regarding~~ relate to the magmatic gas state, ~~pointing out that temperature before emissions might provoke low S~~ suggesting that lower temperatures before emission might promote low sulfate oxidation and moderate SO_4^{2-} formation (Mather et al., 2003) since SO_4^{2-} ~~precursor formed at high temperature~~ precursors are formed at higher temperatures frequently close to the volcanic vent (Roberts et al., 2018). For instance, as ~~evidenced~~ observed in the Volcanic-LRT profile, the Cl-poor plumes might reduce ~~S~~ sulfate oxidation and forward formation of SO_4^{2-} (Mather et al., 2003).

The results have several implications for future research in the region. Modeling studies in the AV have only considered local emissions as inputs (e.g., Henao et al., 2020; Hernández et al., 2022)~~, and~~ . Therefore, it has been highlighted ~~the need to include~~ that external pollution sources ~~as input in~~ need to be included as additional inputs into chemical transport models to have a more complete representation of air quality in the ~~city~~region. Further, this study suggests conducting shorter chemical campaigns and human health impact studies for target elements and sources, especially in the valley and upwind. For instance, special attention to the carbonaceous matter, F^- and Cd emitted from BB in open fires is needed for human and ecosystem health studies in Colombia (Tuomisto et al., 2008; Jayarathne et al., 2014), since this source is particularly important for the region. The infrequent contribution ~~to Sahara~~ of Saharan dust is still ~~significant~~relevant, and there is particular interest in ~~the northern part of the country, where Sahara's~~ northern Colombia, where the most intense events significantly ~~increase~~ increases the mortality relative risk (Arregocés et al., 2023). On the other hand, Colombia has 12 active volcanoes, highlighting the importance of degassing activity in the country. Monitoring emissions and human health exposure is a primary need for municipalities, especially those close to active volcanoes such as Nevado del Ruíz. Both BB and ~~Volcanic event contribution to heavy metals raise alerts around possible body metal accumulation and~~ volcanic events contribute to an increase in heavy metal concentrations. This can adversely impact human health when metal accumulation occurs in the body, leading to diseases such as thyroid cancer (Malandrino et al., 2020; Vigneri et al., 2017). The ~~inhalation intake of these heavy metals sums up to other~~

~~exposures, such as~~ entry of heavy metals into the body via multiple routes (e.g. inhalation, ingestion and dermal ~~route, which might also be higher near~~ absorption) increases the risk of exposure, which is more pronounced nearer the source (e.g., ~~from water and crop pollution~~contamination of the air, water and crops). The increment in Mn and Cr might ~~provoke~~ enhance $PM_{2.5}$ toxicity in lung epithelial cells (Yuan et al., 2019). A deep assessment of the Cr(VI) and Cr(III) needs to be done to evaluate the correct toxicity status of Cr.

Finally, this study ~~advertises~~ highlights the need for cooperation among local, national, and international entities to manage complex aerosol sources/emissions and human exposure.

5 Conclusions

This study evaluates the impact of BB-LRT, Dust-LRT, and Volcanic-LRT aerosols ~~in~~ on the $PM_{2.5}$ ~~concentration and~~ concentrations and its chemical composition in the AV, a densely populated and mountainous region in the tropical Andes. Events ~~for each aerosol type were identified using data with different anomalies from CAMS reanalysis products~~of the LRT of aerosols and their precursors (e.g. volcanic SO_2) were categorised when substantial enhancements (i.e. non-local) in the relevant tracers above background levels from the CAM reanalyses were identified over the campaign site in the AV. Meteorological data from ERA5 and GPM, along with a back-trajectories analysis, were used to link the aerosol events in the AV with the sources and to characterize the meteorological patterns that determine the LRT of aerosols. The impact of these regional events on the local $PM_{2.5}$ was finally evaluated ~~with local concentration comparisons and characterized through~~ using field campaign measurements of local $PM_{2.5}$ and its chemical composition in conjunction with PMF modeling. This ~~study used a unique~~ field campaign represents a unique long-term $PM_{2.5}$ composition ~~campaign~~ dataset from April 2019 to October 2020.

The methodology allowed for identifying periods ~~with LRT of~~ influenced by the LRT of air pollutants, separating the days before, during, and after the LRT events. During LRT event days, the back-trajectories ~~of air masses arrive from zones~~ identified air masses arriving from regions previously reported as critical sources of aerosols in the region. This enables the connection between aerosol events in the AV and the sources. The sources were identified as regional fires for BB-LRT, Saharan ~~sands~~ dust for Dust-LRT, and emissions from the Nevado del Ruíz volcano for volcanic-LRT. The different types of events ~~showed a very marked annual variability~~ were associated with substantial seasonal variability (i.e. pronounced season cylce). BB-LRT events occurred mainly in February, March, and April, with a lower occurrence during August. Dust-LRT events showed a higher incidence from April to August. Volcanic-LRT showed a high occurrence ~~at the beginning of the second semester (~~from June to September ~~) and during January~~ and January to February. No events were identified during October, November ~~,~~ and December.

The weather patterns for BB-LRT events exhibited anomalous ~~low-level~~ lower-tropospheric northeasterly winds, favoring regional transport of aerosols. Additionally, drier conditions in northern Colombia and Venezuela promoted the occurrence and spread of fires in these regions. For Dust-LRT, more intense winds from the Caribbean ~~at medium levels favor~~ in the mid-troposphere favoured transport to the AV when the aerosol ~~load is~~ loading was high. Moreover, during Volcanic-LRT events, anomalous southeasterly winds ~~at low and medium levels~~ throughout the lower- and mid-troposphere were present, along with ~~low~~ lower precipitation in the southeast of the AV, where the Nevado del Ruíz volcano is located, enabling the ~~transport to the AV~~LRT.

When evaluating the impact of LRT events on ground-level $PM_{2.5}$, BB-LRT events resulted in more significant increases in concentrations. The relative increment on $PM_{2.5}$ from Dust-LRT and Volcanic-LRT events was smaller but still ~~significant~~prominent. Similarly, underpinned by literature-supported profiles, the PMF results ~~evidenced the event's influence on the~~ highlighted the importance of each LRT event type on influencing $PM_{2.5}$ chemical composition. OC species ~~such as~~ (e.g. OC1 and OC2~~, anions as~~), anions (e.g. F^- ~~and the traces of secondary pollutants~~) and secondary pollutants (e.g. SO_4^{2-} and NO_3^- ~~primarily determine the profile identification for~~) were the dominant components in the BB-LRT profile. On the other hand, general crustal minerals, together with Ca and Ti, ~~define~~ defined the Sahara-influenced Dust-LRT profile. Regarding Volcanic-LRT, SO_4^{2-} and characteristic minerals (e.g. Na, Al, Ca, Fe, Si, K, and Mg~~represented the~~) were substantially present in the corresponding profile. Further comparison ~~in positive contributed event days supports~~ of the observations, on days which had a positive contribution factor, supported the PMF results although identified no substantial SO_4^{2-} rise during volcanic degassing events. This aligned with the observed low Cl^- contribution, supporting the reduced S oxidation hypothesis and forward production of SO_4^{2-}. Carbon and ion identification increments were mainly observed for BB-LRT and Volcanic-LRT. Lastly, the ~~profile~~ profiles associated with significant increments in the heavy metals Cr, Mn, Cd, and Ni ~~raises concerns~~highlight the potential risk to local air quality from LRT and are a cause for concern for local and national public health authorities. Future studies using chemical models, larger ~~events~~ event datasets, and more detailed measurement and characterization techniques may offer further explanations for this increase in observed heavy metal tracers.

The findings have several implications for future research. For instance, modeling studies in the region have only considered local emissions as inputs, and external pollution sources, especially volcanic degassing, need to be included as inputs in chemical transport ~~models~~modeling work/studies.

Code availability. A Python code for calculating the back-trajectories is available at https://github.com/MariaPa96/Basic_Trajectories_python.

Data availability. The CAMS reanalysis (Inness et al., 2019) data were downloaded from the Atmosphere Data Store at https://ads.atmosphere.copernicus.eu/cdsapp#!/dataset/cams-global-reanalysis-eac4?tab=overview. Meteorological data of the ERA5 reanalysis were downloaded from the Climate Data Store at https://cds.climate.copernicus.eu/cdsapp#!/dataset/reanalysis-era5-pressure-levels and GPM precipitation data was retrieved from the NASA Earthdata at https://disc.gsfc.nasa.gov/datasets/GPM_3IMERGHH_07/summary. The hourly $PM_{2.5}$ concentration were download from the official network platform at https://siata.gov.co/siata_nuevo/. The datasets of wind for the trajectories calculation were downloaded from NCEP-NCAR Reanalysis 1 (https://psl.noaa.gov). The $PM_{2.5}$ chemical campaign datasets are available upon request to MGM (mgomez@elpoli.edu.co).

Author contributions. MPVG: Conceptualization, Methodology, Software, Formal analysis, Data Curation, Writing–Original Draft, Visualization. KSH: Conceptualization, Methodology, Software, Writing-Original Draft, Writing - Review & Editing, Visualization. JAV: Data curation, Formal analysis, Software. RJP: Writing-review & editing. MGM: Conceptualization, Data Curation, Formal analysis, Methodology, Resources. AMR: Conceptualization, Methodology, Writing - Review & Editing, Supervision, Project administration.

Competing interests. The authors declare that they have no conflict of interest

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