Source apportionment study on particulate air pollution in two high-altitude Bolivian cities: La Paz and El Alto

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Abstract. La Paz and El Alto are two fast-growing high-altitude Bolivian cities forming the second-largest metropolitan area in the country. Located between 3200 and 4050 m a.s.l., these cities are home to a burgeoning population of approximately 1.8 million residents. The air quality in this conurbation is heavily influenced by urbanization; however, there are no comprehensive studies evaluating the sources of air pollution and their health impacts. Despite their proximity, the substantial variation in altitude, topography, and socio-economic activities between La Paz and El Alto result in distinct sources, dynamics, and transport of particulate matter (PM). In this investigation, PM_{10} samples were collected at two urban background stations located in La Paz and El Alto between April 2016 and June 2017. The samples were later analyzed for a wide range of chemical species including numerous source tracers (OC, EC, water-soluble ions, sugar anhydrides, sugar alcohols, trace metals, and molecular organic species). The US-EPA Positive Matrix Factorization (PMF v.5.0) receptor model was employed for the source apportionment of PM_{10}. This is one of the first source apportionment studies in South America incorporating an extensive suite of organic markers, including levoglucosan, polycyclic aromatic hydrocarbons (PAHs), hopanes, and alkanes, alongside inorganic species. The multisite PMF resolved 11 main sources of PM. The largest annual contribution to PM_{10} came from two major sources: the ensemble of the four vehicular emissions sources (exhaust and non-exhaust), accountable for 35% and 25% of the measured PM in La Paz and El Alto, respectively, and dust, which contributed 20% and 32% to the total PM

mass. Secondary aerosols accounted for 22% (24%) in La Paz (El Alto). Agricultural smoke resulting from biomass burning in the Bolivian lowlands and neighboring countries contributed to 9% (8%) of the total PM_{10} mass annually, increasing to 17% (13%) between August-October. Primary biogenic emissions were responsible for 13% (7%) of the measured PM_{10} mass. Additionally, a profile associated with open waste burning occurring from May to August was identified. Although this source contributed only to 2% (5%) of the total PM_{10} mass, it constitutes the second largest source of PAHs, compounds potentially hazardous to health. Our analysis additionally resolved two different traffic-related factors, as well as a lubricant source (not frequently identified) and a non-exhaust emissions source. Overall, this study demonstrates that PM_{10} concentrations in La Paz and El Alto region are predominantly influenced by a limited number of local sources. In conclusion, to improve air quality in both cities, efforts should primarily focus on addressing dust, traffic emissions, open waste burning, and biomass burning.

1 Introduction

Outdoor air pollution has undeniably proven to be an important threat for public health, being responsible for about 4.2 million yearly premature deaths around the world every year (WHO, 2021a). The exposure to air pollution becomes more complex at higher altitudes due to the decrease in oxygen per volume of air, as people have developed a higher lung capacity in order to fulfill the body oxygen demand (Frisancho, 1977; Frisancho, 2013; Frisancho et al., 1999; Madueño et al., 2020; U.S. EPA, 2011).

Many of the high-altitude large cities in the world (> 2000 m a.s.l., ≳2 million inhabitants) are located in Latin American low and middle-income countries, among which are Mexico City, Bogotá and Quito. These cities are also subject of a developing industry and a growing vehicular fleet that results in a constantly increasing energy consumption, heavily dependent on non-renewable energy sources (Pardo-Martínez, 2015; Molina et al., 2019; Castro-Verdezoto et al., 2019). Most of the cities in this region, for which data is available, face a deteriorated air quality, with particulate matter (PM) concentrations that exceed the World Health Organization (WHO) guidelines (Gutiérrez-Castillo et al., 2005; Mugica et al., 2009; Ramírez, et al., 2018; Zalakeviciute, et al., 2020, WHO, 2021b).

High-altitude cities exhibit distinct characteristics due to complex topography and associated meteorology, influencing the transport, accumulation and dispersion of air pollution. Moreover, high altitude is linked to strong solar radiation that favors photochemical activity and high daily temperature variations. Compared to other regions at similar latitudes, high altitude cities in South America experience lower temperature, lower atmospheric pressure and saturation vapor pressures, as well as complex wind patterns and reduced precipitation (Zalakeviciute et al., 2018; Vega et al., 2010). Previous studies have shown that these specific high-altitude atmospheric and thermodynamic conditions can strongly favor new aerosol particle formation (NPF) (Boulon et al., 2010; Brines et al., 2015; Hallar et al., 2011; Sellegri et al., 2019; Singla et al., 2018; Sorribas et al., 2015). Additionally, it has been observed that low oxygen environments alter the performance and reduce the efficiency of

combustion engines (Martínez et al., 2022; Wang, et al., 2013a), thus, changing the vehicular emissions of gaseous and particulate pollutants (Bishop et al., 2001; Giraldo & Huertas, 2019; He et al., 2011; Nagpure et al., 2011; Wang et al., 2013b). Listed amongst the highest metropolitan areas in the world, La Paz (between 3200-3600 m a.s.l.) and El Alto (4050 m a.s.l.) are two Bolivian cities constituting a conurbation with a population of approximately 1.8 million people. Despite their close proximity, significant topographical, meteorological and socio-economic differences exist between them. While Bolivian legislation regulates concentrations of certain pollutants (CO, SO_2, NO_2, O_3, TSP, PM_{10}, Pb; Table S1), very few air quality studies which include long term measurements at moderate time resolution have been performed in the country or in the region. The few previous existing studies have reported PM_{10} mass concentrations ranging from 10 and 100 µg m^{-3} measured at urban and urban-background stations in La Paz and El Alto (Red MoniCA, 2016 a,b , 2017, 2018; Wiedensohler et al., 2018). However, no particle chemical speciation has been conducted to identify the major sources contributing to the high PM concentrations. Furthermore, measurements taken at the nearby Chacaltaya GAW station (CHC-GAW: 16.350500°S, 68.131389°W, 5240 m a.s.l.) show that the emissions of the city not only impact the local environment but also act as a point source influencing regional atmospheric composition (Aliaga et al., 2021).

While little is known about the sources of PM in the country, since industry is not largely developed, vehicular emissions potentially represent an important contributor to air pollution, particularly considering the absence of restrictions on the age of the vehicle fleet. Statistics indicate that 43% of the circulating vehicles are less than 10 years old, 15% are 10-20 years old and another 24% are 20-30 years old (INE, 2020a, b). At a regional scale, agricultural biomass burning in the Bolivian and Brazilian valleys and rain-forests constitutes an important seasonal source of particulate pollutants (Mataveli et al., 2021). The latter has a significant impact on the air quality of the cities close to where the fires take place (Nawaz and Henze, 2020) and can be transported over large distances. Studies have shown that air masses coming from the Amazon can traverse the Andes, carrying pollutants and ultimately reaching the Bolivian Altiplano (Chauvigne et al., 2019; Magalhães et al., 2019; Segura et al., 2020; Bourgeois et al., 2015). Additionally, previous studies based on emission inventories adapted to the data availability in LP-EA pointed out road dust, food industry, cooking and vehicle emissions as the major sources of PM_{10}, whereas for Cochabamba (the third largest urban area in Bolivia) estimations showed mobile sources to be responsible for almost 90% of PM_{10} emissions (Herbst, 2007; Pareja et al., 2011). Although there are some indications of the most outstanding sources of particulate matter in La Paz and El Alto, currently there is not comprehensive study on the composition and sources of particulate matter air pollution. Therefore, the aim of this study is to apportion and characterize the sources of PM that affect air quality in the metropolis of La Paz-El Alto, which can be used as a baseline for future policy making.

To achieve this goal, the EPA-Positive Matrix Factorization (PMF v.5.0) receptor model was applied on the PM_{10} chemical speciation obtained from 24-h filter samples collected simultaneously in La Paz and El Alto over a 15-months campaign. This study represents one of the few conducting PM characterization in Bolivia over an extended period. Given the limited number of studies in this region, identifying the sources and chemical profiles of PM in the study sites proved to be more challenging. The analysis included a comprehensive chemical speciation, encompassing ionic species, monosaccharide anhydrides, polyols,

metals, PAHs, alkanes, and hopanes. To the best of our knowledge, this is the first study on source apportionment at high-altitude cities that incorporates such a large set of organic and inorganic species.

2 Method

2.1 Sampling sites

Significant topographical differences exist between the two study sites, La Paz (LP) and El Alto (EA), in addition to the notable disparity in altitude and pressure. While the city of El Alto lies on the open and flat Altiplano plateau, the city of La Paz sprawls along the mountain valleys formed below the Altiplano in a closed area with steep and complex topography. The meteorological conditions throughout the year are governed by the seasonal transition between a dry and a wet season, typical of tropical regions. Temperature and wind patterns vary substantially between the two cities due to the differences in altitude and local topography.

Moreover, the city of El Alto originally developed as a peri-urban zone of the city of La Paz, welcoming migrants from nearby towns and communities who settled on the outskirts of the city of La Paz (Fernández, 2021). This gave rise to significant economic and social disparities between the cities that, to some extent, persist and are evident among the general population (Foster and Irusta, 2003). Such differences could have an impact on air pollutant emissions, due to the different practices in each of the cities, in addition to the daily commute of a significant part of the population of El Alto towards the city of La Paz. The few existing industries are mostly located within or in the surroundings of El Alto, and the vehicular fleet observed in both cities is not homogeneous. The density of heavy vehicle traffic – trucks and buses, is more prevalent in El Alto, since it is the main regional and international connection from and to the metropolis. These factors uphold the need for having independent representative sampling sites for each city rather than relying on a single site, despite both being part of the same conurbation.

The sampling campaign was carried between April 2016 and June 2017. Several ambient and meteorological parameters were measured simultaneously at two urban background sites, one in each city. The sampling sites were located 7 km apart, with an altitude difference of over 400 m, and located at approximately 20 km from the Chacaltaya Global Atmosphere Watch (CHC-GAW) monitoring station (Fig. 1).

The El Alto measurement site was installed within the El Alto International Airport, in the facilities of the meteorological observatory (16.5100° S, 68.1987° W, 4025 m a.s.l.). The observatory is situated at a distance of approximately 250 m from the airport runway and 500 m from the nearest major road and has been described elsewhere (Wiedensohler et al., 2018). Pre-campaign measurements were conducted to assess whether the takeoff and landing of airplanes had any significant influence on the measurements, revealing no substantial impact on CO_2, PM_1 and $PM_{2.5}$ during each airplane arrival and departure. Road traffic within the airport was minimal. The area around the sampling site is unpaved, hence dusty, and there are no other buildings in the proximity of the observatory. In March 2016, just prior the beginning of the sampling, the airport

administration cleared the ground within the perimeter fence of the meteorological observatory, leaving the site dustier than the rest of the airport.

La Paz measurement site (LP) was placed on the rooftop of the city's Museum Pipiripi (Espacio Interactivo Memoria y Futuro Pipiripi: 16.5013°S, 68.1259°W, 3600 m a.s.l.). This municipal building is located atop a small hill in downtown La Paz. Unlike the EA site, within a 1 km radius, the LP site is surrounded by many busy roads and dense residential areas, with a horizontal and vertical minimum distance to the nearest road of approximately 70 and 45 m respectively. Otherwise, the site's immediate surroundings (~100 m radius) are covered by green areas and a municipality buses parking lot at the base of the hill.

2.2 Sampling methods

High-volume samplers (MCV CAV-A/mb with an MCV PM1025UNE (PM_{10}) head) were employed to collect 24-h filter samples of PM every third day at both sites. Sampling started at 9:00 a.m. and the flow was automatically maintained at 30 m^3 h^{-1}. To avoid interference of near-ground particle resuspension, the samplers were placed on the rooftop of the buildings. Throughout the analyzed period of the present study, an impactor with a 50% collection efficiency for aerosol particles with an aerodynamic equivalent diameter of 10 µm was installed at the inlet of the samplers at both sites to establish an upper size-cut.

The mass concentrations measured at both sampling sites are hereafter reported in ambient conditions (EA: $\overline{T} = 280.8\ K, \overline{P} = 628.2\ hPa$, LP: $\overline{T} = 286.0\ K, \overline{P} = 664.7\ hPa$), unless stated otherwise (e.g. when compared to literature reported concentrations). To convert to standard conditions of temperature and pressure ($\overline{T} = 273\ K, \overline{P} = 1013.5\ hPa$) the concentrations must be multiplied by a factor of 1.66 and 1.60 in El Alto and La Paz, respectively. Since the difference in ambient concentrations between the sites resulting from a difference in mean temperature and pressure is of approximately 4%, ambient concentrations are directly compared between the sites in the subsequent sections.

The aerosol particles were collected onto pre-heated (8 hours at 500°C) and pre-weighted 150 mm-diameter quartz fiber filters (Pallflex 2500QAT-UP). After sampling, the filters were folded and wrapped in aluminum foil, sealed in impermeable plastic bags, and stored in a cool environment prior to transportation for analysis. Mass concentrations were initially determined gravimetrically, and then the samples were divided for chemical analysis among three European laboratories. The resulting chemical speciation comprised elemental carbon (EC), organic carbon (OC), sugar anhydrides (Levoglucosan, mannosan), sugar alcohols (arabitol, mannitol), water soluble ions (SO_4^{2-}, NO_3^-, Cl^-, MSA^-, NH_4^+, Na^+, K^+, Mg^{2+}, Ca^{2+}) measured at IGE, Grenoble, France; metals (Al, Ca, K, Na , Mg, Fe, Ti, V, Mn, Cu, Zn, Rb, Sn, Sb, Pb) measured at IDAEA, CSIC, Barcelona, Spain; Polycyclic aromatic hydrocarbons (PAHs: Fla, Pyr, Tri, BaA, Chr, BaP, BghiP, IP, BbF, Cor), alkanes (C21-C26), methyl PAHs, thiophens, hopanes (HP3-HP4) alkane methoxyphenols, and methylnitricatechols measured at EDYTEM, Chambéry, France (Table S2). A total of 92 and 103 filter-samples were collected in the cities of El Alto and La Paz, respectively, excluding samples having sampling flow issues or influenced by specific events (c.a. San Juan local festivity,

Christmas, New Year's Eve). In addition, laboratory blank filters were used to calculate the limits of quantification (QL). The average concentrations measured from the laboratory-blanks were then subtracted from the atmospheric concentrations measured from the filter samples.

2.3 Source apportionment (PMF)

The Positive Matrix Factor PMF 5.0 tool (Norris et al., 2014; Paatero and Tapper, 1994), developed by the U.S. Environmental Protection Agency (EPA), was used to apportion the sources that contribute to the observed particulate material in the collected samples at both sites. This non-negative multivariate factor analysis seeks to solve the chemical mass balance equation [1], applying a weighted least-squares fit algorithm, x_{ij} representing each of the elements of the concentration matrix (having n number of samples and m number of chemical species measured), g_{ik} are the contributions of each k factor to the ith sample, f_{kj} are the chemical profile of the factors, and e_{ij} are the residuals (i.e. the difference between the calculated and the measured concentration).

$$x_{ij} = \sum_{k=1}^{p} g_{ik} f_{kj} + e_{ij} \quad (1)$$

The optimal solution is then achieved by minimizing the function Q defined as:

$$Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \left[\frac{x_{ij} - \sum_{k=1}^{p} g_{ik} f_{kj}}{u_{ij}} \right]^2 \quad (2)$$

where u_{ij} are the uncertainties associated to each measurement.

2.3.1 Sample and chemical species selection

Out of the 197 PM_{10} samples initially included, 12 of them were later excluded from the analysis for having over 6 species with missing values (EA: 19 Sep 2016, 11 Jan 2017; LP: 14 May 2016, 07 Jun 2016, 12 Dec 2016, 02 May 2017) or because they presented unusual concentrations of PM or multiple species (LP: 04 Apr 2016, 22 May 2017, 30 May 2017, 11 Jun 2017, 15 May 2017, 19 Jun 2017). A total of 178 chemical species were measured for each filter. Species displaying irregularities in their time series were excluded from the analysis, as were those with over 25% of the data falling below the quantification limit (<QL, defined as the mean field-blank concentrations measured per specie, plus two times the standard deviation). From the remaining 86 species, the ones that were measured through both Ion Chromatography (IC) and Inductive Coupled Plasma-Mass Spectrometry (ICP-MS), only the ICP-MS metals were included in order to avoid duplicative counting, except for K^+, for which the IC measurements were used since water soluble K^+ is a known tracer for biomass burning (BB), soil resuspension, and fertilizers (Li et al., 2021; Urban et al., 2012). Galactosan and sorbitol were deemed unnecessary tracers for biomass burning and primary biogenic aerosols, respectively, as other specific tracers such as levoglucosan, mannosan, mannitol and arabitol were present. Consequently, they were excluded from the analysis. Additionally, other non-specific-tracer metal species were excluded after several attempts to including them in the PMF input data, as they introduced instability to the solution. Following the findings of Samaké et al., (2019a), arabitol and mannitol were added as one representative polyol

specie, given that they are emitted by the same source and have a Pearson correlation of r>0.7, at both sites. The same was done for PAHs that presented a r>0.9 (PAH_1: [BghiP]+[IP]+[BbF]; PAH_2: [Fla]+[Pyr]). Finally, in the PMF analysis, OC was substituted with OC*, which represents the difference between the measured OC concentrations and the carbon mass concentration of all the included organic compounds, to avoid double counting (e.g. Weber et al., 2019):

$$OC^{*} = OC - \left(\begin{array}{c} 0.12 \cdot [MSA] + 0.40 \cdot [Polyols] + 0.44 \cdot ([Levoglucosan] + [Mannosan]) + \\ 0.95 \cdot ([BghiP] + [IP] + [BbF] + [Fla] + [Pyr] + [BaA] + [Chr] + [Tri] + [BaP] + [Cor]) + \\ 0.85 \cdot ([C21] + [C22] + [C23] + [C24] + [C25] + [C26]) + \\ 0.87 \cdot ([HP3] + [HP4]) \end{array} \right) \quad (3)$$

2.3.2 Uncertainty calculation and species weight-assignment

In the uncertainty matrix, a 10% uncertainty was assigned to PM mass concentrations. The uncertainty calculation for polyols, monosaccharide anhydrides, and ions was followed the formula proposed by Gianini et al. (2012), employing the variation coefficients (CV) and the additional coefficients of variation (a) proposed and used by Weber et al. (2019), with the average QL associated to each species instead of DL. The uncertainties associated to EC, OC, and metals, were calculated following the method proposed by Amato et al. (2009) and Escrig et al. (2009). Furthermore, the uncertainties assigned to the molecular organic species were calculated according to the formulas proposed by Polissar et al. (1998) and Reff et al. (2007), with the substitution of DL values by QL.

Values below the QL in the concentration matrix were replaced by the average of QL divided by 2 for each specie. The corresponding uncertainties were then set to $\frac{5}{6}$ QL (Norris et al., 2014). The outliers encountered in the time series of some species (a total of 4 values) were replaced by NA. Subsequently, the missing values in the input file were set to be replaced in the software by the median value of the corresponding species and their associated uncertainty was automatically set to four times the species-specific median.

The weight of the species in the factor analysis was determined based on their signal to noise ratio (S/N). Species with a S/N>2 were set as strong. Species with a signal to noise ratio: $0.2 \leq S/N \leq 2$ were defined as weak, resulting in a down-weighing of their influence in the analysis by triplicating their uncertainties. Species with a S/N<0.2 were not included in the analysis. Finally, PM was set as total variable, thus setting it as a weak variable. After conducting several tests, certain variables were also set as weak (K^{+}, V), for setting them as strong variables resulted in the creation of artificial factors, without any geochemical meaning. The PAHs, alkanes and hopanes were set as weak species to prevent them from driving the solution.

2.3.3 Solution evaluation criteria

A range of solutions, spanning from 8 to 13 factors, was examined to determine the suitable number of factors contributing to each site. Subsequently, a final solution was chosen based on the evaluation of various statistical and geochemical control parameters, as described by Belis et al. (2019):

- $Q_{true}/Q_{robust} < 1.5$.
- Residuals per species were centered and exhibited a symmetrical distribution around 0, falling within the range of -3 and 3, with a few exceptions for outliers.
- Bootstrap (BS) evaluation of the statistical robustness of the selected base run having a correlation coefficient for every factor > 0.8 after 100 iterations, before and after constraints.
- Displacements (DISP) analysis was performed to evaluate the rotational ambiguity and the solution's tolerance to minor perturbations (No rotations were observed for dQmax = 4, 8).
- Geochemical consistency of the obtained factor chemical profiles based on literature and knowledge of the study site.

2.3.4 Multisite PMF

Initial parallel runs of single-site PMF analysis revealed similar main sources contributing to particulate matter. Increasing the number of factors showed potential for separating the traffic profile, albeit with a compromise on the statistical stability of the solution. Motivated by these findings, a multisite PMF analysis was conducted. Such approach has proven its ability to reduce the rotational ambiguity in factor analyses (Dai et al., 2020; Hopke, 2021; Pandolfi et al., 2020; Hernández-Pellón and Fernández-Olmo, 2019), increasing the statistical robustness through an increased number of samples.

To combine both datasets into a single dataset (EA-LP) the dates in the La Paz dataset were shifted in time by two years and then appended to El Alto's dataset. Thus, duplicated dates were avoided and while composing a single input matrix for PMF that respected the natural seasonal variability of the original datasets. The dimensions of the resulting matrix were 185 rows (samples) x 40 columns (species). The multisite approach stands on the hypothesis that the major sources contributing to PM_{10} in both sites are similar and exhibit similar chemical profiles, which has been verified within the single site solutions.

2.3.5 Set of constraints

Once the optimum number of factors was selected in the multisite base solution, a set of "soft" constraints (Table 1) was applied to the selected solution based on previous studies (Borlaza et al., 2021; Samaké, et al., 2019b; Weber et al., 2019)

2.3.6 Additional analysis of one local specific source: fuel chemical fingerprint

To further investigate the differences between the two main types of fuel used in LP-EA, 3 samples of both gasoline and diesel were taken at 3 randomly chosen gas-stations located in different areas of the city. The main metal composition of these samples was subsequently analyzed using the following procedure: 1 ml of sample (gasoline, diesel) was transferred into a Teflon microwave vessel (Anton Paar microwave laboratory unit). Then, 10 ml of HNO_3 (double distilled, suprapure level) were added and the solution was decomposed by increasing temperature and pressure (175°C and 10 bar). In the microwave, the EPA 3051A method was run twice to assure that the solutions were indeed decomposed (US EPA, 2007). After cooling

down the vessels, the solutions were diluted by a factor of 10 and directly measured using inductively coupled plasma mass spectrometer (ICP-MS). A complete descriptive table of the analyzed species can be found in the SI (Table S3).

3 Results

3.1 Seasonal variations of chemical components of PM_{10}

A yearly alternation between the dry and the wet season as presented in Fig. 2, shows an annual maximum of PM_{10} concentrations coinciding with the middle of the dry season (Southern hemisphere winter). During this season, negligible wet deposition occurs and favorable conditions for particle resuspension are prevalent. Maximum daily ambient PM_{10} concentrations of 37.2 ± 10.5 µg m^{-3} and 33.2 ± 7.5 µg m^{-3} were measured during this period (May-August) in El Alto and La Paz, respectively. Conversely, the wet season (Southern hemisphere summer, December-March) exhibits frequent precipitation events and the highest daily minimum temperatures.

Similar PM_{10} variability and concentrations were observed at the International Airport of El Alto, using the C^{14} beta-attenuation technique, between 2011 and 2015 (ranging between ca. 10-50 µg m^{-3} throughout the year, Red MoniCA, 2016b). In the case of La Paz, the variability observed while also using the C^{14} beta-attenuation technique was similar to the one observed in the present study. However, the reported PM_{10} concentrations were higher (Red MoniCA, 2016a, 2017, 2018). The discrepancy in the measured concentrations in the case of La Paz can likely be attributed the different measurement site locations, as the sampling site in La Paz described in the Red MoniCA (2016a, 2017, 2018) reports was located in the downtown area, next to a busy avenue.

Among all the samples collected during the measurements campaign, 5 and 12% of the daily samples collected in La Paz and El Alto exceeded, respectively, the 24-hour PM_{10} concentration of 45 µg m^{-3} not to be exceeded more than 3-4 days per year, according to the short-term PM_{10} Air Quality Guideline (AQG) level recommended by the World Health Organization (WHO, 2021). Moreover, the annual PM_{10} concentrations in both cities are at least 1.2 times higher than the PM_{10} levels of 15 µg m^{-3} recommended as annual AQG by the same organization (WHO, 2021b). Average measured PM_{10} concentrations were found to be 29.9 ± 12.0 µg m^{-3} (STP: 49.6 ± 19.9 µg m^{-3}) in El Alto and 27.2 ± 8.9 µg m^{-3} in La Paz (STP: 43.5 ± 14.2 µg m^{-3}). However, the annual average values can be relatively lower due to the under sampling during the wet season.

The observed concentrations are lower compared to those reported for Mexico City, a high-altitude (2850 m a.s.l) Latin-American megacity (Table 2), but higher than those observed in the cities of Bogotá and Quito. The average concentrations found in La Paz-El Alto are nearly double the reported average concentrations for most suburban and urban background sites in Europe, and similar to those measured in Turkey, certain regions in Poland (Rybnik: 44.1 µg m^{-3}), Bulgaria (Vidin: 41.3 µg m^{-3}), North Macedonia (Skopje: 48.7 µg m^{-3}) and Italy (Napoli: 46.9 µg m^{-3}) in 2019 (EEA, 2020, 2022).

The reconstruction of the measured PM_{10} mass resulted from the mass closure of the major components of PM, as described in Favez et al. (2010), Putaud et al. (2004), Seinfeld & Pandis (1998), Chan et al., (1997), Pérez et al. (2008), and Cesari et al., (2016). Thus:

$$\mathrm{PM(recons)} = \{(1.8[\mathrm{OC}])\} + \{[\mathrm{EC}]\} + \{([\mathrm{SO_4^{2-}}] - 0.252[\mathrm{Na^+}]) + [\mathrm{NO_3^-}] + [NH_4^-]]\} + \{2.54[Na^+]\} + \left\{1.15 \cdot \left((1.89[\mathrm{Al}]) + \left(2.14 \cdot (2.65[\mathrm{Al}])\right) + 1.67[\mathrm{Ti}] + \left(1.4 \cdot ([\mathrm{Ca}] - [\mathrm{Ca^{2+}}])\right) + \left(1.2 \cdot ([\mathrm{K}] - [\mathrm{K^+}])\right) + 1.36[\mathrm{Fe}]\right) + (1.5[\mathrm{Ca^{2+}}] + 2.5[\mathrm{Mg^{2+}}])\right\} \quad (4)$$

where the first curly bracket accounts for the organic matter, the third one accounts for the sum of the mass of secondary inorganic aerosol particles (non-sea-salt sulfate, nitrate, and ammonium), the fourth accounts for sea salt, and the fifth curly bracket accounts for the mass of the main components of crustal material: Al_2O_3, SiO_2, TiO_2, CaO, K_2O, FeO and Fe_2O_3 (multiplied by 1.15 to take into account sodium and magnesium oxides), and the mass of unmeasured carbonates.

Average PM_{10} (recons.) / PM_{10} (meas.) ratios of 0.91 in El Alto and 0.82 in La Paz were found. The remaining unidentified mass fraction may be attributed to the loss of volatile organic matter and secondary aerosols post-weighing, during the transport of the filter fractions to be analyzed. The difference can also be associated to the presence of non-measured species (i.e. carbonates) or to the adsorption of water in the aerosol particles or the filter (Pio et al., 2013). Moreover A 10% uncertainty associated with the gravimetry measurements could also have a role in the observed difference.

The average percentage contribution of the chemical species that significantly contribute to the measured PM_{10} concentrations in El Alto was: 22±5% OM (i.e. 1.8·OC), 5±2% EC, 9±5% secondary inorganic aerosols (NH_4^+, NO_3^-, and SO_4^{2-}), and 12±3% of crustal material (Al, Fe, Ti, Ca, K, Mg, Mn, P). In La Paz, 25±5% OM, 6±2% EC, 8±5% secondary inorganic aerosols, and 10±2% of crustal material. Moreover, Fig. S3 in the SI shows the monthly behavior of the principal species contributing to PM, along with certain specific source tracers.

Mean OC/EC mass ratios of 2.6±1.1 and 2.8±1.6 were found for El Alto and La Paz, respectively, during the measurements period. This average OC/EC ratio results from the combination of various sources including vehicle emissions and other primary and secondary local and regional sources of carbonaceous particles (such as biomass burning, primary biogenic emissions and secondary organic aerosols). The highest OC/EC ratios, with the largest standard deviation, were observed between August and October, peaking in September. The mean OC/EC ratios during this period is of 3.5±1.3 for El Alto and 3.8±1.6, indicating to the long-range influence of biomass burning emissions at the end of the agricultural year, as well as the the influence of primary organic emissions (Brines et al. 2019; Hays et al. 2002; Robert et al. 2007a, b; Samaké, et al. 2019a, b; Waked et al. 2014). It was observed that biomass burning tracers peak in August, while polyols display an increase in concentrations peaking in September. In contrast, minimum OC/EC ratios that display a smaller dispersion around the mean were observed between March and April: 1.9±0.6 and 2.0±0.6 in El Alto and in La Paz, respectively.

3.2 Source apportionment

After approaching the analysis individually for each site and observing that both sites shared similar sources, as well as considering the proximity of both cities, the multisite approach allowed to overcome the challenge posed by the relatively low

number of samples compared to the number of species included in the single site analysis. This approach immediately provided a solution with greater stability, maintaining the previously observed profiles and making it possible to achieve a stable 11-factor solution. Figure 3 displays the percentage contribution attributed by the PMF analysis to each of the resolved sources after applying the constraints described in the previous section.

The measured PM_{10} concentrations versus the modeled PM_{10} concentrations through the multisite approach exhibited a linear relationship with a slope of 1.01 and an R^2=0.95, indicating that the factor analysis adequately reproduced the measured concentrations. The 11 resolved sources include dust, secondary sulfate, secondary nitrate, primary biogenic aerosols (PBA), MSA-rich, biomass burning (BB), traffic 1 (TR2), traffic 2 (TR2), lubricant, non-exhaust emissions, and waste burning (Fig. 3). Most of the resolved sources are consistent with the emission sources observed in previous studies performed in other sites (Weber et al., 2019; Waked et al., 2014; Yang et al., 2016; Chevrier, 2016). A comparison of the chemical profile of the sources resolved in the present study, and the chemical profile of the sources resolved by Borlaza et al. (2021) and Weber et al. (2019) using the PD-SID method described in Belis et al. (2015) and, Pernigotti and Belis (2018) can be found in the SI. Furthermore, a separation of the traffic exhaust emissions (TR1, TR2) linked to the type of fuel used will also be presented in the following sections.

Dust and the ensemble of vehicular contributions (i.e. Traffic 1, Traffic 2, Lubricant, Non-exhaust emissions) together account for 55% and 57% of the measured PM_{10} mass concentrations in La Paz and El Alto, respectively. The dust factor exhibits outstanding contributions of 32% in the city of El Alto, making it the dominant source in this city. Although the volume sampler was placed on the roof of the observatory building, it cannot be excluded that the samples were influenced by the local dust**.** In La Paz, the vehicular emissions take the lead in terms of percentage contributions (35%). The factors associated with secondary aerosol particles (secondary sulfate, secondary nitrate, MSA-rich) were responsible for nearly 22% and 24% of total PM (La Paz and El Alto respectively), only a slight difference can be observed between the cities except for the nitrate rich profile. Finally, the biomass burning factor was responsible for an average of 9 and 8% of the total measured PM_{10} (in LP and EA, respectively). The chemical profiles and seasonality of each factor are displayed in Fig. 4 and Fig. 5, respectively, and One of the advantages of performing a multisite PMF in the present study is the possibility to differentiate between two traffic profiles that could hardly be observed in the individual solutions. Similarly, some factor profiles that remained mixed in the single-site-solution for one site were polished as a result of combining both datasets. That was the case for the dust, MSA-rich, traffic 2, and non-exhaust profiles (Single site solutions can be found in the SI for comparison with the multisite solution.

3.2.1 Dust

This factor is the major contributor to the observed PM_{10} mass at both sites and is traced by crustal elements, such as Al, Fe, Ti, Mg, Mn, Ca, Na, K, V, Rb. The confidence interval for these species is narrow around the average displacement value, indicating that these species are mainly the ones that define this source profile. The presence of other elements, including sulfate, OC, Zn and Pb (with tight confidence interval), along with EC and Cu (with confidence intervals that allow negligible

concentrations), supports the influence of road traffic in this source, through road dust resuspension. This factor has an average contribution of 32% (Ambient: 10.6±7.6 μg m^{-3}, STP: 15.7±11.2 μg m^{-3}) to the total PM_{10} mass observed in El Alto during the measurements period, and 20% (Ambient: 5.5±4.1 μg m^{-3}, STP: 8.0±5.7 μg m^{-3}) in the city of La Paz. This factor significantly contributes to the difference in PM mass concentrations observed between La Paz and El Alto. The factor contribution can rise up to 46% of the mass in El Alto during winter time (specifically in June), whereas its percentage contribution in La Paz reached their maximum during the transition month of October (27%).

The difference in contribution between these two sites can be attributed to difference in La Paz and El Alto characteristics. Particularly, El Alto is a fast-growing city located on the edge of the Altiplano region, a dry and arid area with mostly unpaved streets and active construction works. On the other hand, the city of La Paz shows to be less influenced by this factor, likely due to a higher fraction of paved roads compared to El Alto. Additionally, La Paz is situated at a lower elevation, surrounded by mountains and hillsides, which reduces the impact of strong winds from the Altiplano. Although both stations were considered to represent urban background, the terrain surrounding the two stations is very different. The El Alto station is located in the middle of the airport facilities, in a rather dusty area, while the La Paz station is located on the rooftop of a building located in the middle of the city. Nevertheless, combining the time series obtained from the PMF analysis for this factor and the meteorological information from both sites, it was observed that the highest contributions from this factor were associated with higher wind speeds coming from the North West (NW). The seasonality observed in this factor is also consistent with the variation in precipitation favoring the main removal mechanism of dust in air (i.e., wet deposition). Similar contributions of dust to PM_{10} (with comparable or lower mass concentrations) have been reported by other studies in South America, like Sao Paulo: 25.7% (11.3 μg m^{-3}, Pereira et al. 2017a), Bogotá: 28% (10.5 μg m^{-3} (STP), Ramírez, et al 2018), and Quito: 19.11-20.79.% (4.8-5.3 μg m^{-3}, Zalakeviciute et al. 2020) (Absolute mass concentrations of dust [μg m^{-3}] were calculated based on the percentage contributions reported on the studies mentioned and the reported average PM mass concentrations).

3.2.2 Primary biogenic aerosol (PBA)

The Primary biogenic aerosol (PBA) factor is associated with the highest fraction of polyols, which serve as tracers of soil and fungi activity, as well as plant debris (Elbert et al., 2007; Samaké, et al., 2019a, b). The following most important contributors to this factor, with narrow confidence intervals are OC, K^+ and heavier alkanes, species that have been observed accompanying this source in other similar studies (Borlaza, et al., 2021; Chevrier, 2016). On average, PBA contributes 7 and 13% (1.5±1.0 μg m^{-3} and 2.8±1.8 μg m^{-3}) to the annual PM_{10} mass observed in El Alto and La Paz, respectively. However, its contribution increased up to 11 and 17% (2.1±1.1 μg m^{-3} and 3.4±1.7 μg m^{-3}) of the mass concentrations during early autumn (March-April). Minimum concentrations were observed during winter. Similar results were found in France by Chevrier (2016) and Samaké et al. (2019b) where maximum concentrations of primary biogenic tracers were observed between late spring and early autumn. Highest contribution of this factor was observed in late summer (February) in La Paz 4.4±2.4 μg m^{-3}, becoming

the second largest source in terms of mass during this month (28%). However, it should be noted that fewer number of samples collected in the rest of the summer months. Higher contributions of this factor were consistently observed in LP compared to EA, most likely due to its closer proximity to vegetation (both, local and in the valleys to the East).

3.2.3 MSA rich

This factor is predominantly identified by MSA, accounting for 100% of the MSA present in the samples. A small fraction of OC, V, Mn, Zn, and certain heavy alkanes is also present in this factor, suggesting a potential minor contribution from anthropogenic sources. It contributes to 7% (2.0±0.9 μg m^{-3} and 2.0±1.4 μg m^{-3}) to the observed PM_{10} mass in El Alto and La Paz. MSA is known to result from the oxidation of the primary emissions of dimethylsulfide (DMS) typically produced by marine phytoplankton, however studies have shown other possible sources of DMS as terrestrial biogenic sources, forest biota or lacustrine phytoplankton (Saltzman et al., 1983; Jardine et al., 2015; Du et al., 2017; Ganor et al., 2000). No clear seasonality was observed, except for the slight decrease in concentrations in the months of March and October.

Neither back trajectory analysis nor association with local wind direction were useful to elucidate on the specific origin of this factor. However, Aliaga et al. (2021) showed that air masses passing by the Titicaca Lake formed part of the third main air mass pathway arriving to the nearest GAW station (CHC-GAW) between December 2017 and May 2018. Moreover, Scholz et al. (2022) showed that the observed DMS in CHC-GAW during the same period was mostly linked to long-range transport of marine air masses, with a smaller contribution from the Titicaca Lake. Considering that air masses originating from the coast do not represent an important source of PM in the conurbation, terrestrial or lacustrine sources could be more likely to be the origin of this factor. The Titicaca Lake, the largest freshwater lake in South America, is located about 50 kilometers outside the metropolitan area (about 50 km) and long-range transport of air masses from the Amazon can also be observed at the sampling sites.

3.2.4 Secondary sulfate

This factor contributes to 8% of the total observed mass concentrations at both sites (1.9±2.1 μg m^{-3} and 2.0±2.1 μg m^{-3} in El Alto and La Paz, respectively) and is characterized by the presence of sulfate and ammonium. This factor is generally associated to long range transport of air masses in preceding European studies (Amato et al., 2016; Borlaza et al., 2021; Waked et al., 2014) due to the time scales and conditions necessary to form ammonium sulfate from its gaseous precursors: sulfuric acid (H_2SO_4) and ammonia (NH_3) (Viana et al., 2008). Additionally, a small fraction of other inorganic elements such as Na, K, Mg, Ca are also found with tight confidence intervals in this factor. These elements have been observed to be associated with sulfate rich factors in previous European studies, occasionally linked to long-range transport factors (aged sea salt) (Weber et al., 2019; Veld et al., 2021; Borlaza et al., 2021; Dai et al., 2020). Nevertheless, the small contribution of Zn and some heavy alkanes in the factor shows there could also be an influence of local sources to this factor. This could be attributed to the relaxed regulations of sulfur concentrations in imported fuels (<5000 ppm for diesel and <500 ppm for gasoline, Decree

1499/2013 of the Bolivian government), which represents 41 to 46% of the national fuel consumption (Correo del Sur, 2022). Furthermore, this factor also includes a small fraction of OC, that could originate either from anthropogenic emissions or from biogenic SOA formation (Borlaza et al., 2021).

The highest contributions from this factor were observed during October and November (local spring) where favorable conditions for ammonium sulfate formation are met, including strong solar radiation, moderate temperature and relative humidity (Korhonen et al., 1999; Karamchandani and Seigneur, 1999). A similar temporal variability was observed in the city of Arequipa (Peru) (Olson et al., 2021), the closest urban high-altitude large agglomeration (ca. 2300 m a.s.l., ~1 million inhabitants) located 300 km to the west of LP-EA. The aforementioned study found urban combustion emissions to be the main sources of sulfate aerosols in the city (50%), followed by dust (20%), despite its proximity to the coast and to the Central Andes volcanic region. However, it is important to highlight that an increase in sulfur concentrations associated to an increase in the regional volcanism activity occurred during the same period (Manrique et al., 2018; Masías et al., 2016), which could contribute to the observed seasonality. Nonetheless, the fact that the average contributions of this factor to total PM_{10} are nearly identical in both cities indicates an even distribution of this factor throughout the metropolitan region. Although the overall contribution of this factor to total PM is relatively low compared to other factors, it accounts for 14-15% of the observed mass in both sites during spring, while comprising only 3-4% of the total mass during winter.

3.2.5 Secondary nitrate

This factor is responsible of 53% of the nitrate found in the samples and represents the second largest source for the ammonium found at both sites (23%). This factor also exhibits a secondary contribution with a narrow confidence interval for EC, OC, Zn, Pb, and heavy alkanes, tracers of traffic emissions. This evidences that the main source of the nitrates observed in La Paz and El Alto is linked to the combustion of fossil fuels, and is mostly locally produced from the oxidation of NO_x emitted from traffic. Previous studies of emission inventories in the country have also estimated that mobile (transportation-related) sources to be the main source of NO_x (Herbst, 2007; Pareja et al., 2011).

The contribution of this factor to total PM_{10} was of 9 and 6% (2.3±2.0 μg m^{-3} and 1.6±1.6 μg m^{-3}) in El Alto and La Paz, respectively. Higher concentrations are observed in El Alto compared to La Paz. Since NO_x concentrations were not monitored at either of the stations, we can only speculate that the difference between La Paz and El Alto is partly attributed to the difference in ambient temperature between both cities, as colder temperatures favor the partitioning of nitrate in particulate phase.

3.2.6 Biomass combustion

The main source of for biomass burning pollution in the tropical South America is agricultural practices and land use change (Mataveli et al., 2021). Although it is not a common practice in the Andean region, long-range transport of air masses coming from the Bolivian lowlands and neighboring countries contributes to PM at both sites. The main species represented in this

factor are OC, levoglucosan, mannosan, and K^+, which are typical tracers of biomass burning (Simoneit and Elias, 2000; Simoneit, 2002; Li et al., 2021). While 100% of mannosan is explained by this factor, only 76% of the levoglucosan present in the samples can be explained by this source (despite the applied constraint). Low contributions of EC to this factor produce a median OC/EC ratio of 17.8. This factor exhibits similar annual contributions of 9% and 8% to PM_{10} concentrations in La Paz and El Alto, with maximum average contributions of 17% and 13% (6.4±5.4 μg m^{-3} and 5.4±4.7 μg m^{-3}) in the middle of the dry season (July-September), peaking in August. In contrast, concentrations during autumn are much lower (1.0±1.0 μg m^{-3} and 1.3±0.9 μg m^{-3}). The median levoglucosan to mannosan ratios (Lev/Man=9.1) of this profile were found to be close to ratios previously reported for sugarcane burning (one of the main plantations in the Brazilian Amazon region) in laboratory and field studies (Hall et al. 2012: 10; Pereira et al. 2017a: 11; Pereira et al. 2017b: 12, Zhang et al., 2015). The difference between cities in the observed concentrations assigned to this factor during the biomass burning season might be explained by the fact that EA, located higher up on Altiplano, is potentially less influenced by long range transport from the low lands. Although agricultural biomass burning practiced in the Andean valleys and the Amazon region of Bolivia and neighboring countries has a relatively low annual contribution, it is important during the dry season. On days when PM_{10} concentrations exceeded the short-term exposure AQG recommended by the WHO (45 μg m^{-3} in 24-hr), the biomass burning factor accounted for 13% of the total mass in EA (7.0±5.9 μg m^{-3}) and 23% in LP (11.9±7.4 μg m^{-3}, making biomass burning the second most important source of PM after dust during those episodes.

3.2.7 Non-exhaust vehicular emissions

This factor is identified by the presence of metals such as Cu, Sn, Sb, and Pb, along with a significant contribution of Fe in terms of mass. These species have been previously identified as tracers for break and tire wears (Amato et al., 2011; Charron et al., 2019; Fukuzaki et al., 1986), generated by vehicles through mechanical abrasion. However, some studies have also found these tracers to be associated with industrial emissions (La Colla et al., 2021), for which we could not entirely neglect the possibility of having an influence of industrial emissions masked within this factor. This factor appeared at an early stage in the single site PMF in El Alto but it was not observable in La Paz. The multisite PMF allowed to clearly identify this factor in La Paz, splitting it from another traffic related source. This factor contributes to 3% of the total PM_{10} mass at both sites, with slightly higher contributions during the dry season, following a similar seasonality pattern as the dust factor. However, this factor frequently presents high concentration spikes in El Alto that are not observed in La Paz.

3.2.8 Open waste burning

With the inclusion of PAHs and alkanes into the PMF analysis, a specific factor tentatively associated to waste burning was identified. This factor is characterized by the presence of levoglucosan, K^+, EC, OC, metal species such as Al, Ti, V, Rb, Pb, PAHs and alkanes. It accounts for 57% of the Triphenylene observed in the samples. This factor also contributes in median to 10-20% of the observed concentrations of PAH_1, PAH_2, BaA and Chr, and 15 to 35% of the measured alkanes. It represents

the second major source of the observed alkanes. Although Cl^- was not included in the final PMF solution due to the instability it added to all the explored solutions, preliminary runs indicated a significant fraction of total Cl^- associated with this factor. A Spearman correlation >0.67 was found between the concentrations of Cl^- and the PM concentrations attributed to this factor (Table S6). These elements are typical byproducts of the combustion of plastic mixed with vegetation or wood (Simoneit, 2002; Cash et al., 2021; Kumar et al., 2018; Rivellini et al., 2017; Lanz et al., 2008; Guttikunda et al., 2019, 2013; Christian et al., 2010; Singh et al., 2008). Similar factors have been observed in prior studies (Pereira et al., 2017a; Zíková et al., 2016; Rai et al., 2020), although only few of these studies were able to distinguish it as a separate factor from biomass burning or traffic, given the ubiquity of some of the tracers.

On an annual average, the total mass of PM_{10} attributed to this factor amounts to only 5% and 2% (1.8±1.8 μg m^{-3} and 0.8±1.2 μg m^{-3}) in El Alto and La Paz, respectively. However, during winter, its contribution can increase to 9 and 6% (3.4±1.6 μg m^{-3} and 2.1±1.2 μg m^{-3}). The seasonality of this factor is evident, with higher contributions in May and decreasing contributions in August. Although the exact source of this factor remains unidentified, the higher contributions in El Alto compared to La Paz suggest the presence of local sources within the El Alto area. Analysis of wind characteristics shows that higher concentrations of this factor are linked to low wind speeds blowing from the North in the case of El Alto, and from the northwest and with higher wind speeds in the case of La Paz (Fig. S7). The local emissions could originate from punctual-sources of waste burning, or the emissions of industrial and open commercial areas in El Alto, later transported to the city of La Paz. Similar behavior was observed when associating Cl- to wind speed and wind direction (not presented here).

3.2.9 Traffic sources 1 and 2 (gasoline/diesel)

The first resolved traffic factor (TR1) is annually responsible for 6 and 8% of the observed PM mass in La Paz and El Alto, respectively (1.9±2.0 μg m^{-3} and 2.3±2.0 μg m^{-3}). The main tracers of this factor are a small fraction of EC and OC, the presence of metals such as Na, Ca, Mg, Al, Fe, Ti, V, Mn, Zn, Rb, Pb, and over 40% of most PAH concentrations, consistent with previously observed vehicular emission factor profiles (Amato et al., 2011; Waked et al., 2014; Charron et al., 2019). Some traces of sulfate, and lighter alkanes can also be observed in the chemical profile of this factor.

The second traffic factor (TR2) contributed with an average of 23% and 13% to total PM_{10} in La Paz and El Alto, respectively (5.7±3.5 μg m^{-3} and 3.6±2.5 μg m^{-3}). The chemical species identified in this factor are similar to those found in TR1, including: EC, OC, Zn, PAH_1 and Cor, with small contributions of sulfate, Na, Ca, Mg and Mn. It is noteworthy that no alkanes and almost no hopanes are found in TR2, even if these compounds are in principle emitted by road traffic.

The median OC/EC ratios obtained from the traffic chemical profiles of TR1 and TR2 are 0.4 and 1.1, respectively The low OC/EC ratios observed in high-altitude conditions are not surprising, as combustion processes are less efficient under low O_2 availability (Wang, et al., 2013a). However, because of the very different conditions for combustion, literature values of the ratio OC/EC (> 1 for gasoline, and < 1 for diesel, Brito et al., 2013, Cheng et al., 2010; Cheng et al., 2021; Wong et al., 2020;

Yang et al., 2019) were not useful to identify which of the traffic factors can be associated to gasoline- or diesel-powered vehicles, being both OC/EC ratios in the present study close to 1.

A key distinction between the two traffic factors is the Mn/Zn ratio, with TR1 exhibiting a ratio greater than 1 and TR2 showing the opposite trend. The fuel analysis (pre-combustion) revealed that the largest difference in the chemical composition between local gasoline and diesel fuels was the relative abundance of Mn compared to Zn. Whilst the measured prior-combustion ratios of Mn/Zn are not preserved, the Mn/Zn ratio remains a characteristic feature of each profile. Additionally, TR1 have higher PAH concentrations, whereas TR2 shows much lower contributions of PAHs. Previous studies have demonstrated that gasoline-powered vehicles indeed emit more long-chain PAHs than diesel fuel (IFP, 2021; Leotz-Garziandia et al., 1999; Zielinska, et al., 2004a). While gasoline-powered vehicles represent over 80% of the vehicle fleet in Bolivia, literature has shown that diesel-powered vehicles can emit 10 to 30 times more particles than gasoline-powered vehicles (Zielinska et al., 2004b).

In terms of contribution, TR2 has a greater overall influence than TR1 in La Paz and is almost twice as influential as TR2 in El Alto. This difference could be related to the difference in the topography, as previous studies have shown that steep slopes can significantly increase the vehicle fuel consumption (Carrese et al., 2013; Wang and Boggio-marzet, 2018). Additionally, the proximity of the LP sampling site to the nearest main avenue (~100 m) and to the parking lot of the municipality buses (~100 m, horizontal distance; ~45 m vertical distance), which are diesel powered vehicles, may play an important role in the respective influences of TR1 and TR2 in LP.

The PD-SID comparison of both traffic factors with the road traffic profiles of several urban/urban-background French sites presented in Borlaza (2021) and Weber (2019) (SI) revealed there is a significant similarity between TR2 and the French road-traffic factors (where diesel is the dominant fuel used). However, TR1 exhibits PD values outside the similarity thresholds established by Pernigotti & Belis (2018).

Based on the previous description of factors TR1 and TR2, we consider likely that TR1 is related to the emissions from gasoline-powered vehicles, whereas TR2 is most likely associated to diesel-powered vehicles. However, the number of registered cars reported by the Municipal Tax Administration in 2011 showed that the number of gasoline-powered vehicles in the city of La Paz (~90% of the registered vehicle fleet in La Paz) was 2.4 times larger than the ones registered in El Alto (~80% of the registered vehicle fleet in El Alto). In contrast, similar number of diesel-powered vehicles were registered at both sites. If these numbers were directly related to the flow of vehicles in the metropolitan area, they could imply the opposite of what can be concluded from the chemical profiles, i.e. TR1 associated to diesel powered-vehicles and TR2 associated to gasoline-powered vehicles. However, it should be noted that vehicle registration does not necessarily imply those are operating vehicles. This could be especially the case for trucks and buses that move between La Paz and El Alto. In addition, it is known that large contributions of emissions could come from a small number of vehicles (Ježek et al., 2015; La Colla et al., 2021; Brito et al., 2013). These factors make it challenging to estimate the contribution of the different types of vehicl es circulating in the metropolitan area to the measurements obtained from the filters.

Together, TR1 and TR2 constitute the major source of particulate matter in La Paz, and the second largest source of PM_{10} particles in El Alto. TR1 displays a slight seasonality with higher concentrations during the dry season of 2016. On the other hand, TR2 does not display significant seasonality, except for higher concentrations observed between April-May 2016 and May-June 2017. Although, one might similar variability for traffic-related profiles, this is not the first study to observe a difference in the yearly variability of gasoline and diesel emissions (Squizzato et al., 2018, for a study in New York State).

3.2.10 Lubricant oil

The inclusion of molecular organic species (PAH, alkanes, and hopanes) enabled the identification of a factor associated with lubricant combustion, likely originating from vehicle emissions. This factor is marked by the presence of hopanes and alkanes in the chemical profile, which serve as unequivocal tracers of oil combustion (Charron et al., 2019; El Haddad et al., 2009). It contributes to 36-47% of the total mass of alkanes present in the samples, and constitutes the major source of hopanes, accounting for 65% of their total mass. Additionally, this factor presents smaller percentage contributions of OC, K^+, Na, Ca, V, Mn, Cu, Zn, and certain PAHs, elements commonly present in fuel combustion emissions. The contribution of this source to annual PM_{10} mass is of 3 and 1% (0.9±0.8 µg m^{-3} and 0.4±0.6 µg m^{-3}) in La Paz and El Alto, respectively. A clear increase in contributions during the coldest months of the year can be observed in the variability of this factor. A similar evolution of the hopanes with maximum concentrations during winter was observed in Marnaz (France) by Chevrier (2016). Likewise, a study in three cities of the United States of America (USA) observed an increase of concentrations of hopanes and alkanes during the coldest months of the year (Kioumourtzoglou et al., 2013). This seasonality could be associated to the cold start of vehicle engines in the early morning and late-night hours, in the period when minimum temperatures decrease.

Overall, the contribution of this factor is greater in the city of La Paz compared to El Alto. This discrepancy can be attributed to the additional strain experienced by vehicle engines while navigating the steep streets of La Paz, a challenge that is less pronounced in El Alto due to its flat topography. Although the contributions of this factor to total PM_{10} mass are relatively low, its significance in terms of air quality should not be underestimated, as it represents one of the major sources of alkanes and hopanes. The latter compound is considered hazardous for human health since it has proven to be associated to systemic inflammation biomarkers (Delfino et al., 2010).

3.3 Methodology discussions

The sampling strategy, the complete chemical characterization, and the multisite PMF, coupled with the specific geographical patterns, enabled this quite unique study to offer an extensive characterization of PM sources in high-altitude cities. The present investigation provides important information that can help policy-making towards better air quality in the region, however, we are aware of some limitations.

- PMF limitations

Having enough samples in the multisite approach and a fairly large chemical speciation including organic tracers allowed the identification of 11 factors in the PMF analysis. It is noteworthy that only a few studies have been able to resolve similar number of sources with good statistical indicators (Chevrier, 2016; Pandolfi et al., 2020; Waked et al., 2014; Weber et al., 2019; Borlaza et al., 2021). Nevertheless, attempting a larger number of factors generated instability in the otherwise geochemically stable profiles. Several factors may contribute to this limitation, including:

- Collinearity between sources, resulting in mixed factors. The presence of OC in both secondary sulfate and primary biogenic emissions could speak of a possible mixing of these factors with biogenic secondary organic aerosols (BSOA). A detachment of BSOA was not possible due to the lack of the specific tracers of this source (3-MBTCA, or cellulose, or methyltetrols).
- Although the industrial sector is not highly developed, there are factories within and the vecinity of the Metropolitan area that were not resolved by the PMF (e.g. cement plants, brickyards, PVC manufactory plants). This could be due to the lack of specific tracers for these sources in the analysis, a similarity of the chemical profile and temporal variability of the emissions compared to the resolved sources, or simply because they represent a very small fraction of PM_{10}
- The removal of chloride from the analysis for bringing instability to the solution. This instability was likely associated to the large variability of this volatile compound.

- Multisite approach limitations

While the multisite approach has added enhanced the findings compared to a single-site approach, it is important to note that *a priori* this method cannot be directly applied to sites that differ greatly from each other. It was important to verify the similarity of the single-site solutions. However, one drawback of the multisite approach is that it enforces the similarity of the common factors found between the two sites, smoothing out the specificity of them. Examples of this forced similarity are as follows:

- The multisite approach successfully separated EC (a traffic tracer) from the dust profile. However, considering that the Altiplano is a major source of dust and that the only path that the air masses take when transporting dust from the Altiplano to La Paz is traversing both cities, it is not surprising that the dust factor in the city of La Paz (single-site solution) is highly influenced by traffic tracers. For the multisite solution the indirect information of the mixing of sources during transport is lost.
- The average molar ratio of sulfate and ammonia concentrations differs between the two cities (2.05 and 1.63 in El Alto and La Paz, respectively), indicating a lower availability of ammonium to neutralize sulfate and nitrate ions in the city of La Paz. However, this distinction is no longer evident in the multisite analysis, which yields a median molar ratio of 1.96 representative of both cities.

- The MSA-rich profile in El Alto exhibits strong mixing with metallic species, among them crustal material, which hinted its path through the Altiplano towards the city of El Alto. This mixing pattern is no longer evident in the multisite analysis.

Considering the advantages of a more specific characterization of sources provided by the multisite approach outweigh the associated drawbacks, we believe it was the most appropriate technique to apply in the metropolitan region of La Paz and El Alto with such database in hand.

4 Conclusions

This study presents innovative information and a unique analysis of air pollution sources in the high-altitude urban environment of the fast-growing cities of La Paz and El Alto in Bolivia. It also provides a detailed description of the chemical profiles of 11 identified source types, resolved by the multisite PMF method, along with their temporal and spatial variability. The extensive and comprehensive dataset, combined with the inclusion of inorganic and organic species in the analysis, enabled an advanced source apportionment beyond classical solutions, allowing for the identification of several biogenic and combustion-related factors that would have remained unresolved otherwise. Notably, waste burning was separated from biomass burning, and traffic exhaust emissions were separated into two independent profiles.

On average, vehicular emissions represent 35 and 25% of the PM_{10} concentrations measured in La Paz and El Alto, respectively. Then, dust emerges as one of the two main sources contributing to 20 and 32%. Factors associated with secondary inorganic aerosols account for 22 and 24% and the primary biogenic emissions account for 7 and 13% annually. Although one of the smallest factors in terms of contribution to the total mass, the non-regulated burning of waste, predominantly occurring in El Alto between May and August, is the second most significant factor responsible for observed PAH concentration levels. The observations in this study were made at urban background sites, representing wider regional pollution levels in La Paz and El Alto. Locally, especially near roads or landfills, the mass concentrations are expected to be higher. While most of the resolved sources are associated with local activities (dust resuspension, primary and secondary vehicular emissions, and waste burning), there is a significant contribution of regional natural and anthropogenic sources of PM (Primary and secondary biogenic emissions, and biomass burning).

Based on our findings, we can outline relevant actions towards the improvement of air quality in La Paz and El Alto:

1) Regulation of vehicular emissions have to improved. As the Metropolitan area continues to grow, more efficient means of transportation and stricter policies and control on combustion practices are needed to ensure that air quality is not further degraded.

2) Waste burning should be prohibited. It is a major source of PAHs and other pollutants with high human health risk factor.

3) Agricultural biomass burning is a seasonal source, a decrease in their emissions would result in a significant improvement in the air quality during the most polluted season, not only for the metropolis but also for the rest of the country.

4) Dust is an important source in terms of mass that has an anthropogenic component (e.g. vehicle resuspension, construction activities, mining) and should be addressed.

5) Updated policies of pollutant emissions are essential to regulate also the growing industry sector.

In order to have a comprehensive understanding of the pollution sources in the metropolitan area of La Paz and El Alto, information on the gaseous components is of utmost importance. A longer sampling time period together with an updated emissions inventory of the resolved sources would be beneficial for a better understanding of the resolved sources and their evolution in time. Furthermore, analyzing the potential impact on health of the resolved sources is crucial for efficiently targeting the most hazardous sources of PM.

Code availability.

The software code is available upon request.

Data availability.

The chemical and PMF datasets are available upon request.

Authors contribution.

GU, MA, PL, JLJ, AA, JLB, RK, IM, NP and AW participated in the conceptualization of the experimental set up and design. IM participated in the data curation. VM, MP and LJB participated in the formal analysis and the development of the methodology. GU, MA, PL, JLJ, AA, JLB, RK, PG were involved in the funding and resource acquisition. JLB, IM, NP and VC contributed to the investigation by organizing the samples collection and performing the experiments. GU, MA, PL, MP, LJB, GM and JLJ helped with mentoring, supervision and validation of the methodology, techniques and results. VM was responsible for the data processing and the writing of the original draft. GU, PL and JLB revised the original draft. All the authors reviewed and edited the manuscript.

Competing interests.

G. Močnik is employed by Haze Instruments d.o.o., the manufacturer of the aerosol instrumentation.

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Figures

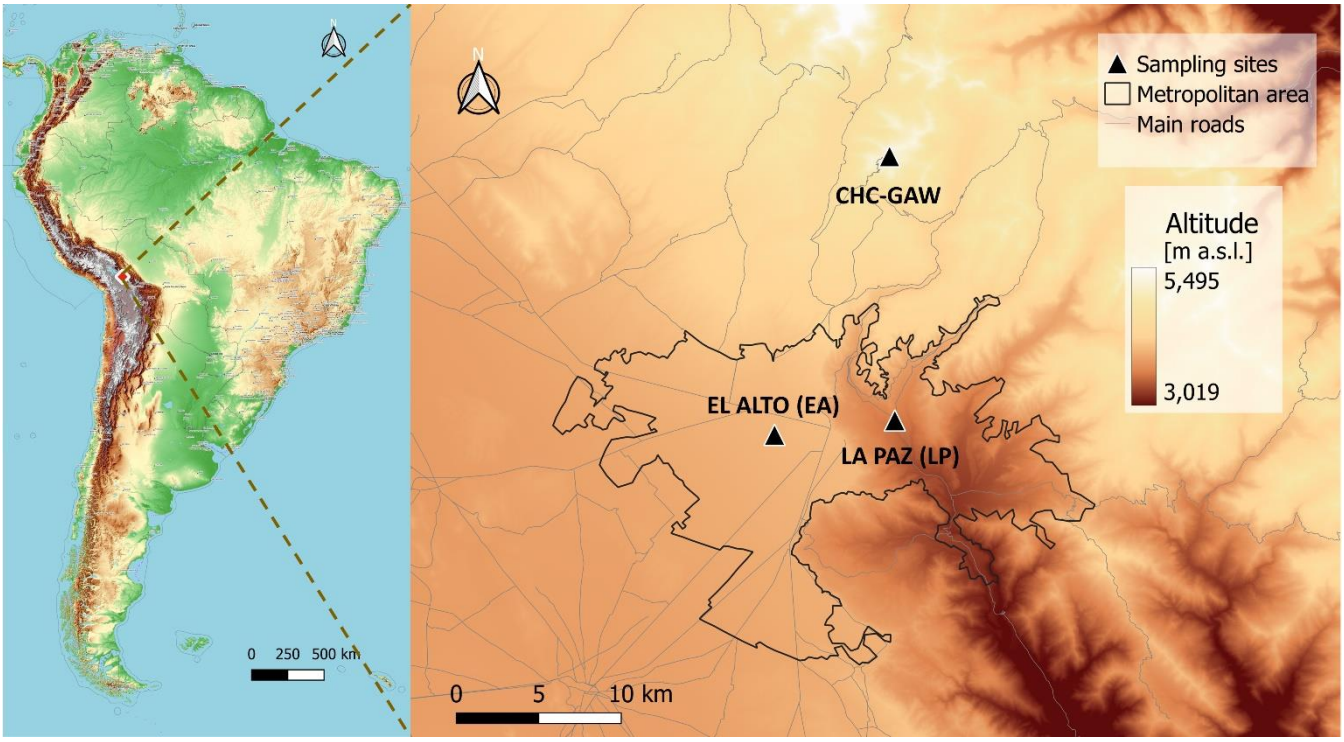

Figure 1. Geographical location of the sampling sites (left panel) La Paz (LP) and El Alto (EA) zoomed in (right panel) and positioned with respect to the regional Chacaltaya-GAW monitoring station (CHC-GAW). Color scale represents the altitude above sea level.

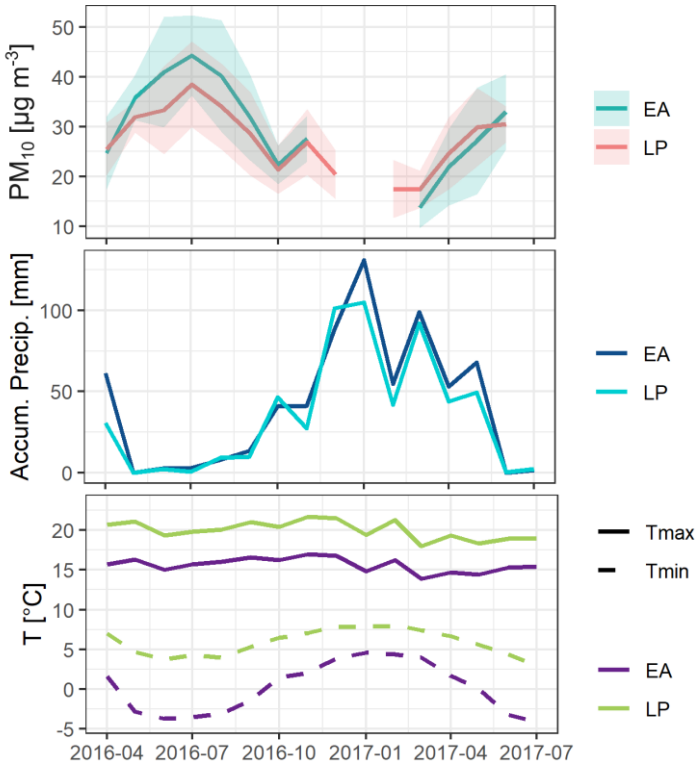

Figure 2. Monthly PM_{10} mean concentrations (µg m^{-3}), monthly accumulated precipitation (Accum. Precip., mm), and monthly mean maximum/minimum temperature (°C).

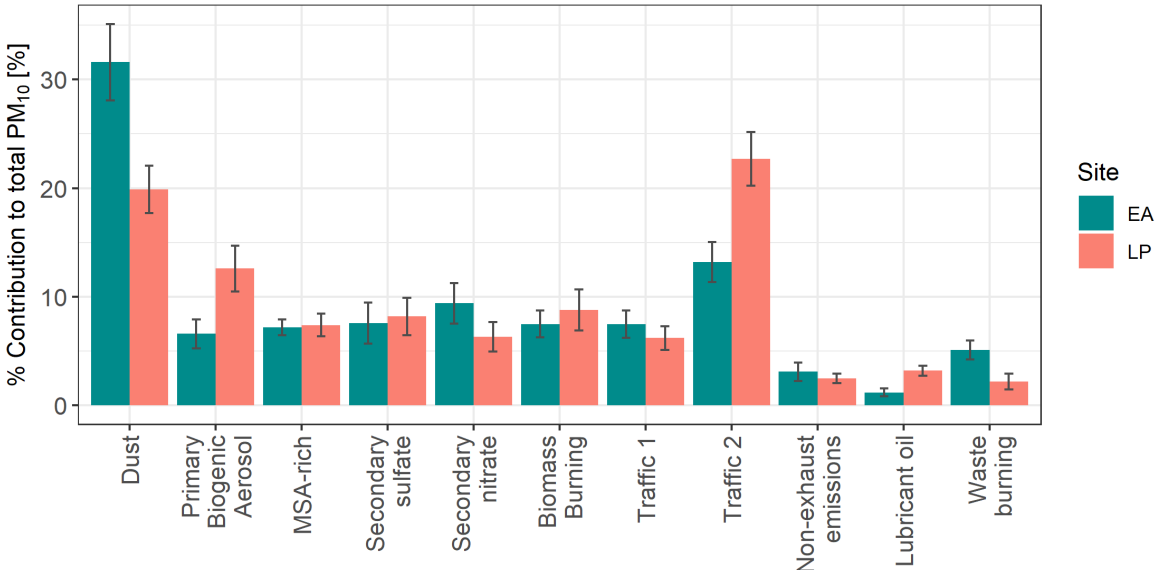

Figure 3. Average factor contributions to total PM_{10} at each site, resulting from the multisite PMF. The bars represent the 95% confidence interval of the mean values.

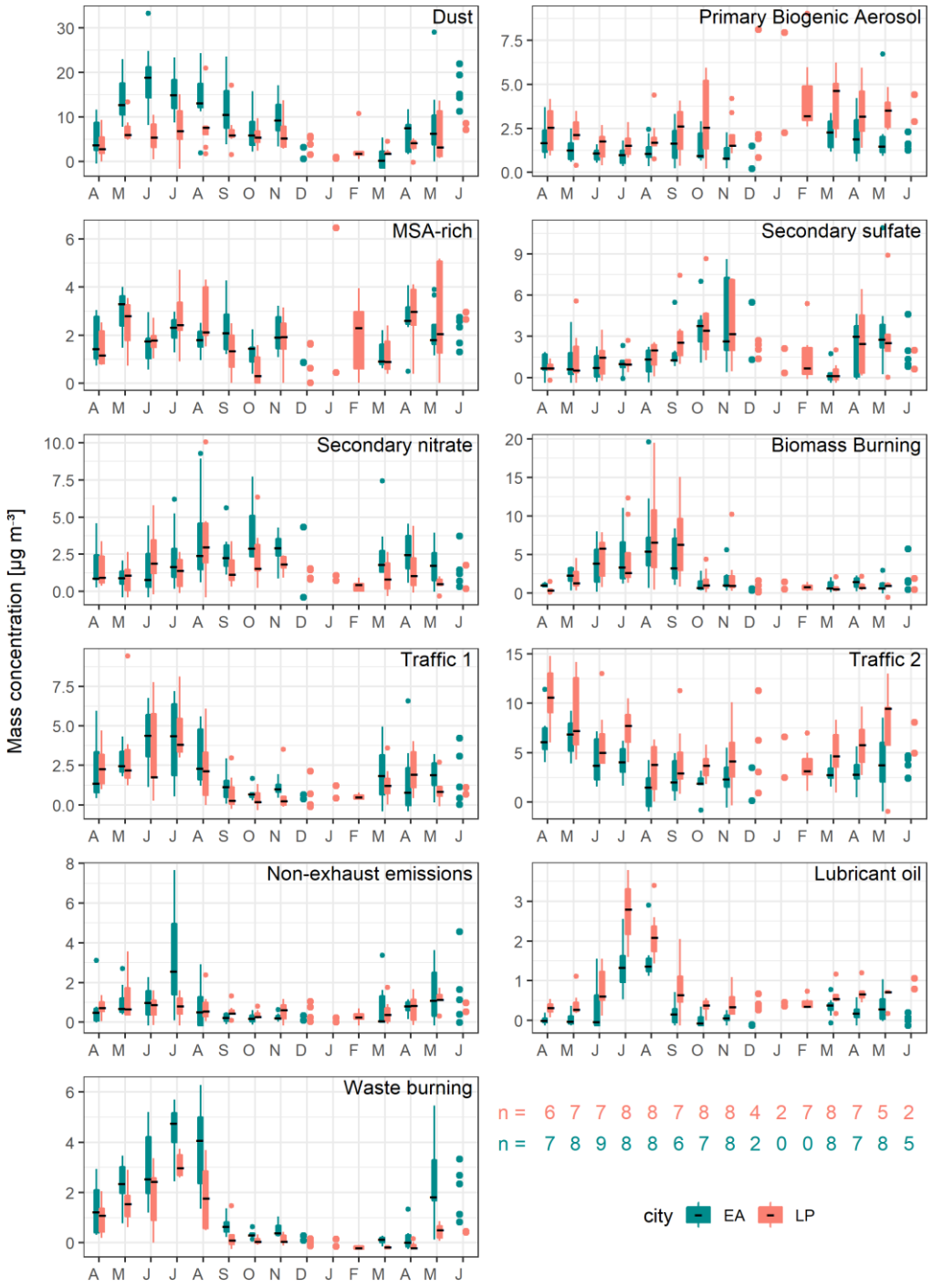

Figure 4. Source mass-contribution monthly variations (n = number of modelled data points included in the average) between April 2016 and July 2017.

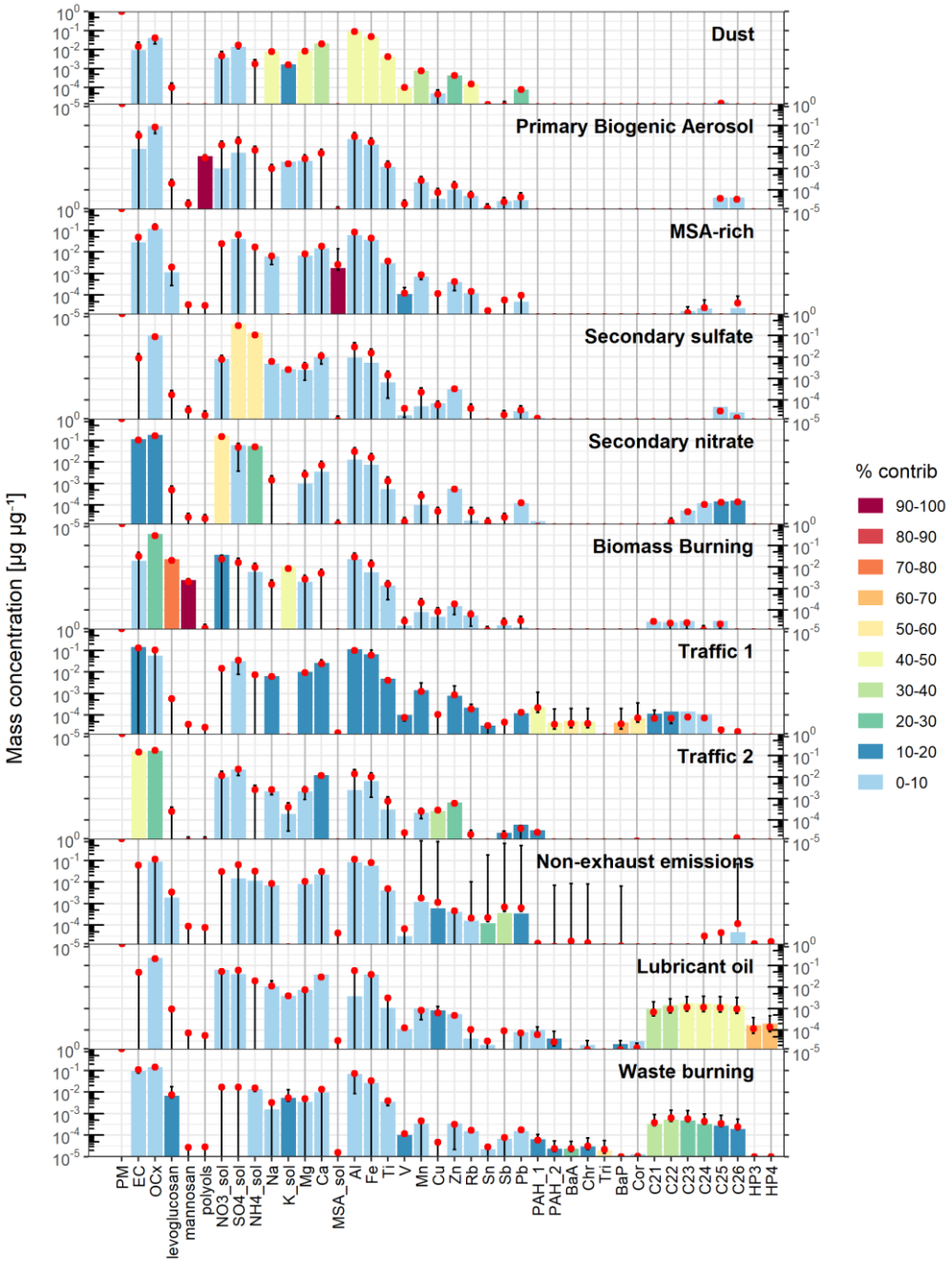

Figure 5. Source chemical profiles (bars representing median bootstrap mass contributions of each species per µg of PM mass attributed to each source in y-axis, red dots represent mean DISP values, error bars represent DISP confidence intervals, color scale represent the contribution in percentage). The name of each source is further described and developed in the individual factor descriptions[1].

[1] PAH_1: [BghiP]+[IP]+[BbF]; PAH_2: [Fla]+[Pyr]).

Table 1. Set of constraints applied to final solution

FACTOR	SPECIE	CONSTRAINT	VALUE
Biomass Burning	Levoglucosan	Pull up maximally	%dQ 0.50
Biomass Burning	Mannosan	Pull up maximally	%dQ 0.50
Primary Biogenic Aerosol	Polyols	Pull up maximally	%dQ 0.50
MSA-Rich	MSA	Pull up maximally	%dQ 0.50

Table 2. Air quality studies at high-altitude Latin American cities.

	Average PM_{10} (Min-Max) [µg m^{-3}]	Period	Study	Population[i]	Altitude [m a.s.l.]
Mexico City, Mexico	(45.38-80.10) [ii]	2015-2016	(Cárdenas-Moreno et al., 2021)	18,457,000	2,850
Quito, Ecuador	24.9-26.2[iii,iv]	Jan 2017-Dec 2018	(Zalakeviciute, et al., 2020)	1,793,000	2,240
Bogota, Colombia	37.5 (9.89-160) [iii, iv]	Jun, 2015-May 2016	(Ramírez, et al., 2018)	9,989,000	2,620
El Alto, Bolivia	29.9 (6.6-59.0) [iii,v]	April 2016-June 2017	Present study		4050
La Paz, Bolivia	27.2 (11.6-50.9) [iii, v]	April 2016-June 2017	Present study		3200-3600

[i] https://populationstat.com/
[ii] Range of spatial variation
[iii] Range of seasonal variation
[iv] Concentrations reported in standard conditions of temperature and pressure
[v] Campaign average PM_{10} concentrations that could slightly over estimate annual mean values due to a low number of samples collected during the wet season, where the minimum mass concentrations expected.