An Investigation of the Precipitation Net Effect on the Particulate Matter Concentration in a Narrow Valley: Role of Lower-Troposphere Stability

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ABSTRACT

Precipitation modulates the pollutant concentration in the atmosphere directly and indirectly through different mechanisms. Wet deposition, including in-cloud scavenging and below-cloud scavenging, is one of the main drivers of particulate matter removal from the atmosphere. Our focus is to assess the net effect of precipitation on the pollutant concentration using observations, a nonparametric conditional analysis, and the signed overlapping coefficient complement index. The study considers not only the direct near-surface aerosol below-cloud scavenging, but also the indirect effect associated with the induced lower-troposphere stability limiting the vertical dispersion, leading to a pollutant concentration increase. We consider the precipitation effects on fine and coarse particulate matter concentration. Probability density functions of the pollutant concentration during precipitation events and under dry conditions are analyzed for every hour of the day. The net effect of precipitation on the fine and coarse particulate matter concentration is strongly dependent on the atmospheric stability. During the nighttime and before midmorning, the atmosphere is stable; a precipitation event during this time induces below-cloud scavenging, reducing the concentration of particulate matter in the atmosphere. During the afternoon, unstable atmospheric conditions are predominant, and aerosols disperse vertically; a precipitation event during this period stabilizes the atmosphere, generating early stabilization that, together with the continued anthropogenic emissions during the day, leads to near-surface pollutant accumulation, offsetting the washout effect of the precipitation.

1. Introduction

During recent decades, atmospheric pollution has become one of the foremost environmental concerns for expanding metropolitan areas around the globe. The population and economic growth, the increase in energy demand, and the expansion of flawed transportation models have led to the rise of pollutant emissions into the atmosphere, worsening air quality conditions in urban areas (Mayer 1999; Li et al. 2015). In megacities such as New Delhi (India) and Beijing (China), extreme air pollution episodes are experienced frequently, with hourly fine particulate matter (PM_{2.5}) concentrations exceeding 400 $\mu\text{g m}^{-3}$ (Yang et al. 2015; Marlier et al. 2016) and average annual

concentrations above 90 $\mu\text{g m}^{-3}$ (Cheng et al. 2016). The air quality problem is widespread globally, and it is part of the ecology of cities where pollution generation outpaces the societal capacity to implement control measures (Grimm et al. 2008). Expanding urban areas in tropical regions have not escaped the deterioration of air quality; on the contrary, often complex topography and highly variable climate conditions emphasize this challenge. In the last five years, the city of Medellín (see Fig. 1a), Colombia, has experienced the onset of critical environmental episodes characterized by poor air quality and, in particular, sudden peaks of PM_{2.5} concentration. The increase in the number of emission sources, especially mobile sources (see Fig. 1b), led to a long-term increase in PM_{2.5} before 2016, exacerbated by critical episodes associated with lower-troposphere stability (Herrera-Mejía and Hoyos 2019). The combination of growing emissions, complex topography, and unfavorable climate and meteorological conditions for the vertical dispersion of pollutants caused PM_{2.5} concentrations to peak during

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FIG. 1. (a) Geographical context of the Aburrá Valley, located in Colombia in the Department of Antioquia. The map shows, in colors from blue to brown, the main topographic features of the region and the distribution of the air quality monitoring stations, the microwave radiometer, and the weather radar. In this study, we use the following monitoring stations: eight in situ PM2.5 and eight PM10 sites. The MWR, a weather station, and a pyranometer are installed at the SIATA main operations center (cyan star). The map also shows the location of a weather station at the western hill (gray circle), the radar wind profiler (maroon circle), and the traffic detection site (orange circle). (b) Yearly evolution of the motorization rate in the Aburrá Valley. The motorization rate corresponds to the number of vehicles per 1000 people. (c) Monthly evolution of the average PM2.5 concentration in the Aburrá Valley. The spatial average is computed using eight PM2.5 stations. The period of the PM2.5 records is from January 2014 to March 2018. The shading corresponds to the mean value ± 1 standard deviation, representing the variability.

March 2016 (see Fig. 1c), with hourly particulate matter concentrations reaching $180 \mu\text{g m}^{-3}$, maximum daily concentrations as high as $118 \mu\text{g m}^{-3}$, and 22 days of unhealthy air quality conditions according to the U.S. Environmental Protection Agency (EPA) standards.

Several studies indicate that exposure to high concentrations of particulate matter, or exposure over extended periods, can contribute to, or even cause, respiratory, cardiovascular, and cognitive diseases (Anderson et al. 2012; Brunekreef and Holgate 2002; Lelieveld et al. 2015; Newby et al. 2015; Münzel et al. 2017; Zhang et al. 2018). Lelieveld et al. (2015) suggested that during 2010, approximately 3.3 million premature deaths worldwide were attributable to high concentrations of PM_{2.5}, and according to model projections this number could double by 2050. Recent evidence suggests that, overall, atmospheric pollution was likely responsible for an estimated 6.5 million premature deaths during 2015 (Landrigan et al. 2018).

In addition to anthropogenic emissions, climate and meteorological conditions modulate air pollution locally: stagnant atmospheres and thin planetary boundary layers contribute to the onset of critical air pollution episodes by controlling whether pollutants are dispersed in the atmosphere or interact among themselves and

with water vapor and radiation to form secondary pollutants (Lazaridis 2011; Elminir 2005). Vertical pollutant dispersion during convective conditions, horizontal advection removal, along-valley and uphill removal, and precipitation scavenging are the main meteorological processes that act to remove gaseous pollutants and suspended particles from the near-surface atmosphere. In the case of particulate matter, wet deposition is arguably one of the most important removal routes, including rainout or in-cloud scavenging and washout or below-cloud scavenging (BCS) (Grömping et al. 1997; Lazaridis 2011). In-cloud scavenging corresponds to the incorporation of pollutants within cloud droplets by condensation during the nucleation phase and incorporation of gases surrounding the droplets by aqueous-phase reactions (Chatterjee et al. 2010; Seinfeld and Pandis 2006; Feng 2007). Collisions and coalescence among particulate matter and rain droplets during the fall, or through turbulent and Brownian diffusion, lead to BCS (Duhanyan and Roustan 2011; Feng 2007; Zikova and Zdimal 2016). Understanding the wet removal processes implies considering multiple spatial scales, from the microscale (10^{-6} m) to the macroscale (10^6 m), and temporal scales from minutes to the inter-annual scale, making wet deposition one of the most

complex atmospheric processes (Seinfeld and Pandis 2006; Pruppacher and Klett 2012).

Most studies on wet deposition have mainly focused on investigating the washout effects by raindrops using a microphysical framework through the analysis of the raindrop size distribution, the terminal velocity of raindrops, and the BCS coefficient for particulate pollutants (Chate and Pranesha 2004; Chate et al. 2011; Bae et al. 2012; Andronache 2003; Wang et al. 2010). These studies evaluate the effectiveness of the washout of solid particles with a specific size under particular rainfall conditions (Olszowski 2016; Zikova and Zdimal 2016; Kyrö et al. 2009). Typically, the wet deposition theory considers different types of hydrometeors, including rain, haze, and snow, in a wide range of diameters: ultrafine, fine, and coarse particles (PM_{10}). The washout efficiency is usually evaluated through the scavenging coefficient on a daily time scale. However, the scavenging coefficient is not easy to estimate due to the complex set of microphysical parameters required; therefore, an approach linking the air quality to observable meteorological parameters such as rainfall is required.

Recent studies have evaluated the washout effect in particulate matter concentrations following an observational approach. Blanco-Alegre et al. (2018) used in situ high-temporal-resolution rainfall and aerosol measurements to estimate scavenging coefficients, finding aerosol size- and raindrop size-dependent scavenging after precipitation. Guo et al. (2016) used data from two cities in China (Guangzhou and Xi'an) to assess the effect of rainfall on the aerosol concentration, finding a minimum cumulative precipitation threshold, different for both cities, for which the air quality improves after a rainfall event. Feng and Wang (2012) computed the precipitation-driven scavenging rate as a measure of the difference in the particle concentration before and after precipitation; their results suggest that, in some cases, factors such as local emissions or atmospheric diffusion conditions predominate over the wet scavenging caused by raindrops, counteracting the washout effect, thus resulting in a higher fine particle concentration after a precipitation event. Feng and Wang (2012) stated that low amounts of precipitation had very little influence on the concentrations of all kinds of particles. Guo et al. (2014) also found that artificial rain interventions may worsen air quality under some scenarios.

Vertical dispersion and rainfall-triggered aerosol removal are especially important in narrow valleys such as the Aburrá Valley, where the complex topography tends to limit the horizontal advection of pollution since the magnitude of surface winds is

usually very weak. Thus, atmospheric stability conditions, the evolution of the atmospheric boundary layer (ABL), and, in particular, the development of a deep convective layer are considered determining factors in pollutant concentrations in the Aburrá Valley, as they modulate the temporal variability of the vertical dispersion efficiency (Herrera-Mejía and Hoyos 2019). A stable atmosphere inhibits atmospheric vertical exchange and favors pollutant accumulation, while unstable convective environments promote pollutant dispersion and mixing (Whiteman 2000). According to Herrera-Mejía and Hoyos (2019), the atmosphere becomes unstable in the Aburrá Valley between 1000 and 1700 LT, favoring thermal convection and mixing, triggering the vertical dispersion of pollutants. On the other hand, stable conditions predominate between 1900 and 0900 LT.

Wet deposition is another potential mechanism for removing pollutants from the near-surface Aburrá Valley's atmosphere that needs to be assessed. As highlighted by Feng and Wang (2012), the net effect of rainfall on the aerosol concentration is not always obvious, as there might be alterations in the atmospheric conditions that counteract the BCS induced by rainfall. The focus of the present study is to estimate the net effect of precipitation on the pollutant concentration using in situ observations, weather-radar-derived precipitation, and atmospheric thermodynamic profiles. The general hypothesis is that the net effect depends not only on the washout efficiency but also on the rainfall-induced stabilization of the lower troposphere. In that sense, and given that the atmospheric stability in the tropics has a strong diurnal cycle induced by the varying sign of the surface energy balance rather than a marked seasonality, it is necessary to examine the net effect on air quality in a subdaily time scale. The proposed approach follows a nonparametric, conditional analysis of hourly records of $PM_{2.5}$ and PM_{10} , given different precipitation scenarios. Section 2 includes the description of the region of study, the main datasets used, the definition of the thermodynamic indices used to evaluate the lower-troposphere stability, and the conditional analysis methodology based on the estimation of an overlapping coefficient among conditional probability density functions (PDFs). Section 3 presents the main results of the methodology for the pollutant concentration during wet and dry conditions in the Aburrá Valley. Finally, section 4 presents the most important conclusions of the study.

2. Data and method

The observational and conditional assessment of the net effect of precipitation on the pollutant concentration

presented in this work relies on in situ air quality measurements from the Aburrá Valley network, rainfall from a weather radar quantitative precipitation estimation (QPE) technique, and thermodynamic indices derived from microwave radiometer (MWR) thermodynamic profiles. We also use information from a radiosonde intensive observation period (IOP) to provide validation for the long-term results obtained using the MWR retrieval, information from automatic weather stations, shortwave radiation from a Kipp and Zonen SMP11 pyranometer (measures the hemispherical solar radiation integrated over the wavelength range from 285 to 2800 nm), high-frequency 3D winds from a Campbell Scientific, Inc., CSAT3 instrument, and traffic information (number of vehicles crossing a downtown detection site per hour) derived from an automatic vehicular camera system provided by the city of Medellín's transit authority. Vehicular counts are available for a year between July 2015 and June 2016. The Medellín metropolitan area includes 10 municipalities settled within the Aburrá Valley, a complex narrow valley with steep hills, located in northern South America, in the central branch of the Andean mountain range ($6^{\circ}15'06''\text{N}$, $75^{\circ}33'48''\text{W}$). The Aburrá Valley is home to approximately three and a half million people living in an area of 1152 km^2 ; the valley is 64 km long, and the widest cross section, from ridgeline to ridgeline, is approximately 18.2 km wide. The height difference between the top and the bottom of the valley is approximately 1.4 km.

a. Data sources

1) AIR QUALITY DATA

Particulate matter and gaseous concentrations in the Aburrá Valley are routinely monitored by the local early warning system [Sistema de Alerta Temprana (SIATA), www.siata.gov.co], a science and technology project established by the local environmental authority [Area Metropolitana del Valle de Aburrá (AMVA)]. In this work, we consider pollutant records longer than three years to guarantee statistically significant results. Figure 1a shows the spatial distribution of the air quality monitoring network instruments used in this research, and Table 1 summarizes the period of the data record used in each case. In this study, we use eight in situ $\text{PM}_{2.5}$ monitoring stations and eight PM_{10} stations.

2) PRECIPITATION DATA

In the absence of good-quality in situ rainfall records in all air quality monitoring stations, a radar-based QPE provides a good alternative to obtain robust precipitation information. The precipitation estimates are based on a technique described in Sepúlveda (2015) and

TABLE 1. Air quality monitoring stations (and data availability).

Station	Type	Initial date	End date
1	$\text{PM}_{2.5}$	1 Jan 2013	10 Sep 2017
	PM_{10}	1 Jan 2013	10 Sep 2017
2	PM_{10}	1 Jan 2013	10 Sep 2017
3	PM_{10}	1 Jan 2013	10 Sep 2017
4	$\text{PM}_{2.5}$	1 Jan 2013	10 Sep 2017
5	$\text{PM}_{2.5}$	1 Jan 2013	10 Sep 2017
	PM_{10}	9 Sep 2015	10 Sep 2017
6	PM_{10}	1 Jan 2013	10 Sep 2017
7	$\text{PM}_{2.5}$	1 Jan 2013	10 Sep 2017
8	$\text{PM}_{2.5}$	16 Sep 2015	10 Sep 2017
9	PM_{10}	1 Jan 2013	10 Sep 2017
10	$\text{PM}_{2.5}$	1 Jan 2013	10 Sep 2017
	PM_{10}	1 Jan 2013	10 Sep 2017
11	$\text{PM}_{2.5}$	4 Feb 2014	10 Sep 2017
12	$\text{PM}_{2.5}$	1 Jan 2013	10 Sep 2017
	PM_{10}	1 Jan 2013	10 Sep 2017

Sepúlveda and Hoyos (2017) using radar reflectivity fields, a disdrometer, and rain gauge information. The QPE technique allows the estimation of precipitation maps over the valley and the locations of interest using retrievals from the Aburrá Valley weather radar, a 350-kW C-band polarimetric and Doppler radar manufactured by Enterprise Electronics Corporation operated by SIATA. The radar scanning strategy allows precipitation information to be obtained every five minutes with a spatial resolution of approximately 128 m, from a 1° tilt plan position indicator sweep. The uncertainty associated with the QPE is relatively low within a 120-km radius from the installation site (less than 10%), and the correlation between hourly in situ rain gauge records and the estimates from the QPE technique is higher than 0.7 (Hoyos et al. 2019). This research uses precipitation products in the period from January 2012 to September 2017. We also conduct the analysis of the net effect of rainfall on the pollutant concentration for one representative air quality monitoring station using radar reflectivity data (as a proxy for precipitation) and in situ rainfall records from a tipping-bucket gauge to ensure that the results are not biased by the QPE technique, adding robustness to the overall assessment.

3) ATMOSPHERIC PROFILES, THERMODYNAMIC STABILITY INDICES, AND ABL HEIGHT

Thermodynamic stability indices are estimated from temperature and humidity profiles measured by the MWR, Radiometrics model MP-3000A, operated by SIATA. The MWR is located at the top of the SIATA main operations center on the valley floor (see Fig. 1), approximately 60 m from the surface, and provides vertical profiles of the thermodynamic state of the atmosphere with variable spatial resolution: 50 m from the

surface to 500 m, 100 m up to 2 km, and 250 m up to 10 km. Thermodynamic profiles are available with a 2-min time resolution, and the data record spans from January 2013 to September 2017. Specifically, the change of potential temperature in the vertical direction at different lower-troposphere levels ($\Delta\Theta_z$), the convective inhibition energy (CINE), and the lifted index (LI) are considered to be proxies of atmospheric stability. The use of $\Delta\Theta_z$ is based on the definition of atmospheric stability using the potential temperature Θ as a conservative variable in adiabatic processes; for unsaturated moist air, $d\Theta/dz > 0$ in stable conditions, $d\Theta/dz = 0$ under neutral conditions, and $d\Theta/dz < 0$ under unstable atmospheres (Peppler 1988; Curry and Webster 1999). The $\Delta\Theta_z$, where z corresponds to the height, in meters, above the surface, is computed as the difference between Θ at height z and Θ at height $z - \Delta z$, $\Theta(z) - \Theta(z - \Delta z)$. We use the temperature difference as a proxy for the average vertical gradient of Θ within each layer. We consider $\Delta z = 200$ -m-thick layers, with the only exception being for $\Delta\Theta_{200}$, computed as $\Theta_{200} - \Theta_{50}$ to avoid the effects of the surface layer.

The CINE indicates the amount of energy that inhibits the updraft of air parcels, and it is an indirect measure of the lower-troposphere stability: as the atmosphere becomes stable, the CINE becomes more negative. Conversely, unstable atmospheres correspond to CINE values close to zero. The CINE is estimated as the integral of the difference between the temperature of the parcel and the ambient temperature from surface to the LFC (if present, otherwise CINE = 0), as follows:

$$\text{CINE} = \int_{\text{SFC}}^{\text{LFC}} g \frac{T'_v - T_v}{T_v} dz,$$

where LFC is the level of free convection, SFC is the surface layer, T_v is the virtual temperature of the environment, and T'_v is the virtual temperature of the parcel (Peppler 1988). The LFC, by definition, corresponds to the height at which an air parcel becomes warmer (and therefore less dense) than the surrounding atmosphere under adiabatic ascent. To estimate the LFC, we follow the *parcel theory* based on the atmospheric profile retrieved using the MWR: (i) From the MWR surface air and dewpoint temperatures, we calculate the lifting condensation level (LCL) as the intersection between the mixing ratio isopleth and the dry adiabatic ascent, finding the level where the rising parcel saturates; (ii) from the LCL, we continue with a moist adiabatic ascent until the top of the atmosphere (in this case 10 km); (iii) then, we calculate the difference (T_{diff}) between the temperature of the parcel and the ambient temperature

profile from the MWR. The LFC is determined as the first level where $T_{\text{diff}} \geq 0$ and the height $H > \text{LCL}$.

The LI is also a proxy of atmospheric stability: as the atmosphere becomes more unstable, the index tends to be increasingly negative. The LI is calculated as

$$\text{LI} = T_{500} - T_{500}^{*},$$

where T_{500} is the environment temperature at 500 hPa and T_{500}^{*} is the temperature of an air parcel lifted adiabatically from 50 hPa above the surface to 500 hPa (Peppler 1988).

A 30-day radiosonde IOP was conducted during March 2018, some days launching three and other days eight radiosondes from the SIATA main operations center, resulting in a total of 114 atmospheric soundings (some radiosonde flights failed, mostly due to communications issues). Radiosonde retrievals are useful to validate the long-term results obtained using the MWR retrievals. Radiosondes provide direct measurements of atmospheric profiles and, hence, constitute an important reference for the estimates in this study. However, the radiosonde measurements are also susceptible to artifacts associated with the Lagrangian nature of the platform: radiosonde profiles do not correspond to the same atmospheric column, and in a narrow valley, the differences between the profiles over the base of the valley and over the hills could potentially be large, introducing uncertainty into the comparisons. In our case (figure not shown here), most of the radiosonde flights below 4500 m are within the area sensed by the MWR in the same altitude range; however, there is a westward bias toward the hills in the radiosonde that could potentially introduce differences in the measurements.

Regarding the ABL height, and despite its importance, there is a lack of a robust tool to detect mixing heights, particularly in complex terrains, where there could be multiple definitions, and hence a multi-layered structure, of the ABL (Seibert et al. 2000; De Wekker and Kossmann 2015; Lehner and Rotach 2018). Various definitions and methods could result in a wide range of estimates (Eresmaa et al. 2012; Lotteraner and Piringer 2016). Among the most widely used methods are the gradient method using radiosondes (Seidel et al. 2010; Lee et al. 2014), the minimum gradient method, maximum variance using ceilometer profiles (Hayden et al. 1997; Stachlewska et al. 2012), and the bulk Richardson number method (Stull 1988; Chandra et al. 2014; Zhang et al. 2014). In this study we use the ABL height estimates obtained by Herrera-Mejía and Hoyos (2019) using a Richardson number-based multisensor methodology. Herrera-Mejía and Hoyos (2019)

FIG. 2. Scatterplots of (a) the daily precipitation vs 24-h-average PM_{2.5} and (b) the daily precipitation vs the 24-h change in the PM_{2.5} concentration (Δ PM_{2.5}). The Δ PM_{2.5} is calculated as the 24-h concentration during a rainy day minus the previous day concentration.

implemented different techniques to estimate the ABL height using ceilometer backscattering profiles and a combination of a radar wind profiler and MWR retrievals (multisensor), concluding that the Richardson multisensor technique is more robust, performing better under stable and unstable conditions than the other methods, and capturing properly the different time scales of ABL variability. The temporal resolution of the Richardson ABL height estimates is 30 min, and the information is available since January 2015.

b. Diurnal cycle conditional analysis

One of the most prominent challenges in the assessment of the net effect of precipitation scavenging atmospheric pollutants in a tropical environment is the determination of the appropriate time scale for the analysis. We argue that using the daily cumulative precipitation for the study could mask the actual net effect of rainfall in the aerosol concentration. The 24-h time scale is artificial and does not distinguish well among the physical processes that lead to precipitation. In the Aburrá Valley, the diurnal cycle of precipitation is bimodal (Poveda et al. 2005), with intense and short-lived (30–50 min) convective rainfall events in the afternoon (from 1400 to 1700 LT) and low-intensity, long-lived stratiform precipitation during the night (from 0000 to 0400 LT). The hypothesis behind the work presented in this study is threefold: (i) The net effect of rainfall on the pollutant concentration depends not only on the efficiency of the washout effect but also on the role of rainfall in stabilizing the atmosphere, thus leading to near-surface pollutant accumulation; (ii) the effect of precipitation in modulating

atmospheric stability has a strong diurnal cycle; and (iii) the time scale for the analysis must allow us to consider intraday variability. Here, we use hourly-resolution datasets, separating the entire record into different categories (48 in total): for each hour of the day and for dry (precipitation $< 2 \text{ mm h}^{-1}$) and wet (precipitation $\geq 2 \text{ mm h}^{-1}$) conditions (24×2). We replicated the analysis using different precipitation thresholds to separate between dry and wet cases (from 0 to 3 mm) and the results are robust (not shown). The analysis for wet cases considers pollutant concentrations one hour after the precipitation event to assess the immediate precipitation net effect.

The method includes a conditional analysis, contrasting the estimated PDFs of various atmospheric pollutants for the different hours of the day and among dry and wet conditions, similar to the methodology described in Agudelo et al. (2011). This framework allows us to examine, in a probabilistic sense, the diurnal cycle of the net effect of precipitation on pollutant concentrations. The separation of the PDFs for different atmospheric pollutants under wet and dry conditions is a measure of the net effect of precipitation on near-surface pollution and, furthermore, a quantitative estimation of the relative net removal efficiency (pollutant removal relative to dry conditions) for the different pollutants and a proxy for discriminating the potential of pollutant concentration based on hourly rainfall data or weather forecasts.

The same procedure is also followed for the thermodynamic indices to analyze the atmospheric stability during dry conditions and after precipitation events for every hour of the day. Additionally, the net effect of

FIG. 3. (a)–(d) Radar reflectivity fields as a proxy for the evolution of the precipitation event on 24 Aug 2016, from 1300 to 1730 LT. (e) Temporal variability of fine particulate matter (black line), rainfall (blue line), and cumulative rainfall (dashed line). (f)–(j) As in (a)–(e), but for the precipitation event on 22–23 Mar 2017.

FIG. 4. Cases with rainfall in the afternoon preceding an increase in the concentration of PM2.5 showing (a),(c) the temporal variability of PM2.5 (black line), rainfall (blue line), and cumulative rainfall (dashed line) and (b),(d) the cumulative precipitation obtained using the QPE technique for the same period as in (a) and (c). (e)–(h) As in (a)–(d), but for cases with rainfall in the evening associated with BCS.

FIG. 5. The (a),(b) PM_{2.5} concentration PDFs for wet (green line) and dry (orange line) conditions at monitoring station 4 and (c),(d) PM₁₀ at monitoring station 5 in Fig. 1 at (left) 0200 and (right) 1400 LT. The QPE-derived precipitation time series is used to discriminate between wet and dry conditions.

precipitation on the pollutant concentration is studied taking into account the role of precipitation intensity using the same conditional approach. For this assessment, the maximum precipitation intensity is computed for events with a cumulative rainfall greater than 5 mm in a 60-min period; the hourly pollutant records are then reclassified in two sets for precipitation intensities below and above the median.

c. Overlapping coefficient

Once all the pollutant PDFs are estimated for each hour of the day and for wet and dry conditions, we propose using an index to summarize the results by

estimating the relative separation or agreement among the PDFs (Inman and Bradley 1989). The index is based on the overlapping coefficient (OC), which corresponds to the area of intersection among the PDFs. Let $f[C(t)] = P[C(t)|WC]$ and $g[C(t)] = P[C(t)|DC]$ be the pollutant concentration $C(t)$ PDFs for wet (WC) and dry (DC) conditions, respectively; the OC in the discrete case is calculated as follows:

$$OC = \sum_x \min\{f[C(t)], g[C(t)]\},$$

with $0 \leq OC \leq 1$. The index corresponds to the signed OC complement (SOCC) defined as follows:

$$SOCC = \begin{cases} OC - 1 & \text{median}\{f[C(t)]\} \leq \text{median}\{g[C(t)]\} \\ 1 - OC & \text{median}\{f[C(t)]\} > \text{median}\{g[C(t)]\} \end{cases},$$

with $-1 \leq SOCC \leq 1$. According to the definition, a SOCC index close to 0 indicates that both distributions share a large amount of area, suggesting, in consequence, that precipitation does not play a significant role

in modulating the net pollutant concentration. On the other hand, a SOCC index closer to ± 1 indicates that the PDFs are more separated, suggesting that precipitation is an important discriminating factor altering the

pollutant concentration. Negative values of the SOCC index correspond to a decrease in the pollutant concentrations associated with rainfall, and positive values are associated with an increase in the pollutant concentrations.

The nonparametric Wilcoxon–Mann–Whitney test (Mann and Whitney 1947; Depuy et al. 2014) is used to assess whether any the two samples $X_1, X_2, \dots, X_n$ and $Y_1, Y_2, \dots, Y_m$ likely originate from the same distribution. The Wilcoxon–Mann–Whitney test does not assume normality or equal variance as does the t test, and it uses the ranks of the data rather than their raw values. In the Wilcoxon–Mann–Whitney test, the null hypothesis is that the distributions of the two concentration populations $f[C(t)]$ and $g[C(t)]$ are identical. The statistic for the Wilcoxon–Mann–Whitney test is computed as

$$U_n = nm + \frac{n(n+1)}{2} - R_n \quad \text{and}$$

$$U_m = nm + \frac{m(m+1)}{2} - R_m,$$

where n is the sample size of $[C(t)|WC]$ (sample 1), m is the sample size of $[C(t)|DC]$ (sample 2), and R_n and R_m are the sum of the ranks in samples 1 and 2, respectively. The R_n and R_m are obtained after ranking all of the $n + m$ observations in a single sample $[C(t)|WC] \cup [P(t)|DC] = C(t)$. The normalized test statistic Z , which is approximately Gaussian for large samples, is defined as

$$Z = \frac{U - m_U}{\sigma_U},$$

where $m_U = nm/2$ and $\sigma_U = [nm(n + m + 1)/12]^{1/2}$. If $|Z|$ is greater than a predetermined cutoff value, then the null hypothesis can be rejected. We use 95% significance as a threshold to reject the null hypothesis.

3. Results

a. Preliminary assessment

An assessment of the the relationship between the total daily precipitation and the 24-h-average $PM_{2.5}$ in the Aburrá Valley shows that high concentrations (over $30 \mu\text{g m}^{-3}$) are more likely to occur when the total daily precipitation is less than 25 mm (Fig. 2a); however, there is not a clear relationship indicating that the 24-h-average $PM_{2.5}$ concentration variability is far from being determined by antecedent rainfall conditions. In addition, the relationship between rainfall and $PM_{2.5}$ is considerably different to the results from theoretical scavenging studies (Wang et al. 2014). In other words, the observations indicate that high $PM_{2.5}$ concentrations are not likely in days with high

FIG. 6. Diurnal cycle of the SOCC index for (a) $PM_{2.5}$ at monitoring station 4 and (b) PM_{10} at monitoring station 5. Similar to Fig. 5, the QPE-derived precipitation time series is used to discriminate between wet and dry conditions. Negative values indicate net precipitation-induced aerosol removal, and positive values suggest that precipitation leads to pollution accumulation. The circles summarize the results of the Wilcoxon–Mann–Whitney test. The null hypothesis states that the PDFs of pollutants under wet and dry conditions are identical. Filled green circles correspond to cases in which the null can be rejected; conversely, open circles correspond to hours in which the null hypothesis cannot be rejected. (c) Diurnal cycle of the potential temperature Θ at 50 m (black line) and $\Delta\Theta_{200}$ (blue line). The yellow area represents the period between sunrise and sunset.

cumulative precipitation, but low precipitation is not necessarily related to a high $PM_{2.5}$ concentration. However, the practical utility of this apparent relationship is limited, especially if we consider forecasting $PM_{2.5}$ concentrations using rainfall as a predictor variable. The critical issue is whether $PM_{2.5}$ concentrations can be predicted based on cumulative precipitation information or whether other processes are considerably more important in modulating the concentration of pollutants in the lower troposphere. Figure 2b suggests that there is no clear reduction of $PM_{2.5}$ (negative $\Delta PM_{2.5}$) even in cases of large cumulative precipitation. On the contrary, there is a considerable number of instances where $\Delta PM_{2.5}$ is positive, emphasizing the fact that the overall precipitation–aerosol concentration relationship is complex.

Recurring contradictory evidence of the net effect of rainfall on the aerosol concentration is observed after precipitation episodes in the region. As an example, a precipitation event took place on 24 August 2016, from

FIG. 7. Conditional analysis for monitoring station 4 in Fig. 1 using (left) radar reflectivity data and (right) in situ rainfall records, showing the PDFs for wet and dry conditions at (a),(d) 0200 and (b),(e) 1400 LT, as well as (c),(f) the diurnal evolution of the corresponding PM_{2.5} SOCC index in each case.

1300 to 1730 LT, over the Medellín metropolitan area (see Figs. 3a–d). Subsequently, after the rainfall event, PM_{2.5} concentrations considerably increased (Fig. 3e), doubling from an average of approximately $30 \mu\text{g m}^{-3}$

in the morning hours to approximately $60 \mu\text{g m}^{-3}$ after 1500 LT. A contrasting case occurred as a result of the nighttime precipitation on 22 March 2017, after which the PM_{2.5} concentrations decreased considerably

FIG. 8. Summary of the SOCC index for all (a) PM_{2.5} and (b) PM₁₀ stations. White squares correspond to hours of the day for which there are not sufficient precipitation cases to estimate a statistically significant SOCC index; X marks correspond to cases in which the null hypothesis, indicating that the distributions of pollutants for wet and dry conditions are identical, cannot be rejected. The numbers on the left correspond to stations in Fig. 1.

FIG. 9. (a) Average number of vehicles per hour crossing a detection site in the city of Medellín, on a downtown street, conditioned to hours with dry events only (orange line), as well as conditioned to hours during (blue line) and after (green line) rainfall events. (b) Diurnal cycle of the number of vehicles per hour for dry weather and the composite diurnal cycle for all cases with rainfall between 1400 and 1500 LT. Also shown are average (c) absolute value and (d) anomaly (composite analysis) of the number of vehicles associated with rainfall events for each hour, respectively. The abscissa axis in (b) and the ordinate axis in (c) and (d) correspond, in each case, to the time of the precipitation events used to generate the composites (around the time of the events, as reference).

(see Figs. 3f–j). The air quality monitoring network registered $\text{PM}_{2.5}$ concentrations above $60 \mu\text{g m}^{-3}$ in all ground stations prior to the precipitation event; after the event, the $\text{PM}_{2.5}$ concentrations declined to less than $10 \mu\text{g m}^{-3}$. Figure 4 presents four additional examples showing two cases with rainfall in the afternoon preceding an increase in the concentration of $\text{PM}_{2.5}$ (Figs. 4a–d), and two cases where rainfall in the evening is associated with efficient BCS.

b. Conditional analysis

1) PM ASSESSMENT

This contrasting behavior is not episodic. The $\text{PM}_{2.5}$ concentration PDFs for wet (blue line) and dry (orange

line) conditions at 0200 LT, using the QPE-derived precipitation for the subset discrimination, show that, at this time of the day, the likelihood of having lower concentrations is considerably larger after precipitation takes place, as a result of aerosol washout (Fig. 5a). In contrast, at 1400 LT, the likelihood of having higher $\text{PM}_{2.5}$ concentrations increases immediately after rainfall events compared to the likelihood during dry days (Fig. 5b). Clearly, the net effect of precipitation on the aerosol concentration is modulated by other factors in addition to the rainfall-induced washout mechanism. The precipitation net effects on the PM_{10} concentration are congruent with the findings for $\text{PM}_{2.5}$ (Figs. 5c,d).

The above results suggest that there is a diurnal modulation of the net effect of precipitation on the

aerosol concentration. Figure 6 expands the assessment to the entire diurnal cycle and summarizes the previous distributions using the SOCC index, with negative values indicating a net precipitation-induced aerosol removal and positive ones suggesting that precipitation leads to pollution accumulation. The overall results for $\text{PM}_{2.5}$ and PM_{10} indicate that the net effect of precipitation during the night is the reduction in the particulate matter concentration, most likely associated with washout aerosol removal (Figs. 6a,b). During the day, between 1000 and 1700 LT, the indirect net effect of precipitation is the opposite, leading to an increase in the particulate matter concentration. The most striking result is that the SOCC index changes sign during the day following the inverse sign of the near-surface stability, with negative values during the night and before midmorning and positive values during the day (Fig. 6c). Since the BCS does not depend on whether the precipitation is nocturnal or occurs during the day, other processes must be indirectly offsetting the BCS effect. Figure 7 presents the same conditional analysis as in Figs. 5 and 6 but using radar reflectivity data and in situ rainfall records, showing the PDFs for wet and dry conditions at 0200 and 1400 LT, as well as the diurnal evolution of the corresponding $\text{PM}_{2.5}$ SOCC index. The evidence confirms the results regarding the diurnal variability of the net effect of precipitation on the aerosol concentration, suggesting that the results obtained using the radar QPE products are robust and do not strongly depend on the precipitation product used to discriminate between wet and dry days, after all, such discrimination is straightforward.

The comparison between the net effect of rainfall in the concentration of $\text{PM}_{2.5}$ and PM_{10} hints to a relevant difference regarding the particle size. While the overall results for $\text{PM}_{2.5}$ and PM_{10} are similar, there is an important difference in the 1000–1700 LT period. During this time, for PM_{10} , the period with a positive SOCC index is shorter (1000–1500 LT) than for $\text{PM}_{2.5}$ (1000–1700 LT); more important, the null hypothesis cannot be rejected, except at 1400 and 1500 LT, and the differences among the PM_{10} PDFs for wet and dry cases are not significant. This finding could be related to the aerosol diameter, as the removal efficiency of coarser particles in the 1–10- μm range is higher than that for finer particles.

The results of the assessment of the particle concentration registered at all monitoring sites are coherent with the previously described findings. Figures 8a and 8b summarize the SOCC index for $\text{PM}_{2.5}$ and PM_{10} , respectively, for all PM monitoring stations in Fig. 1, showing a coherent change in the SOCC sign during the diurnal cycle associated with net particulate matter

FIG. 10. SOCC index for different cumulative precipitation considering the time period from 0000 to 0600 LT, for $\text{PM}_{2.5}$ (green line) and PM_{10} (blue line) recorded at monitoring station 10 (see Fig. 1).

removal during nighttime and the net aerosol concentration increase during the day, after wet conditions. Similarly, the precipitation effects on the PM_{10} concentration are weaker, and the null hypothesis cannot be rejected, in the period between midmorning and before the sunset, potentially due to particle size effects.

The analysis in the previous paragraph is based on an implicit assumption: there is no increase in vehicular emissions during or after a precipitation event during daytime. Figure 9a presents the average number of vehicles per hour crossing a detection site in the city of Medellín, during dry hours only, and hours during and after rainfall events. The figure shows, as expected, a marked diurnal cycle of traffic for all three cases. The traffic variability could be perceived qualitatively as a proxy for emissions. In addition, it is clear that for all hours, the number of vehicles crossing the detection sites during or after rainfall events is less or equal than that for dry conditions. Since all hours of the day are analyzed independently in Fig. 9a, it is necessary to evaluate whether the number of vehicles detected per hour is less due to a significant slowing-down effect of the traffic due to poor meteorological conditions or whether there is an actual reduction of cars on the road associated with people avoiding driving under wet conditions. We argue that the reduction in the number of trips is due to the reduction in the number of non-commuting trips, provided that it is highly unlikely for a person to avoid commuting trips due to rainfall. In other words, people for whom it is not mandatory to drive for commuting purposes are discouraged from using their vehicles during and after rainfall events.

Figure 9b shows a comparison between the diurnal cycle of the number of vehicles per hour for dry weather and the composite diurnal cycle for the cases

FIG. 11. Scatterplots of (a),(b) potential temperature and (c),(d) relative humidity retrieved using MWR and measured using radiosondes during an IOP during March 2018 for measurements at (left) 50 and (right) 200 m above the surface; 114 different radiosonde flights were used in the construction of the plots.

with rainfall between 1400 and 1500 LT, using the same precipitation events as those used to estimate the SOCC index in Fig. 6. The figure presents evidence of a 4-h reduction in the number of vehicles during and after the rainfall event. The evaluation of the reduction of the number of vehicles during precipitation events for each hour of the day is presented in Figs. 9c and 9d, showing in colors, and for every hour, the absolute value and the anomaly of the number of vehicles associated with rainfall events. The figure shows that during working-day hours when the traffic is considerable, there is always a reduction in the number of vehicles on the road and no generalized extension of the rush hours into the evening, which also indicates that there is reduction of the number of trips, likely noncommuting trips, suggesting an actual reduction in

emissions during the day. Based on the vehicular evidence, the increase in particulate matter concentration seen in Fig. 6 is not due to an increase in the number of vehicles on the road; hence, atmospheric processes must be offsetting the BCS.

The SOCC index is also used to analyze the effects of cumulative precipitation on the aerosol concentration (Fig. 10). The figure shows, for the period between 0000 and 0600 LT, changes in the SOCC index for different values of cumulative precipitation. While the cumulative precipitation increases, the SOCC index becomes more negative; that is, the differences between the PDFs for wet and dry conditions are greater, indicating that the net aerosol removal is larger. Thus, as the cumulative precipitation increases, the effects of precipitation become more significant. Aerosol BCS

increases efficiency from 0 to 5 mm cumulative precipitation. After 5 mm, the efficiency does not further increase. This condition is true for both $\text{PM}_{2.5}$ and PM_{10} ; however, the precipitation removal effects for PM_{10} are more significant, showing the dependence of wet deposition on the aerosol size. The removal efficiency quickly stabilizes as the cumulative precipitation reaches 5 mm, implying that most aerosol BCS occurs during the first stages of precipitation. Differences between the PM_{10} and $\text{PM}_{2.5}$ washouts are explained considering the theory of collection efficiency between particles and rain droplets. According to previous studies, the collection efficiency is minimal for particles with an aerodynamic diameter approximately between 50 nm and $2\ \mu\text{m}$ (Qurel et al. 2014; Cherrier et al. 2017); these particles are in the so-called Greenfield gap (Greenfield 1957).

The evidence suggests that the influence of precipitation on the aerosol concentration is strongly dependent on the diurnal cycle of the state of the atmosphere, most likely of the atmospheric stability, with a positive SOCC index corresponding to times during the day in which the lower troposphere is typically unstable within the valley, which is consistent with the findings by Herrera-Meja and Hoyos (2019). The evidence in Fig. 6 shows that around the transition times between stable and unstable conditions (0900–1000 and 1700–1800 LT), the null hypothesis cannot be rejected, indicating that during these periods, the differences in the aerosol concentrations in wet and dry conditions are not statistically significant and that there is no certainty about the effects of precipitation on the aerosol concentrations.

2) STABILITY ASSESSMENT

Thermodynamic indices are used to evaluate the effect of precipitation events on the lower-troposphere stability also following the conditional methodology. Before assessing the overall changes in the lower-troposphere stability during and after rainfall events relative to dry conditions, it is important to evaluate the representativeness of the temperature and moisture profiles retrieved using the MWR, in particular considering the performance of radiometer retrievals in complex terrains (e.g., Massaro et al. 2015). Radiosondes are widely regarded as an accurate method for measuring temperature and humidity profiles in the atmosphere, including the lower troposphere, with applications in boundary layer studies (e.g., Seidel et al. 2010). However, the limited temporal resolution between radiosonde flights does not allow a detailed evaluation of changes in atmospheric stability in different time scales. Despite the potential artifacts due to their Lagrangian

FIG. 12. Correlation, for different levels above the surface, between relative humidity, temperature, potential temperature, virtual potential temperature, and vapor pressure retrieved using the MWR and the same variables measured by radiosondes during the March 2018 IOP; 114 different radiosonde flights were used in the construction of the plots.

nature, radiosonde profiles from the March 2018 IOP (114 soundings) are used to evaluate the validity of the MWR retrievals inside the valley, near the surface, where atmospheric pollution is measured. Figure 11 presents the scatterplots of potential temperature and relative humidity retrieved using the MWR and measured using radiosondes at 50 and 200 m above the surface, showing a good agreement during the entire IOP. In all four cases in Fig. 11, the correlation is larger than 0.93. Figure 12 shows the correlation, at different levels above the surface, between relative humidity, temperature, potential temperature, virtual potential temperature, and vapor pressure retrieved using the MWR and measured by the radiosondes during the IOP. The figure shows correlations that are greater than 0.5 for all variables up to 1750 m above the surface. In particular, the correlation in the case of potential temperature (and virtual potential temperature) is greater than 0.85 up to 1100 m above the surface, which coincides with the average depth of the valley (dashed black horizontal line in Fig. 12). The correlation for relative humidity, which is more difficult to measure, is greater than 0.65 below 1100 m above the surface. The correlation coefficient for $\Delta\theta$ at different layers, estimated from radiosondes and the MWR, is in all cases greater than 0.63. The low correlations above the top of the ridgetop are most likely explained by the Lagrangian nature of the radiosondes, and not due to an artifact of the MWR: once the radiosondes enter the free atmosphere, trade

FIG. 13. (a),(b) CINE and (c),(d) LI PDFs at (left) 0200 and (right) 1400 LT for wet (green line) and dry (orange line) conditions.

winds advect them westward away from the valley, while the MWR temperature and humidity retrievals correspond to the same column above the base of the valley. Even if there were artifacts above 2500 m, the focus in this work is on the lower-troposphere stability, therefore the results suggest MWR retrievals are useful for continuous long-term lower-troposphere stability assessment.

The PDFs of CINE for wet and dry conditions show that, at 0200 LT, the CINE distributions for the wet and dry cases are quite similar (Fig. 13a). In contrast, at 1400 LT, the results suggest that the CINE PDF for wet conditions is considerably different than that for dry conditions, with larger negative values, indicating a lower-troposphere stabilization as a result of the precipitation event occurring during unstable conditions (Fig. 13b). The diurnal precipitation leads to atmospheric stabilization, hence decreasing the occurrence of vertical updrafts. The PDFs of LI at both times suggest a slight change toward a more stable atmosphere immediately after the precipitation event. These results differ from the CINE PDFs for 0200 LT: while CINE PDFs for this period do not show any change, the LI PDF shows a more stable atmospheric column. It is important to consider that while CINE

represents the inhibition of local convection, from bottom to top, the LI corresponds to a midtropospheric index. The observed difference is likely to be associated with the nature of both indices: precipitation does appear to be associated with changes in the midtroposphere stability even if the atmosphere is already stable, most likely due to the release of latent heat, while it does not seem to affect the lower-troposphere stability. In general, atmospheric stabilization restricts the rise of pollutants from the surface and over the top of the valley (where they can be advected away by the trade winds), leading to pollutant accumulation within the valley in association with continued anthropogenic emissions. During the night, and considering that rainfall occurs already in a stable environment, precipitation does not have a significant effect on the atmospheric stability near the surface and, consequently, the CINE PDF does not change between the wet and dry cases.

In addition to CINE and LI, we also use potential temperature gradients in different lower-troposphere layers ($\Delta\theta$) to characterize the atmospheric stability during dry and wet conditions. Overall, the sign and the magnitude of the SOCC index for $\Delta\theta_{200}$, to $\Delta\theta_{1400}$ every 200 m, suggests there is atmospheric

FIG. 14. (a) Evolution of the SOCC index for potential temperature gradients throughout the day, for different layers, including from $\Delta\Theta_{200}$ to $\Delta\Theta_{1400}$, every 200 m, where $\Delta\Theta_z$ is defined as $\Theta(z) - \Theta(z - 200)$. (b), (c) Also shown is the SOCC index (b) for the incoming radiation change at the surface level, from 0600 to 1700 LT, and (c) for the ABL height with time.

stabilization after a rainfall event, regardless of the hour of the day (Fig. 14). The SOCC index, which summarizes the differences in PDF for dry and wet conditions, also suggests that the stabilization is larger in magnitude during the daytime, from 0700 to 1700 LT. Figures 15a–d, show, in colors, and for every hour of the day, the absolute value and the anomaly of $\Delta\Theta_{200}$ and CINE for precipitation events, relative to dry conditions. The figure confirms that the amplitude of the near-surface stabilization is larger during the daytime as a response, mainly, to precipitation events. On the other hand, CINE, which is a column-derived index, shows evidence of stabilization prior, during, and after the rainfall events as a result of cloud

radiation forcing as it will be discussed in the following paragraphs.

The stabilization mechanisms, in a topographically constrained environment, are associated with a combination of (i) the negative radiative cloud forcing associated with nimbus clouds and other precedent cloudiness and (ii) the formation of a convective cold pool due to evaporatively cooled air transported to the surface through convective downdrafts, modifying the thermodynamic properties and moisture structure in the subcloud layer and generating stable stratification (Simpson 1969; Charba 1974; Tompkins 2001). Figure 14b shows the SOCC index for incoming radiation at the surface level, from 0600 to 1700 LT. The data, as

FIG. 15. Composite analysis for the (left) absolute value and (right) the anomaly, for every hour of the day, of (a),(b) $\Delta\Theta_{200}$ and (c),(d) CINE for precipitation events, relative to dry conditions. The abscissa axis in (b) and the ordinate axis in (c) and (d) correspond, in each case, to the time of the precipitation events used to generate the composites (around the time of the events, as reference).

expected, suggest there is an important change in surface radiation associated with the occurrence of precipitation. Figures 16a–d shows a similar analysis as the one presented in Fig. 9 for traffic anomalies, but for radiation anomalies associated with wet conditions. Figure 16a shows the average shortwave radiation per hour measured at the same location as the MWR, conditioned to hours with dry events only, as well as conditioned to hours during and after rainfall events. The figure shows a marked diurnal cycle for all three cases, and a considerable decrease in incoming solar shortwave radiation both during and after precipitation events, reaching up to 600 W m^{-2} anomalies around noon. Associated with the latter magnitude of shortwave radiation anomalies, net radiation negative anomalies during rainfall events are of approximately 400 W m^{-2} , and positive downward anomalies of longwave radiation before the precipitation event of $20\text{--}30 \text{ W m}^{-2}$. Similarly as in Fig. 9, since all hours are analyzed independently, it is important to evaluate

radiation anomalies using composites for different lags to study the time span in which radiation is reduced around a precipitation event. Figure 16b shows a comparison between the radiation diurnal cycle for dry weather and the composite diurnal cycle for the cases with rainfall between 1400 and 1500 LT. The figure presents evidence of a 4-h reduction in the radiation after the rainfall event. Figures 16c and 16d show, in colors, and for every hour of the day, the absolute value and the anomaly of radiation associated with rainfall events. The figure shows that during the daytime, there is always a reduction in the radiation reaching the surface, with a longer effect when rainfall occurs in the morning time, which is not very likely in the region (Poveda et al. 2005). Considering Figs. 14a, 14b, and 16a–d, it is clear that the stabilization effect is due to the negative radiation anomalies together with the generation of a cold pool, both as a result of rainfall events. So far, the evidence for the cold pool is somewhat indirect: even in the absence of solar radiation,

FIG. 16. As in Fig. 9, but for (a)–(d) surface radiation, (e)–(h) ABL height, and (i)–(l) TKE.

all the lower-troposphere levels show positive anomalies of $\Delta\theta$ which implies a lower-troposphere cooling. Figure 17 presents evidence of the cold pool formation, showing the onset of considerable negative surface air temperature anomalies during the night during and after precipitation events (Figs. 17a–c), that suddenly stabilize the lower troposphere. These anomalies, on the other hand, do not appear as a result of cloudiness (see Fig. 17d).

Figures 16e–h and 16i–l show a similar analysis as in Figs. 16a–d, for the ABL and the turbulent kinetic energy, respectively. The figures provide additional evidence of the overall stabilization of the atmosphere, showing a considerable reduction of the ABL height and the TKE under wet conditions. The diurnal cycle of ABL height shows a minimum at sunrise, with values around 400 m, and a maximum around noon, with the ABL reaching 1400 m. Under wet conditions, the magnitude of the negative ABL height anomalies during daytime is between 600 and 800 m. The magnitude of the ABL height anomalies are larger after the event than before, but precedent cloudiness also appears to play a role modulating the ABL height. Figure 14c presents the SOCC index for ABL height, showing a considerable and statistically significant separation under dry and wet conditions. The behavior of TKE is very similar to the observed changes in ABL height as a response to a precipitation event, showing a 50% reduction in the afternoon. On the other hand, one-minute average surface winds do not change magnitude significantly in any hour of the day in association with precipitation events, as suggested by the results shown in Fig. 18. The figure shows, the long-term wind rose at SIATA main operations building located at the base of the valley (MWR site), for the period from 0200 to 0300 LT under dry conditions (Fig. 18a) and after precipitation events (Fig. 18b), and for the period from 1400 to 1500 LT under dry conditions (Fig. 18c) and after precipitation events (Fig. 18d). Figures 18e–h are similar to Figs. 18a–d, for a weather station located over the western hill. In general, the different wind roses for the 0200 and 1400 LT periods show a slight decrease in wind speeds after rainfall events, with the exception of the 0200 LT period at the base of the valley, which shows a slight increase. Figures 18i and 18j show the PDF of wind speed at the base of the valley and over the western hill, respectively, for dry and wet conditions, without conditioning the data by the time of the day. In general, the differences suggest a slight decrease in winds after rainfall events; however, these differences are not very significant in terms of the magnitude, particularly over the western hill,

FIG. 17. (a) Surface air temperature during the evening conditioned to hours characterized by cloud-free atmospheres (orange line), cloudy skies (blue line), and after rainfall events (green line). (b) Surface air temperature during the evening for all cases with rainfall between 0300 and 0400 LT. Average absolute value of temperature associated with (c) rainfall events and (d) cloudy (but dry) conditions for each hour. The abscissa axis in (b) and the ordinate axis in (c) and (d) correspond, in each case, to the time of the precipitation events used to generate the composites (around the time of the events, as reference).

where the winds are weak, regardless of the occurrence of rainfall, with magnitudes typically less than 2 m s^{-1} . In summary, the conditions associated with rainfall events show a considerable stabilization of the atmosphere, a reduction of the ABL height, and a decline in the TKE, especially during the daytime, leading to the observed pollutant accumulation.

3) PRECIPITATION INTENSITY ASSESSMENT

An assessment of the role of precipitation intensity in aerosol removal, using the median intensity as a threshold to separate the full set into high and low precipitation intensity events, suggests that $\text{PM}_{2.5}$ concentration is lower after low-intensity precipitation events than after high-intensity cases (Fig. 19a). Before conclusions are drawn about the higher efficiency of low-intensity events, it is important to consider that, in general, convective systems over the Aburrá Valley occur during the afternoon hours and are characterized by their high intensities and short life cycles. Stratiform systems occur mainly during

the night hours and are characterized by their long life and low/medium intensity. Figure 19b presents the distribution of the precipitation intensity for all wet cases, the median precipitation intensity used as a threshold to separate low-intensity from high-intensity cases, and the intensity distribution for nighttime and daytime precipitation. The figure suggests a different precipitation intensity distribution depending on the period of the day, also indicating that low $\text{PM}_{2.5}$ concentrations do not depend on the intensity as could have been wrongly concluded from Fig. 19a, but, again, on the time of the precipitation. Thus, the apparent role of the precipitation intensity is the result of the varying net effect of precipitation on the aerosol concentration during the course of the day.

4) CLIMATOLOGICAL ASSESSMENT: AN EXAMPLE

In general, precipitation in the valley presents a marked diurnal cycle, with two peaks, one of them at approximately 0100 LT and the other at approximately 1500 LT (Poveda et al. 2005), as indicated by the

FIG. 18. Long-term wind roses (top) at SIATA main operations building located at the base of the valley (MWR site) and (middle) for a weather station located over the western hill (a),(c),(e),(g) under dry conditions and (b),(d),(f),(h) after precipitation events for the period (left) from 0200 to 0300 LT and (right) from 1400 to 1500 LT. Also shown are the PDF of wind speed at (i) the base of the valley and (j) over the western hill for dry and wet conditions for the entire data record.

continuous black line in Fig. 20a. As a result of the divergent effects of nighttime and daytime precipitation on the aerosol concentration, the month-to-month variability of the diurnal cycle of precipitation strongly modulates the air quality, partly determining the likelihood of occurrence of critical air quality episodes. The monthly precipitation over the Aburrá Valley also shows a pronounced annual cycle of precipitation (see Fig. 20b) and a bimodal structure as a result of the seasonality of the intertropical convergence zone, with high precipitation during March–May and September–November. Despite this regularity, the diurnal cycle of precipitation changes considerably from year to year and from month to

month. Figure 20a presents the diurnal cycle of precipitation during March 2016 and 2017 and the long-term mean for the month. It is evident that, in addition to the differences in total precipitation, March 2016 is characterized by afternoon convective precipitation, while nighttime precipitation is negligible. Based on the results of the present study, the precipitation diurnal cycle is one of the main factors determining the March 2016 critical air quality episode shown in Fig. 20d. The peak in the aerosol concentration during the transition season in March 2016 is not observed during October 2016 or during March 2017, the two subsequent transition seasons. During October 2016, the precipitation diurnal cycle exhibits considerable

FIG. 19. (a) PDFs of PM_{2.5} concentration, both in wet conditions, for a high (green line) and low (orange line) precipitation intensity. The median intensity is used as a threshold to separate the full set. (b) Distribution of the precipitation intensity for all wet cases (purple line), with the median precipitation intensity (vertical line) used as a threshold to separate the low- and high-intensity cases, and the intensity distribution for nighttime (blue line) and daytime (coral line) precipitation.

overnight rainfall (Fig. 20c), leading to a net aerosol removal compared to the circumstances during March 2016. Similarly, the March 2017 nighttime precipitation is also higher than that during March 2016,

helping to control, together with the implementation of restrictive emission policies, sudden increases in aerosol concentrations. These results emphasize the need for a better understanding and more skillful

FIG. 20. Diurnal cycles of precipitation in the Aburrá valley for (a) the entire period (black line), the month of March (blue line), March 2016 (orange line), and March 2017 (lime line) and (c) the month of October (blue line), October 2016 (orange line), and October 2017 (lime line). (b) Annual cycle of precipitation over the Aburrá Valley (black line) and monthly precipitation during 2016 (orange line) and 2017 (lime line). (d) Monthly PM_{2.5} concentration during 2016 (orange line) and 2017 (lime line).

forecasting of the seasonal and interannual changes in the monthly cumulative daytime and nighttime precipitation.

4. Conclusions

This work introduces an alternative observational methodology to study the processes associated with wet deposition, and more generally, with the net effect of precipitation on pollution concentration near the surface, adding to the important microphysical approach that has been key in the fields of air quality and cloud formation. The method, a nonparametric conditional analysis using the signed overlapping coefficient complement, allows us to assess the net effect of precipitation on the pollutant concentration considering not only the direct near-surface aerosol removal effect associated with below-cloud scavenging but also the indirect effect associated with the induced changes in the lower troposphere, leading to pollutant concentration. In particular, daytime precipitation alters the lower troposphere, inducing near-surface stabilization, which in turn affects the dispersion of pollutants.

The net effect of precipitation on the pollutant concentrations was studied using the monitoring stations located in the Aburrá Valley metropolitan area, considering different times during the day. The results of the study suggest that the effects of precipitation on $PM_{2.5}$ and PM_{10} are strongly dependent on the atmospheric stability. Figure 21 presents a schematic diagram summarizing the main results. During the nighttime (see Fig. 21a) and before midmorning, the potential temperature increases with height, indicating a stable atmosphere; a precipitation event during this time generates below-cloud scavenging, reducing the concentration of the particulate matter in the atmosphere (see Fig. 21b). The nighttime precipitation event slightly stabilizes the midtroposphere, but the near-surface troposphere stability does not change considerably (Fig. 21b). The midtropospheric stabilization is probably associated with warming as a result of latent heating. Nevertheless, the atmosphere was already stable, and the differences are not significant.

During afternoon hours, unstable atmospheric conditions are dominant, as represented by the potential temperature profile in Fig. 21c; convective processes are triggered, and aerosols disperse vertically and away from the valley surface, inducing a considerable reduction in the primary pollutant concentration; however, rainfall stabilizes the atmosphere earlier than under dry conditions, as shown in Fig. 21d. The stabilizing effect of rainfall was confirmed based on the behavior of the ABL, TKE, and thermodynamic indices such as CINE and LI, as a result of shortwave surface radiation forcing.

FIG. 21. Schematic diagram summarizing the net effect of precipitation on the pollutant concentration. (a) During the nighttime and before midmorning, the potential temperature increases with increasing height, indicating that the atmosphere is stable; (b) a precipitation event during this time generates below-cloud scavenging, reducing the concentration of particulate matter in the atmosphere. A nighttime precipitation event slightly stabilizes the midtroposphere, but the near-surface troposphere stability does not change considerably. (c) During afternoon hours, unstable atmospheric conditions are dominant as represented by the potential temperature profile; convective processes are triggered, and aerosols disperse vertically and away from the valley surface, generating a considerable reduction in the primary pollutant concentration; however, (d) rainfall stabilizes the atmosphere, generating early stabilization. Furthermore, anthropogenic emissions continue as represented by the vehicles in the diagram. On a dry day, because of the efficient expansion of the convective boundary layer, pollutant emissions escape the valley. In contrast, on rainy days, the early stabilization leads to near-surface pollutant accumulation, offsetting the washout effect of precipitation.

Moreover, anthropogenic emissions continue as represented by the vehicles in Figs. 21c and 21d. On a dry day, due to the efficient expansion of the convective boundary layer, pollutant emissions escape the valley. In contrast, on rainy days, the early stabilization leads to the near-surface pollutant accumulation offsetting the washout effect (Fig. 21d); in this case, the net effect of precipitation is to increase the concentration of particulate matter.

The method, using the signed overlapping coefficient complement, allowed us to detect the contrasting effect of precipitation at different hours of the day as a result of the competing effects of below-cloud scavenging and atmospheric stability. Additionally, it was possible to see that as the hourly cumulative precipitation increases,

the effects are more significant. On the other hand, the results indicate that PM_{10} was more efficiently removed than was $\text{PM}_{2.5}$, showing the important influence of the particulate matter size on the scavenging process. During nighttime, the reduction of $\text{PM}_{2.5}$ as a result of a precipitation event is on average $10\mu\text{g m}^{-3}$; for PM_{10} , the reduction is on average $17.5\mu\text{g m}^{-3}$. The results also suggest that the year-to-year and month-to-month varying precipitation diurnal cycle modulates the annual cycle of particulate matter concentration, in particular, of $\text{PM}_{2.5}$. This finding underlines the need to study in detail the interannual and long-term variability of the precipitation diurnal cycle; while there is significant research on the interannual precipitation variability on a monthly time scale, not enough research has focused on the long-term variability of the subdaily structure of rainfall, including the frequency of convective versus stratiform precipitation locally.

The results of this work are useful not only for a better understanding of the general mechanisms controlling pollutant accumulation near the surface but also as a tool for policy makers regarding decisions related to air quality control via emission restrictions in the midst of environments characterized by prevalent atmospheric stability and scarce nighttime rainfall, allowing the development of better environmental policies for the prevention or mitigation of air pollution around cities.

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