PM_{10} CONCENTRATIONS MEASURED IN OPEN PIT COAL MINES IN NORTHERN COLOMBIA: SEASONAL VARIATIONS, TRENDS AND SOURCE

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ABSTRACT
PM_{10} particulate material is one of the criteria pollutants with the greatest monitoring in Colombia. Daily concentrations of PM_{10} were evaluated in an open pit coal mine (Cerrejón) located in northern Colombia, during 2012–2017. The annual mean concentration for PM_{10} varied from 22.89 to 41.54 μg/m^3. The annual averages of all sampling sites exceed the WHO guidelines for PM_{10} (20 μg/m^3). Daily PM_{10} concentrations exceeded the WHO PM_{10} guideline limit (50 μg/m^3) for approximately 15.70% of the sampling days. PM_{10} concentrations at all sites presented a significant correlation (r ranged from 0.40 to 0.84). These strong relationships indicate the existence of common sources of PM_{10}. A trend analysis was performed in all monitoring sites. PM_{10} concentrations showed positive trends for four sampling sites. The results of the conditional probability function (CPF) analysis indicate that open pit mining is the main source of emissions of PM_{10}, contributing to concentrations higher than 45 ug/m^3. The Cerrejón mine needs a plan to reduce particulate matter to comply with the new Colombian standards.
Keywords: PM_{10}, conditional probability function, trends, open pit mining, coal mine, Cerrejón Colombia.

1 INTRODUCTION

In the report *Ambient Air Pollution: A Global Assessment of Exposure and Burden of Disease* the World Health Organization (WHO) concluded that 9 out of 10 people live with air that does not comply with its guidelines [1]. These conditions increase mortality at a rate of 3 million people annually in the world.

In Colombia, the total economic burden attributable to environmental risk factors is approximately $2.7 billion. About 70% of this load is attributable to risk factors due to air pollution [2]. PM_{10} (particles $\leq$ 10 μm) is the criteria pollutant of greatest monitoring in Colombia through environmental corporations. This critical pollutant is identified in annual air quality reports in the country for exceeding the maximum permissible levels in monitoring stations located in urban centers and industrial processes [3].

Extraction activities in open pit mining cause emissions of particulate matter by large-scale manipulation of soil and coal [4]. The sources of air pollution in areas of open pit coal mining generally include activities such as drilling, blasting, loading and unloading of overburden, loading and unloading of coal, traffic on unpaved surfaces, coal storage piles (wind erosion and maintenance) and maintenance of roads [4], [5].

The environmental impacts of an open pit coal mine are related to six areas of interest: air quality, water quality and quantity, acid mine drainage, land impacts, ecological impacts and economic impacts [6]. Airborne material has been identified as the main source of air pollution in the area of influence of extractions, especially total suspended particles (TSP) and particles smaller than 10 microns (PM_{10}) [7], [8]. The particulate material originating in the extraction of coal in open pit mines varies in size, morphology, and chemical and physical composition. Atmospheric PM is an interest in the scientific, due to its important impact on public health. Exposure to particulate matter has been associated with excess morbidity and

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mortality [9]–[11]. Open pit coal mines are important sources of particles. These extraction activities have been related to high concentrations of particulate material in their area of influence.

PM_{10} concentrations in open pit coal mines vary from country to country and depend on the capacity of the mine and the season of the year. For example, for PM_{10}, research has registered levels of 50 $\mu g/m^3$ [12], 97 $\mu g/m^3$ [13], 144.8 $\mu g/m^3$, [14], 196 $\mu g/m^3$ [15]; all averages were above of the PM_{10} annual mean established by the guideline values of the World Health Organization [16]. Among the open pit coal mines in the Colombian Caribbean region, the Cerrejón mine is the biggest in the world and is situated on the Guajira peninsula. In Colombia's national air quality report, open pit mining is classified as an important source of PM_{10} emission. It is vital to monitor the historical concentrations of PM_{10} statistically and measure the trend to improve planning and the exploration area.

Meteorological factors have significant impacts on the dilution and diffusion of PM_{10}, causing variation in the concentration levels [17]. Another factor to consider in studies of air pollutants is the trend over time. Studying the temporal behavior of PM_{10} and its correlation with meteorology can provide relevant results for more effective action planning in areas of constant emissions [18].

This study analyzed 6 years of data of PM_{10} from the Cerrejón Air Quality Network (CAQN) to quantify the spatial and temporal behavior of PM_{10}. The trend was calculated to find new strategies for regulating air quality in open pit extraction areas.

2 MATERIALS AND METHODS

2.1 Study area

The study was conducted in the area of influence of the Cerrejón mine (11°5′ N; 72°40′ W) located in the peninsula of La Guajira, in the north of Colombia. The Cerrejón mine is between 85 and 192 m above sea level, with an intervention area of ~142 km^2. The total population of the department of La Guajira by 2019 would be close to 1,067,063 inhabitants. Around the coalfield, there are 80,312 people, of which 34% are indigenous. 50% of this population is located less than 10 km from the site (Fig. 1).

The Cerrejón mine has expansion plans to increase coal production. However, the production of 2019 decreased by about 13% compared to the year 2018. The exploitation is carried out in six extraction areas: Patilla pit (PTp), 100 pit (T100p), Comunero pit (CMp), Oreganal pit (OGp), Tabaco pit (TBp) and La Puente pit (LPp). Fig. 1 is a topographic map of the extraction area, indicating the locations of exploitation pits and population centers.

The semi-desert mining influence zone has an average annual rainfall of about 489 mm, most of which falls from September to November and April to May. The region's average annual temperature is 28.87°C, with little difference between rainy and dry seasons. The wind speed is between 2.53 and 6.04 m/s, with an annual mean of 3.11 m/s. The highest frequency in the wind direction is presented from the northeast.

2.2 Environmental data

The monitoring of particulate material PM_{10} was conducted between 2012 and 2017. In the area of influence of the mining excavations, there were 11 air quality monitoring stations managed by the CAQN. Five monitoring stations of the Network were selected for the analysis. The sites were chosen to cover the entire mine, choosing stations downwind and upwind of the excavation.

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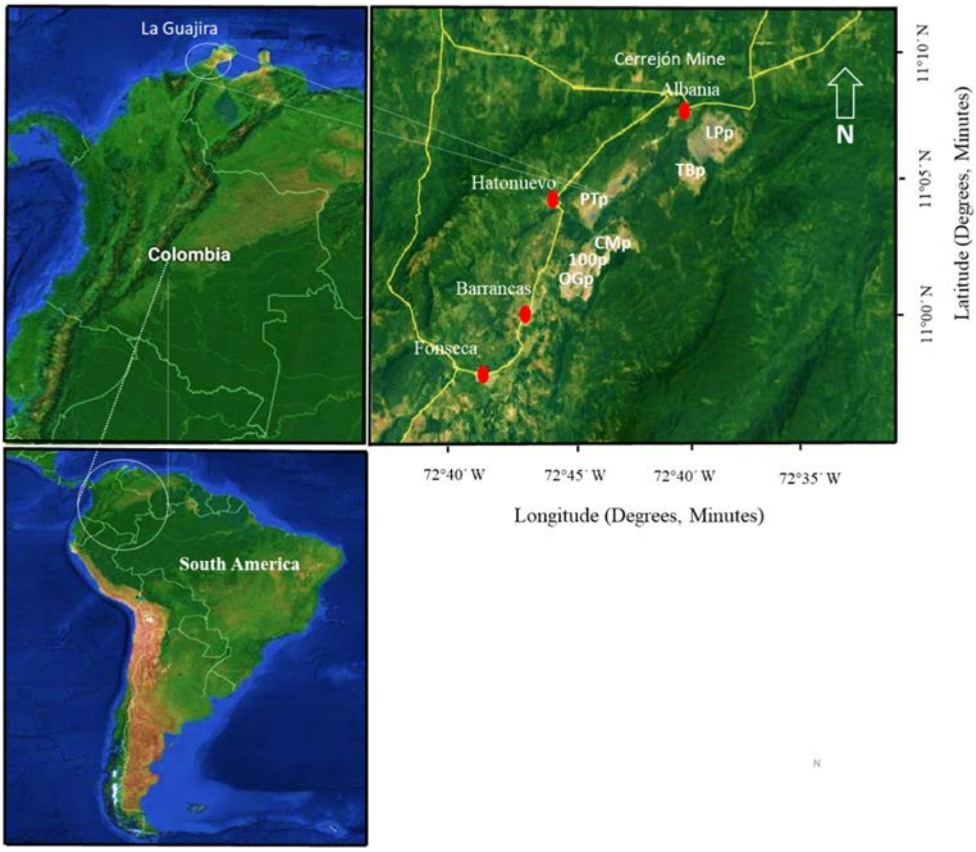

Figure 1: Geographical location of the Cerrejón coal mine in La Guajira (Caribbean region of Colombia). Red circles refer to urban centers. *(Source: Google Earth v7.3.2.5776, 15 Nov. 2019, DigitalGlobe 2019, www.earth.google.com, Accessed on: 22 Jan. 2020.)*

The PM_{10} data were obtained from the Cerrejón Air Quality Network (CAQN). The CAQN processed and analyzed all samples using the reference method for the determination of PM_{10} in the atmosphere [19]. PM_{10} samples were collected continuously from 2012 to 2017 using a high-volume sampler (Hi-Vol), with 24 h samples every 6 days in the period 2012–2014 and every 3 days in the period 2015–2017 for a total of 1,762 PM_{10} samples. Quartz filters were used for the collection of PM_{10}.

The meteorology of the area of influence of the Cerrejón mine was characterized by data collected from three meteorological stations in the same period. The hourly average meteorological parameters such as wind speed (ws, m/s), wind direction (wd, °) temperature (T, °C), relative humidity (RH, %), and rain (P, mm) were collected. Table 1 shows the characteristics of the meteorological stations selected for this study, and Fig. 2 shows geographic location.

Table 1: Location and elevation of the Cerrejón meteorological network.

Site ID	CAQN site	Latitude	Longitude	Elevation (m)	Parameters measured
SySo	Sol y Sombra	11.143	−72.519	117	PM_{10}
PaTi	Patilla	11.050	72.671	115	PM_{10}
PrVi	Provincial	11.022	−72.742	156	PM_{10}, ws, wd, T, P
BaRr	Barrancas	10.960	−72.780	150	PM_{10}, ws, wd, T, P
LsCa	Las Casitas	10.955	−72.741	162	PM_{10}, ws, wd, T, P

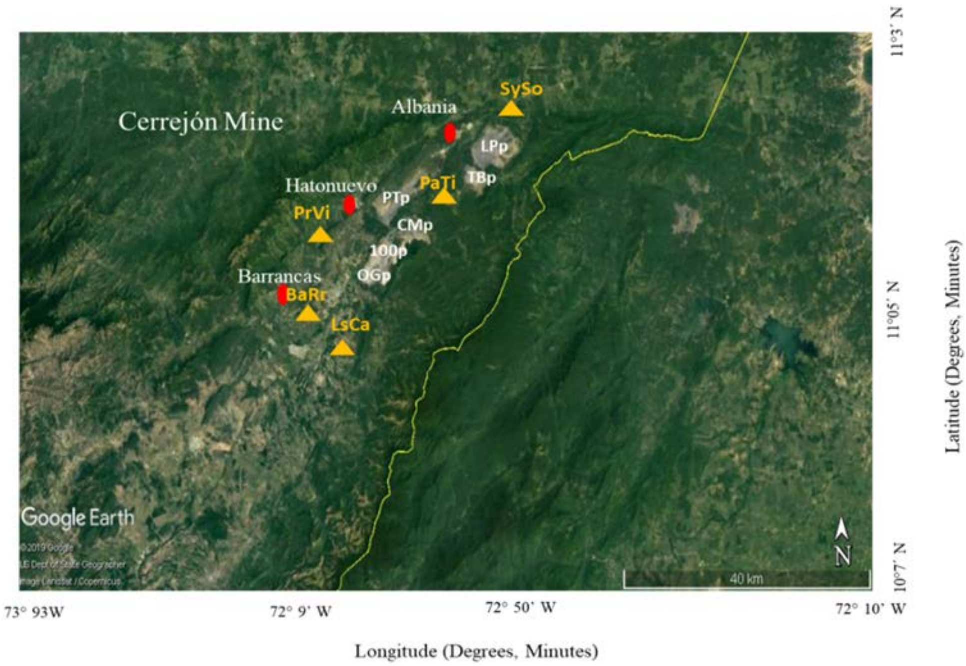

Figure 2: Geographical location of the sampling sites from Cerrejón Air Quality Network. The yellow triangles and the white letters are the monitoring stations and the excavation pits, respectively. *(Source: Google Earth v7.3.2.5776, 15 Nov. 2019, DigitalGlobe 2019, www.earth.google.com. Accessed on: 22 Jan. 2020.)*

2.3 Data analysis

The study was performed using statistical analysis with R statistical programming software (www.r-project.org, R Foundation for Statistical Computing) using Openair (Tools for the analysis of air pollution data) [20]. The relationships between meteorological parameters and PM_{10} were analyzed using Pearson coefficients. Temporal variability of the averages of PM_{10} concentrations was evaluated using the TheilSen function of the Openair package [20], [21]. The polarPlot tool of Openair was used to analyze the PM_{10} concentrations and variability by the wind speed and wind direction.

3 RESULTS AND DISCUSSION

3.1 Meteorological conditions and PM_{10} concentrations

Meteorological parameters were obtained in three meteorological stations (PrVi, BaRr and LsCa) (Table 1). Table 2 shows daily means of temperature (T, °C), relative humidity (RH, %), rain (P, mm), wind speed (ws, m/s), and wind direction (wd, °).

Table 2: Daily averages of the meteorological parameters in the study period (2012–2017). Standard deviation and maximum and minimum values are also presented.

Station	Temperature (°C)				Relative humidity (%)			
	Mean	St. Dev	Min	Max	Mean	St. Dev	Min	Max
PrVi	28.08	2.94	12.80	35.76	60.11	9.57	18.00	96.56
BaRr	28.81	2.01	24.22	34.07	70.02	7.82	40.45	92.25
LsCa	29.80	2.11	13.32	37.01	66.21	9.80	18.00	96.56
Station	Rain (mm)				Wind speed (m/s)			
	Mean	St. Dev	Min	Max	Mean	St. Dev	Min	Max
PrVi	2.03	5.89	0.10	76.60	3.12	1.12	0.00	5.82
BaRr	2.99	7.00	1.02	110.30	2.86	1.04	0.00	5.33
LsCa	1.82	6.89	0.98	107.20	3.12	1.23	0.00	5.73

In the study area, two behaviors were differentiated: dry period and rainy period. The dry period is associated with high temperature ranges between 28.01 and 37.46°C. Relative humidity in the dry period ranges between 18 and 96%. The wind speeds vary from 1.3 to 5.7 m/s with a maximum during the dry period and minimum in the rainy period. The maximum rainfall occurred in October and November, with a daily average of 10.47 mm.

The PM_{10} levels showed a high degree of similarity and common patterns at the sampling sites. Periods of maximum and minimum levels were observed repeating in the same months throughout the 5 years studied. Fig. 3 presents the time series of PM_{10} concentrations for the period studied. Table 3 shows the statistical data on the concentrations of PM_{10} (µg/m^3) by sampling sites.

Table 3: Statistical data of the PM_{10} concentrations (µg/m^3) by sampling sites in the period 2012 to 2017.

	PM_{10} (µg/m^3)					
CAQN site	Mean	St. Dev	Min	Max	Lower $CI_{95\%}$	Upper $CI_{95\%}$
SySo	22.89	22.89	4.87	89.98	33.77	36.12
PaTi	35.42	13.78	9.05	103.77	40.20	42.89
PrVi	38.76	14.87	5.94	128.00	34.17	36.66
BaRr	34.94	14.39	6.65	98.00	37.52	39,98
LsCa	41.54	16.48	4.77	98.00	21.87	23.90

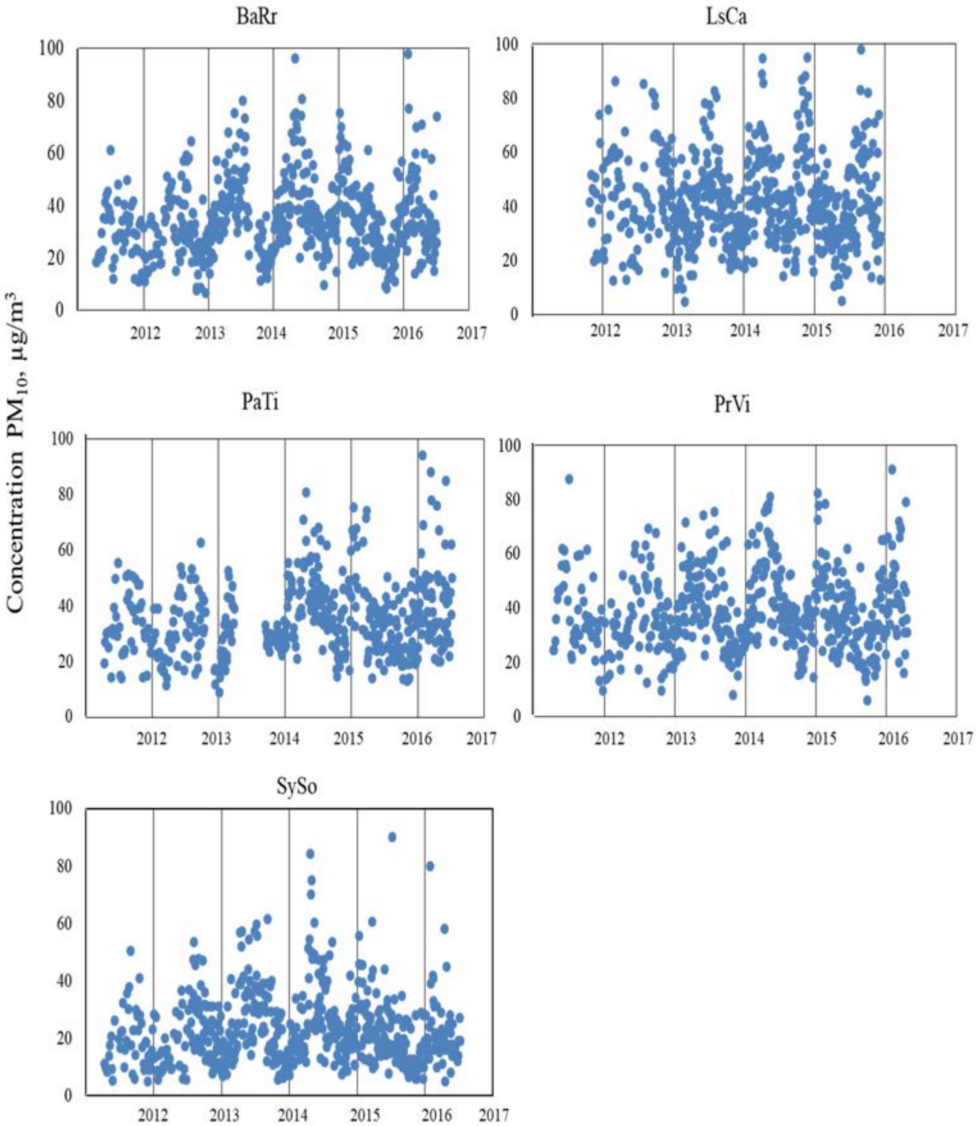

Figure 3: Time series of daily average PM_{10} concentrations for all selected sampling sites during 2012–2017.

The annual mean PM_{10} concentrations were calculated from the daily values and varied from 22.89 to 41.54 µg/m^3, corresponding to the SySo and LsCa sites. These sites are located upwind and downwind of the excavations, respectively (Fig. 2). PM_{10} concentrations were higher at sites located downwind of the excavation.

In the annual averages of the concentrations of PM_{10}, no excess of the annual maximum permissible level (50 µg/m^3) established in the Colombian standard [22] was observed. The annual averages of all sampling sites exceed the WHO guidelines for PM_{10} (20 µg/m^3). The

Colombian standard establishes the annual maximum permissible levels (30 µg/m^3) after January 2030. Under these conditions, open pit extraction would have to intensify controls to comply with national standards. Two 24-hour samples exceeded the daily maximum permissible level for PM_{10} of the Colombian standard (100 µg/m^3). Daily PM_{10} concentrations exceeded the WHO PM_{10} guideline limit (50 µg/m^3) for approximately 15.70% of the sampling days. These levels could increase short-term mortality by about 1.2% over the WHO air quality guideline (AQG) [23].

Pearson correlation coefficients (*r*) were calculated for the relationship of the PM_{10} concentrations upwind and downwind of the excavations (Table 4). PM_{10} concentrations between all sites presented a significant correlation (*r* ranged from 0.40 to 0.84). These strong relationships indicate the existence of common sources of PM_{10}. The results showed coincidences with studies in the same area with fewer years of analysis [24].

Table 4: Pearson correlation coefficients calculated for PM_{10} concentrations in all sites in the period 2012–2017.

	r				
CAQN site	SySo	PaTi	PrVi	BaRr	LsCa
SySo	1	–	–	–	–
PaTi	0.63	1	–	–	–
PrVi	0.56	0.67	1	–	–
BaRr	0.65	0.68	0.84	1	–
LsCa	0.40	0.52	0.58	0.60	1

3.2 Time trends of PM_{10} concentrations

A slight increase is observed for the years 2015 to 2017 for PM_{10} concentrations in the five sampling sites (Fig. 2). Fig. 4 shows the 2012–2017 trends for PM_{10}. In the figures, the numbers show the percent increase or decrease in the trend. The numbers and the symbols show in the figures for each trend estimate are: Percent increase or decrease in trend and how statistically significant the trend estimate is: *** means $p < 0.001$, ** means $p < 0.01$, * means $p < 0.05$, + means $p < 0.1$, and no symbol means no significant trend ($p > 0.1$). The growth rate of PM_{10} concentrations varies from −0.01 to 2.72 µg/m^3/year, at sampling sites BaRr (0.11–2.47), PaTi (0.01–2.72), PrVi (−1.20–2.20) and SySo (−0.11–2.47). The LsCa sampling site presented a decreasing rate of PM_{10} concentrations. BaRr, LsCa and PaTi stations presented statistically significant trends (Table 5).

3.3 PM_{10} source location

The conditional probability function (CPF) analyzes the impacts of point sources in different wind directions, using PM_{10} concentrations and the wind direction measured at the site. The CPF estimates the probability that the contribution of PM_{10} of a source related to the wind direction exceeds a predetermined threshold. Specifically, the CPF is defined as:

$$CPF = \frac{m_{\theta j}}{n_{\theta j}}, \quad (1)$$

where $m_{\theta j}$ is the number of samples in the wind sector θ and wind speed interval j with mixing ratios greater than some "high" concentration, and $n_{\theta j}$ is the total number of samples in the same wind direction–speed interval [25]. Fig. 5 shows a conventional CPF plot calculated

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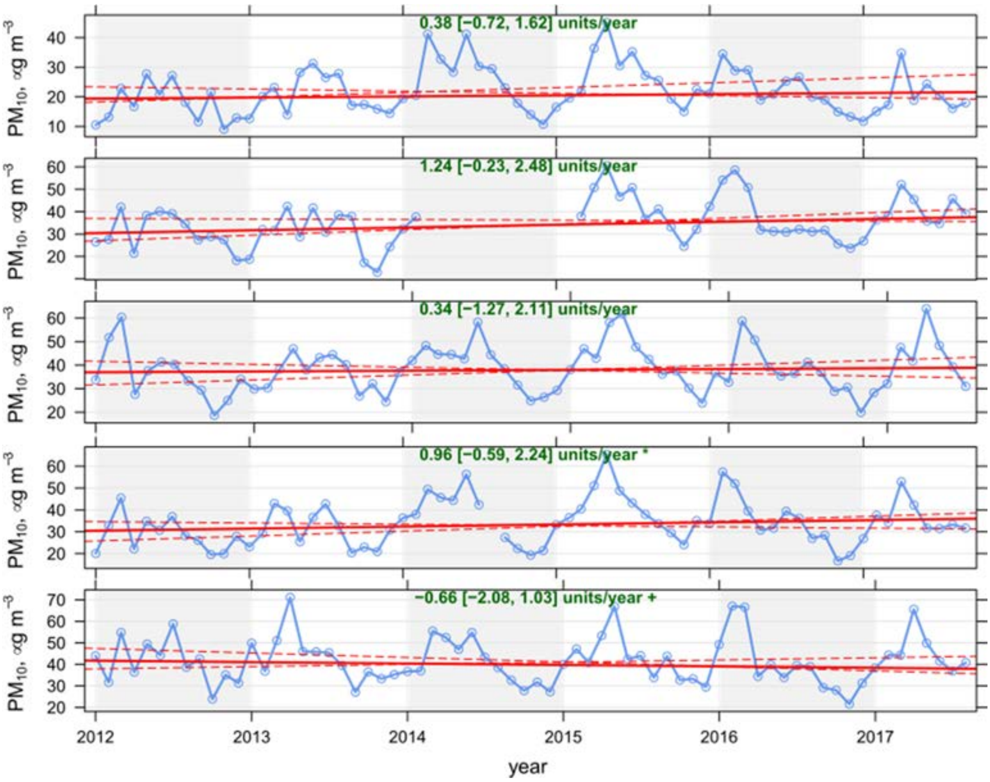

Figure 4: Time trends of annual PM_{10} concentrations at selected sites during 2012–2017.

Table 5: Time trend analysis using the de-seasoned Sen–Theil method. The median slope in %/year, 95% CI of the slope and *p*-values are also shown.

PM_{10}, µg/m^3				
CAQN site	slope, (µg/m^3/year)	95% CI, (µg/m^3/year)		*p*-value
BaRr	1.24	−0.11	2.47	< 0.1
LsCa	−1.34	−2.74	0.34	< 0.1
PaTi	1.24	0.01	2.72	< 0.05
PrVi	0.34	−1.20	2.20	> 0.10
SySo	0.38	−0.72	1.57	> 0.10

with the tool polarPlot package of open-air. CPF was calculated for a 75th percentile PM_{10} concentration consistent with significant emissions from open pit mines equivalent to a PM_{10} concentration (PM_{1075th} > 44 µg/m^3). CPF analysis showed that the highest concentrations of PM_{10} were observed between wind directions 2–5 m/s, with predominant winds from the NNE (BaRr site), NE (LsCa site) and NEE (PrVi site). This analysis indicated the presence of a major source nearby, leading to the contribution of PM_{10}. Sources PTp and CMp showed

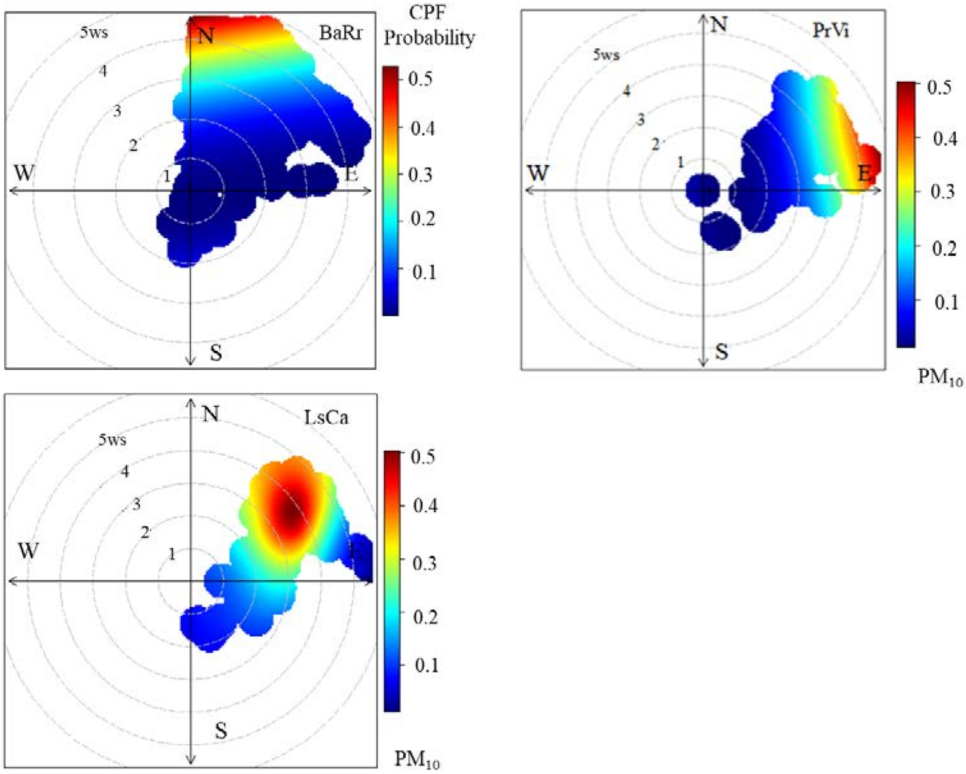

Figure 5: CPF plots for the highest 75% of the PM_{10} mass contributions.

strong influence of their emissions on the PrVi sampling site. CMp, 100p and OGp influenced the BaRr sampling site. Similarly, the LsCa sampling site was influenced by the OGp and 100p sources. CPF analysis with wind directions between 2 to 5 m/s indicates the presence of a significant source nearby [26], [27].

CPF was calculated for a 25th percentile PM_{10} concentration (PM_{1075th} < 28 µg/m^3). The results showed a source contribution at wind speeds of 1 to 2 m/s, indicating the presence of emission sources very close to the sampling site. As cited by [24]: "Further studies on the characterization and distribution of particulate matter collected from the area are needed for comprehensive source apportionment".

4 CONCLUSIONS

The PM_{10} concentrations measured in the area of influence of the Cerrejón mine exceeded the annual mean of the WHO international guidelines at all sampling sites. However, the annual limits of the Colombian standard in these sites were not exceeded. Sampling sites located in the center of the excavations exceeded the WHO PM_{10} guideline limit for approximately 15.70% of the sampling days. The correlation of PM_{10} between sites presented a significant correlation, indicating the existence of common sources. PM_{10} showed increasing trends in concentration for four of the five sites selected in the study. The Cerrejón mine needs to implement a plan to decrease the levels of particulate material systematically to comply with the new Colombian standards by 2030. The results of the CPF analysis clearly showed contributions from PM_{10} from nearby sources. The analysis indicates that open pit mining is the main source of emissions of PM_{10}. The reduction actions planned by the Cerrejón mine in the future would be an important contribution to improving the air quality in the area.

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