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Investigation of PM_{10} sources in Santa Catarina, Brazil through graphical interpretation analysis combined with receptor modelling

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Epidemiological studies have documented that elevated airborne particulate matter (PM) concentrations, especially those with an aerodynamic diameter less than 10 μm (PM_{10}), are associated with adverse health effects. Two receptor models, UNMIX and positive matrix factorization (PMF), were used to identify and quantify the sources of PM_{10} concentrations in Tubarão and Capivari de Baixo, Santa Catarina, Brazil. This region is known for its high pollution levels due to intense industrial activity and exploitation of natural resources. PM_{10} samples were collected using high volume samplers at two sites in the region and statistical exploratory analysis techniques were applied to identify and assess PM_{10} sources. The two primary PM_{10} sources were identified as soil re-suspension/road dust emissions and coal burning emissions, contributing 65–75% and 15–25% of the PM_{10}, respectively. The study confirmed the significance of the influence of local PM_{10} emissions (power plants, soil re-suspension and road dust emissions) on regional air quality, although no violations of the Brazilian PM_{10} standards (limit of 150 $\mu g/m^3$) were observed, with a mean concentration of 27.6 $\mu g/m^3$ measured in this study. This study demonstrated the usefulness of statistical exploratory analysis techniques in assessing the validity of modelling results and contributing to the interpretation of ambient air quality data.

Keywords: air pollution; PM_{10}; statistical exploratory analysis; UNMIX; PMF

1. Introduction

Airborne particulate matter (PM) can be emitted to the atmosphere by a wide range of natural and anthropogenic sources. Elevated concentration of coarse particles, particularly those with an aerodynamic diameter less than 10 μm (PM_{10}) has been associated with adverse health effects in local and regional communities. [1–4] Numerous epidemiological studies have shown an increased morbidity and mortality due to elevated PM_{10} concentration. [5–8] In addition, atmospheric aerosols are implicated in a variety of environmental problems including acid rain, reduced visibility, modification of the earth's radiative balance and changes in cloud properties. [6]

PM properties (size and chemical composition) are a function of emission source, atmospheric reactions and meteorological conditions. Generally, coarse airborne particles are composed of inorganic components (sulphates, nitrates, ammonium, chloride and trace metals), elemental and organic carbon, biological components (bacteria, spores and pollens) and adsorbed volatile and semi-volatile organic compounds. [5] PM_{10} originates from both anthropogenic and natural sources. Combustion processes such as motor vehicle emissions, fossil fuel burning, industrial processes and biomass burning are the dominant anthropogenic sources of PM_{10}, [9,10] while significant natural sources of PM_{10} include volcanic emissions and sea spray. [9]

According to Hopke [11] and Gildemeister et al. [7], the management of air quality is a difficult problem that involves the identification of emission sources of interest, estimation of emission rates and understanding the atmospheric transport of the substances and chemical and physical transformation during transport. Emission source identification and evaluation is a critical step in developing effective management strategies and ultimately reducing environmental PM_{10} concentrations. [6]

Existing research efforts have focused on the development of techniques to identify PM_{10} sources, [9,12] including analyses of spatial and temporal PM concentration variations to identify dominant emission sources and provide a greater understanding of transport. Receptor models, which attribute pollution to sources through statistical interpretation of data, are also a useful tool to support such an analysis. [3,11,13–15] These models focus on the behaviour of the ambient environment at the point of impact and have been used to unravel the contributions of sources to observed ambient PM_{10} concentrations. [16] The most commonly used receptor models are available via the United States Environmental Protection Agency (USEPA):

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the chemical mass balance; UNMIX; positive matrix factorization (PMF) and also principal component analysis (PCA). [17,18]

Pollution (concentration) roses are another technique to identify PM sources. This approach consists of evaluating variations of meteorological conditions and PM concentrations through graphical interpretation. Such an approach allows, over a long period, the determination of the geographical origin and relative importance of PM sources affecting each site. [19–21]

The combination of graphical interpretation analysis and receptor modelling (PMF and UNMIX were the receptor models utilized in the study) has the potential to provide improved identification and prioritization of pollutant sources impacting ambient air quality. The objective of this study is to assess this technique using ambient air quality data for a region where a more conventional source identification study has been previously conducted (to provide a basis for comparison).

The southeast region of the state of Santa Catarina, Brazil, is known for its high pollution levels due to intense industrial activity and the exploitation of natural resources. [22] The largest coal burning thermoelectric complex in Latin America, the Thermoelectric Complex Jorge Lacerda (TCJL), is located in this region. This thermoelectric complex consists of seven power plants with a total capacity of 832 MW that consume approximately 2.9 × 10^6 tons of coal per year. The TCJL power plant has electrostatic precipitators operating at approximately 98% efficiency to remove the particulates in the gaseous effluent. To promote dispersion, the emissions are discharged from two stacks (100 m and 200 m, respectively). [22] Despite these measures, local government and community groups remain concerned about the impact of PM on the public health of residents in the south area of Santa Catarina.

To date, only one study has been completed in this region to identify principal air pollution sources. [22] Godoy et al. [22] utilized a purely statistical method consisting of PCA. A limitation of this approach (and other receptor models) was the inability to distinguish between spatially and temporally correlated sources, such as vehicular and road dust sources, which are perceived as a single source because they almost always impact the receptor at the same time. As the road dust is re-suspended in the air, the motor vehicle passes over the road. [11] Furthermore, PCA can assume several valid statistical results without physical meaning. This study investigated the principal sources of PM_{10} in this region to estimate the contribution of the TCJL and other sources to local and regional atmospheric PM_{10} loading and compare these results with those of the previous PCA-based study.

2. Material and methods

2.1. Sampling and analysis

PM_{10} concentration data were collected in the cities of Tubarão and Capivari de Baixo in the state of Santa Catarina using two high-volume air samplers (AGVMP$_{10}$; Energética Qualidade do Ar, Brazil) equipped with a PM_{10}

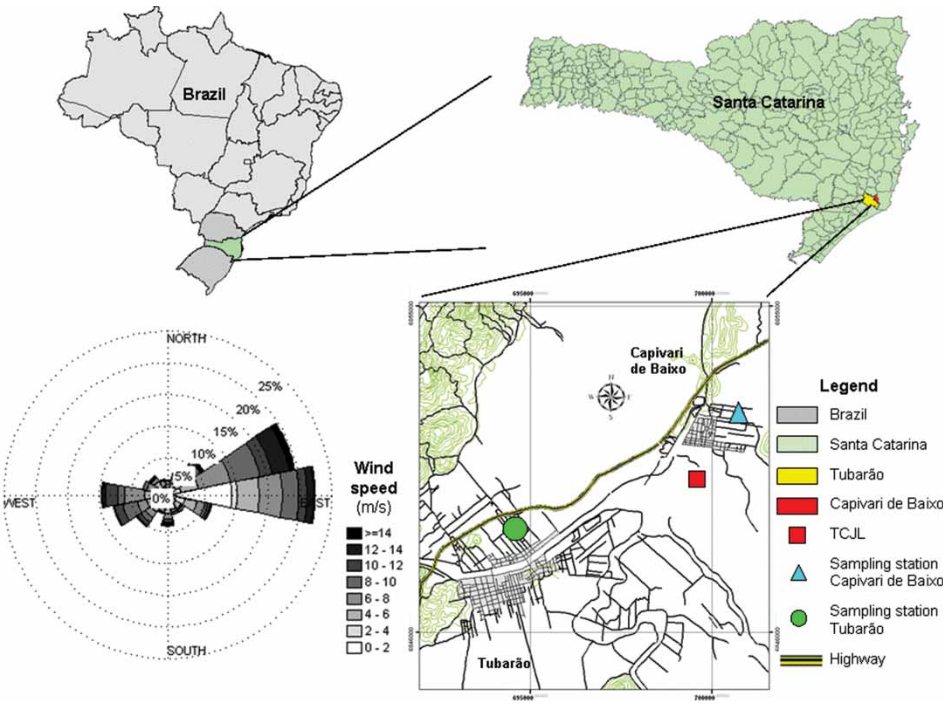

Figure 1. Sampling station location and wind rose.

cut-off inlet to collect particulate samples on a fibreglass filter (0.6 µm pore size; 20.3 cm × 25.4 cm). Soil re-suspension, road dust, marine spray, biomass burning and TCJL emissions are the most probable sources of PM_{10} at Tubarão. The locations of the sampling stations were selected based on the wind frequency (Figure 1) and the distance between the sampling points was less than 8 km. A total of 82 samples were collected between December 2008 to December 2009, with 55 samples collected in Tubarão and 27 in Capivari de Baixo. The flow (around $100\,m^3/h$) and time (around 24 hours) of sampling were regulated by the sampling device AVG-PM_{10} (Energética Qualidade do Ar).

A meteorological station (Davis Vantage PRO 2, USA) was installed in Tubarão at the PM sampling station to provide rain, wind direction, wind intensity, temperature, humidity and atmospheric pressure data. The installation was located on top of a tower (approximately 20 m above ground level) with a 360° unobstructed horizon and unrestricted exposure to the meteorological parameters of interest.

PM_{10} mass concentrations were obtained through gravimetric analysis using an electronic micro balance (AY 220; Shimadzu, Japan). PM_{10} concentrations were determined by the mass rate (collected on fibreglass filters) and volume (regulated by the flow times and the sampling time).

A subset of 50 samples (28 from Tubarão and 22 from Capivari de Baixo) were selected randomly and analysed for metals. The fibreglass filters containing the PM were dissolved in high-purity HNO_3 and H_2O_2 using microwave digestion (Microwave Oven ETHOS PLUS-Sorisole, Italy) in closed Teflon vessels. The resultant solution was then diluted in 50 mL of ultra-pure water to optimize detection. The concentration of arsenic (As), lead (Pb), cadmium (Cd), chromium (Cr), manganese (Mn), mercury (Hg) and nickel (Ni) were determined using a PerkinElmer ELAN 6000 ICP-MS instrument (PerkinElmer, USA). To improve accuracy and precision of the analysis, the system was operated in full quantitative mode. [22,23] The detection limits are based on a 98% confidence level (3 SD).

2.2. *Statistical exploratory analysis*

A statistical exploratory analysis approach was utilized to investigate and characterize the main meteorological influences on the PM_{10} concentrations. Spearman's nonparametric correlations were employed using MATLAB® (MathWorks Inc, USA) and Statistica® (Statsoft, USA) to verify the meteorological relationships for the PM_{10} concentrations at Tubarão and Capivari de Baixo. Pollution roses were applied for graphical interpretation to identify the most polluted wind direction. This approach allowed for the determination of the geographical origin and relative importance of the pollution sources affecting each site. [19,20,24]

2.3. *Receptor models*

The receptor models UNMIX 6.0 (USEPA, USA) and PMF 3.0 (USEPA, USA) were applied to the Tubarão and Capivari de Baixo data in order to estimate the source profiles and contributions. These locations were modelled in a single domain due to their proximity and similar pollution sources. The fundamental principle of receptor modelling is that mass conservation (Equation (1)) can be assumed and a mass balance analysis can be used to identify and apportion sources of airborne PM in the atmosphere [11,25]:

$$x_{ij} = \sum_{p=1}^{p} g_{ip} f_{pj} + e_{ij} \tag{1}$$

where: x_{ij} is the measured concentration of the j^{th} species in the i^{th} sample; f_{pj} is the concentration of the j^{th} species in the material emitted by the source p; g_{ip} is the contribution of the measured i^{th} sample and e_{ij} is the portion of the measurement that cannot be fitted by the model.

UNMIX solves the mass balance problem by utilizing an assumption that the data at the receptor site are a linear combination of an unknown number of sources with unknown chemical profiles. [13,15] The model uses a new transformation method based on self-modelling curve resolution (SMCR) techniques. Since a unique solution is not possible, the SMCR technique restricts the feasible region of the real solution into a small region with explicit physical constraints, such as requiring source compositions to be greater than or equal to zero. Further description of this model can be found in Hopke [11] and USEPA. [17]

In both the PMF and UNMIX receptor models, sources are constrained to have non-negative species concentration and no sample can have a negative source contribution. [15] The primary difference between the PMF and UNMIX models is that the PMF model considers the uncertainties in the measured variables. PMF takes the approach of an explicit least-squares approach, in which the method minimizes the ratio established between residual modelled and analytical error (Equation (2)) with the constraint that each of the elements of g and f is to be non-negative [11]:

$$Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \frac{\left(x_{ij} - \sum_{p=1}^{p} g_{ip} f_{pj}\right)^2}{s_{ij}^2} \tag{2}$$

where Q is the ratio between residual modelled and analytical error and s_{ij} is an estimative of the uncertainty in the j^{th} variable measured in the i^{th} sample. [11,17,18]

In this study, eight variables were measured; however, the number of species used in the models was reduced to seven (PM_{10}, As, Mn, Ni, Cr, Pb and Cd) due to the Hg concentrations being below the analytical detection limit. These metals were chosen based on previous work conducted in the area (Evaluation of the environmental quality of the coal basin in Santa Catarina). [22] For the variables applied, no detrimental points (*i.e.* extreme outliers and/or

Table 1. Average PM$_{10}$ and species concentrations measured at the two sites.[1]

Site	Parameter	Number of samples	Mean ± SD (ng/m^3)	Min (ng/m^3)	Max (ng/m^3)	MRL (ng/m^3)	Detec. Limits^a (ng/m^3)
Tubarão	PM$_{10}^{b}$	55	27.62 ± 11.14	8.44	60.03	150^c	1
	Mn	28	21.04 ± 0.22	6.0	52.0	3000	4.70
	Ni	28	2.5 ± 0.06	0.25	13.0	90	1.11
	As	28	3.23 ± 0.10	–	16.0	5000	2.45
	Cd	28	0.35 ± 0.03	–	1.3	10	0.17
	Pb	28	12.73 ± 0.16	3.8	38.0	1000	3.66
	Cr	28	8.13 ± 0.57	–	24.58	300	2.45
	Hg	28	0.020 ± 0.001	–	0.11	200	0.07
Capivari de Baixo	PM$_{10}^{b}$	27	22.99 ± 10, 44	4.19	48.23	150^c	1
	Mn	22	19.72 ± 0.07	7.1	44.0	3000	5.44
	Ni	22	2.18 ± 0.10	0.52	7.6	90	1.14
	As	22	3.48 ± 0.02	0.06	13.4	5000	2.99
	Cd	22	0.34 ± 0.17	–	0.77	10	0.12
	Pb	22	14.31 ± 1, 7	01.8	39.0	1000	5.20
	Cr	22	8.29 ± 0.01	0.45	38.0	300	3.61
	Hg	22	0.040 ± 0.001	–	0.24	200	0.18

Notes: MRL = minimal risk level. [26]

[a] Average detection limits (3 SD of each sample).

[b] Concentrations in μg/m^3.

[c] Brazilian and USA national ambient PM$_{10}$standards in μg/m^3. Not to be exceeded more than once per year on average over 3 years.

concentrations below the detection limit) were identified; therefore, all data points were included in the modelling.

3. Results and discussion

3.1. Statistical exploratory analysis

The average PM$_{10}$ concentrations and corresponding chemical composition during the sampling period at the two sites are shown in Table 1. The average PM$_{10}$ concentrations for the entire sampling period was 27.62 ± 11.14 μg/m^3 in Tubarão and 22.99 ± 10.44 μg/m^3 in Capivari de Baixo. During the sampling periods, no exceedance of the Brazilian PM$_{10}$ standards (150 μg/m^3), the USEPA PM$_{10}$ standards or the minimal risk level for hazardous substances listed by the Agency for Toxic Substances and Diseases Registry were observed.

Observed PM$_{10}$ concentrations were similar at the two sampling sites during the monitoring period (Figures 2 and 3), suggesting a strong similarity between the source of the particle emissions at the sampling sites. The slightly higher PM$_{10}$ concentration at Tubarão could be associated with its larger population and higher level of urban activity (96,529 inhabitants compared to 19,934 inhabitants in Capivari de Baixo) resulting in increased PM$_{10}$ emissions from road dust and soil re-suspension. As seen in Figure 1, the wind directions from the northeast and east are the most frequent, suggesting that the particulate emissions from the TCJL are most likely impacting Tubarão. However, if the TCJL strongly contributed to the PM$_{10}$ emissions in Tubarão, the concentration of PM$_{10}$ at both sites should not be similar, as previously observed by Godoy. [22]

As data for other meteorological parameters such as atmospheric stability, solar radiation and cloud cover were not directly available, their influence could not be evaluated. Data from meteorological models would be required to obtain these parameters.

While the spatial PM$_{10}$ concentration variations appeared to be low, significant temporal variations were observed at the two sampling sites, with the greatest PM$_{10}$ variability being observed during autumn (March to June), winter (June to September) and spring (September to December). These seasons are the drier seasons, with a relatively lower frequency of rain, which favours atmospheric accumulation of pollutants. The highest PM$_{10}$ concentrations were also observed during these seasons. The large concentration increase during the summer of 2010 is due to the small pool of data that coincided with high concentration events that are not expected to be representative of typical summer conditions.

No significant chemical composition differences were observed between the PM$_{10}$ collected from both sampling sites (Figure 4). The concentration of Mn and Pb were found to be the highest in the PM. While the PM was analysed for Hg, many samples were below the method detection limit (0.07 and 0.18 ng/m^3).

A non-parametric correlation (Spearman ranking) between the meteorological data and the PM$_{10}$ concentrations at Tubarão and Capivari de Baixo is presented in Table 2. As expected, rain (mm/day) was the meteorological parameter with the strongest correlation to the measured PM$_{10}$ concentrations at the two sampling sites. Other daily average meteorological parameters (with the exception of

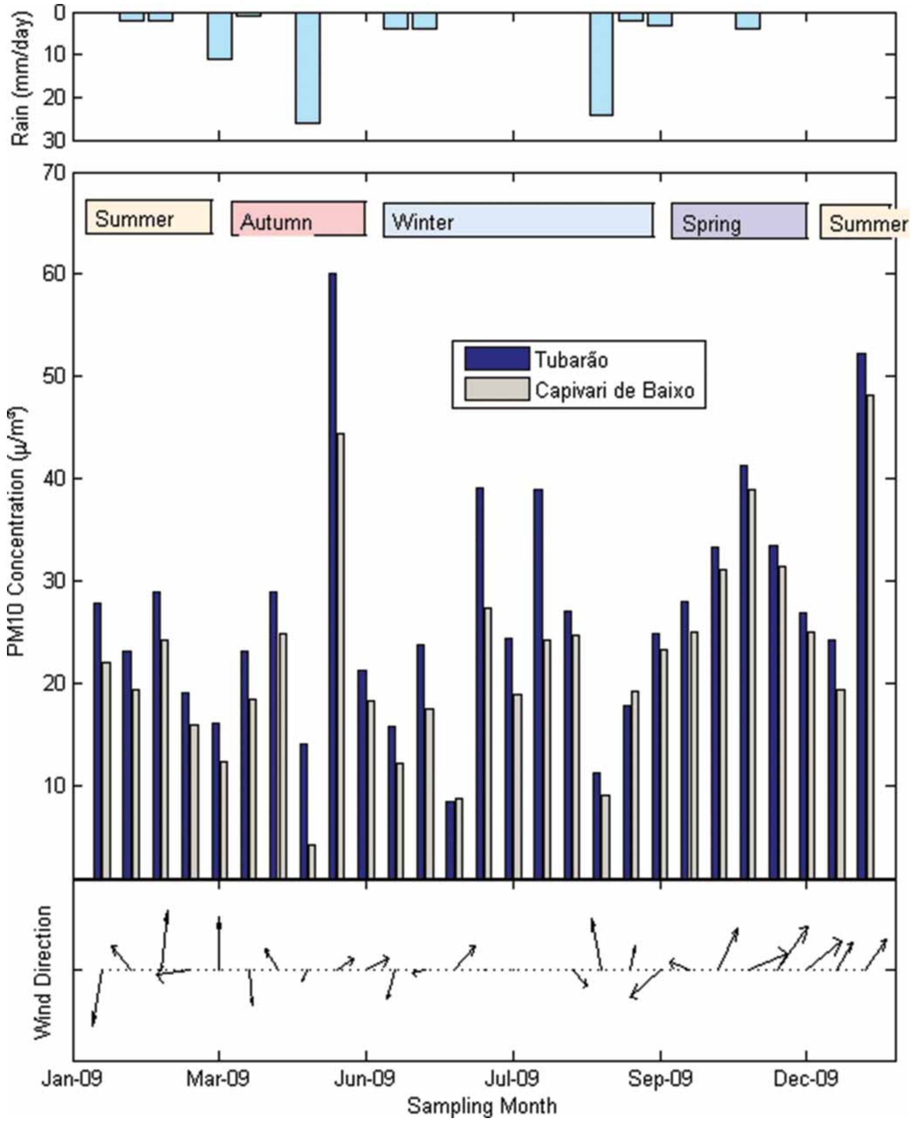

Figure 2. PM_{10} concentration observations at Tubarão and Capivari de Baixo. Precipitation in mm/day during the sampling date. Wind direction (arrowhead indicates wind provenance) and daily average speed (arrow size indicates the intensity). Sampling camping frequency was not equispaced.

wind speed and direction) did not have a strong correlation with the observed PM_{10} concentrations. The relationship between wind speed and PM_{10} concentration is not yet fully understood and, as noted by Weiner and Matthews, [27] a more frequent and longer sampling period could be needed to explore this relationship further.

The pollution roses (Figure 5) indicated that the winds from the east-northeast (ENE) direction resulted in the highest PM_{10} concentrations at Tubarão and Capivari de Baixo. The winds from the ENE are already moderately polluted (with PM_{10} up to 40–50 μ g/m^3) before reaching Capivari de Baixo and the atmospheric PM_{10} concentration of this ENE wind increased after it passed through the cities of Capivari de Baixo and Tubarão(PM_{10} up to 60–70 μg/m^3).

Wind events with high PM_{10} concentrations arriving in Capivari de Baixo are likely a result of the drier nature of wind from the North, Northeast and ENE directions. Under a prevailing ENE wind, suspension of soil particulates, TCJL emissions and other emissions related to urban activities could contribute to the observed increase in PM_{10} concentration in wind reaching Tubarão after passing through Capivari de Baixo. In particular, the up-wind (during the prevailing ENE winds) location of the coal-fired TCJL power station relative to the city of Tubarão (Figure 1) could contribute to increase the PM_{10} levels. However, the PM_{10} measured concentration at both sites did not suggest a significant impact of the TCJL. The origin of the pollution sources remains unclear. A longer sampling campaign could help to explain the uncertainties.

As shown in Figure 5, winds from the southern quadrant had much lower PM_{10} concentrations relative to ENE winds. In this region, southern winds are commonly

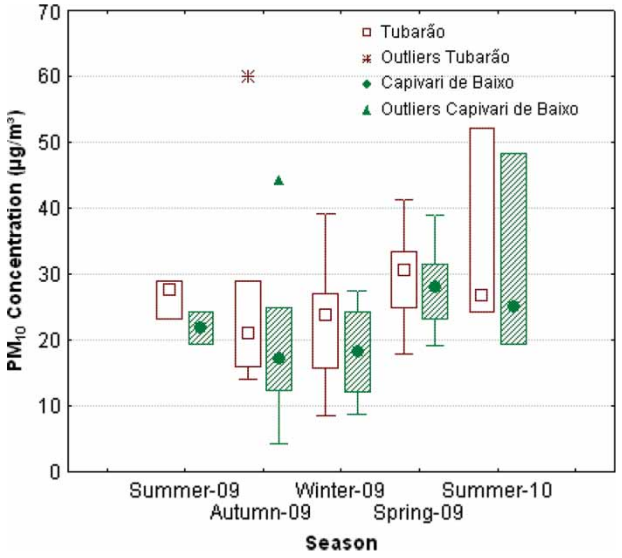

Figure 3. Seasonal variations in PM_{10} concentrations at in Tubarão and Capivari de Baixo. Summer (from December to March), autumn (from March to June), winter (from June to September) and spring (from September to December).

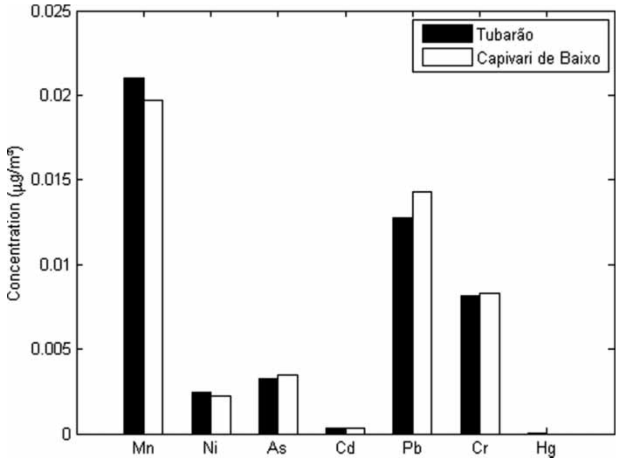

Figure 4. PM_{10} chemical composition during the sampling campaign at the two sites. Average concentrations of the analysed samples during the sampling campaign.

accompanied by cold fronts and rain in the south of Santa Catarina; therefore, a southerly wind direction is apparently less favourable for PM_{10} air pollution, probably due to rain deposition of suspended particulates.

3.2. Source apportionment by UNMIX and PMF

Previous work by Godoy et al. (2005) identified sea spray as a significant source of the coarse fraction of the PM_{10} (35–82% of the amount of $PM_{2.0-10}$) at the studied site, but not for the fine particles. Soil re-suspension (13–42%), followed by TCJL emissions (5–15%) and road dust (5–8%) are the other $PM_{2.0-10}$ potential sources suggested by Godoy. [22]

3.2.1. UNMIX

The UNMIX model was applied to the PM_{10} chemical composition dataset (with the exception of Hg). Since UNMIX is unable to perform these calculations using only two sources, the modelling was performed using three source apportionments. The overall particulate matter (PM_{10}) concentration predictions compared well with the measured data, with a coefficient of determination (r^2) of 0.72 (Figure 6).

Two sources (soil re-suspension and TCJL) accounted for 81.5% of the original dataset covariance, while the third source only accounted for 0.5%. The magnitude of the third source did not allow its identification, even though it contributed approximately 40% of the Ni concentration. The normalized source profiles found in the present work are presented in Table 3.

The first source had the largest contributions to ambient PM_{10} and Pb levels and was also responsible for approximately 80% of the PM_{10} and 40% of the ambient Mn. It is likely that this source was associated with soil re-suspension and road dust emissions, as described previously by Godoy. [22] Emissions arising from road vehicles are generally contributed by a mix of tailpipe emissions, wear and tear of brakes and tyres and re-suspension of road dust. Genuine overlaps in chemical composition between soil and road dust are common in urban regions. [28]

The second source was similar to the TCJL emission profile, with significant contributions from the measured Cr, Pb, As, Cd, Ni and Mn concentrations. Overall, source 2 contributed approximately 20% of the measured PM_{10} concentration, which was in agreement with the results modelled using PCA by Godoy. It was not possible to identify source 3 due to limitations in the data, a greater number of samples and additional analysed species would be required to complete the identification.

Table 2. Spearman-rank non-parametric correlation between meteorological data and PM_{10} concentrations in Tubarão and Capivari de Baixo.

	Temperature					
Location	Mean (° C)	Max (° C)	Min (° C)	Rain (mm)	ATM pressure (hPa)	RH (%)
PM_{10} - Tubarão	0.11	0.21	0.04	**−0.43**	0.01	−0.20
PM_{10} - Capivari	0.23	0.36	0.11	**−0.54**	−0.09	−0.31

Notes: ATM = atmospheric; RH = relative humidity.
Statistical significance of 5%.
Numbers in bold indicate valid statistical correlation. All parameters were computed as daily averages.

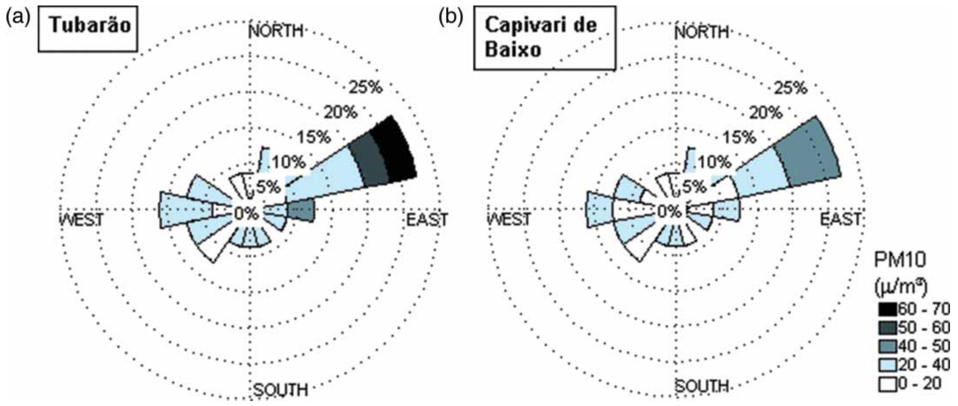

Figure 5. PM_{10} pollution roses in Tubarão (a) and Capivari de Baixo (b). PM_{10} concentrations in $\mu g/m^3$ and wind direction. Frequency of the concentration events in percentage.

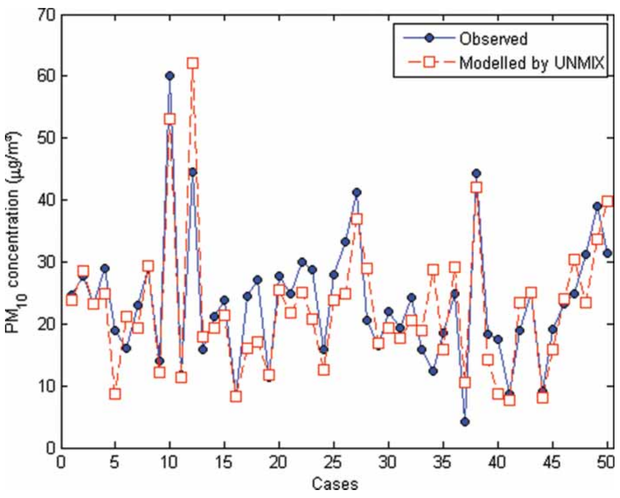

Figure 6. Comparison between observed and modelled time series by UNMIX for PM_{10} concentration.

Table 3. Source profiles modelled by UNMIX.

Element	Source 1 Soil re-susp (ng/m^3)	Source 2 TCJL (ng/m^3)	Source 3 Unknown (ng/m^3)	(r^2)
PM_{10}^a	17.40	4.23	0.84	0.72
Mn	8.34	10.8	1.13	0.85
Ni	0.01	1.40	0.94	0.99
As	0.00	3.43	0.07	0.81
Cd	0.15	0.21	0.01	0.66
Pb	7.45	6.29	0.07	0.58
Cr	0	9.17	0	0.84

Notes: re-susp = re-suspension.
[a]Concentrations in $\mu g/m^3$.

3.2.2. *PMF*

The PMF model was applied to the same dataset used for the UNMIX modelling, with the normalized source profiles presented in Table 4. The model had good predictive capacity for PM_{10}, Mn, Pb and Cr (Table 4); however, the Ni concentration predictions were poor ($r^2 = 0.37$).

Table 4. Source profiles modelled by PMF.

Element	Source 1 Soil re-susp (ng/m^3)	Source 2 TCJL (ng/m^3)	Source 3 Unknown (ng/m^3)	(r^2)
PM_{10}^a	15.77	0.00	5.83	0.74
Mn	11.1	5.34	3.47	0.96
Ni	1.22	0.88	0.16	0.37
As	0.53	2.58	0.00	0.58
Cd	0.15	0.14	0.01	0.52
Pb	2.66	0.01	10.5	1.00
Cr	0.00	7.71	0.10	0.97

Notes: re-susp = re-suspension.
[a]Concentrations in $\mu g/m^3$.

The predicted contribution of each of the three sources using the PMF model was similar to those obtained using UNMIX (Tables 3 and 4). This was as expected due to the similar approaches utilized by the models. [11] According to Hopke, [11] in most cases, the source profiles are similar and, despite some discrepancies, Poirot et al. [29] also concluded that sources identified by UNMIX and PMF were similar.

Source 3 did not contribute sufficiently to explain the variation in the data, similar to the UNMIX modelling. However, source 3 appears to be the main contributor to Pb emissions and about 25% of PM_{10}, suggesting that another localized and/or unidentified sources exist. The absence of elements such as Fe, magnesium (Mg), silica (Si), sodium (Na) and other organic and inorganic species could lead to the models being insensitive to other particulate emissions sources at Tubarão and Capivari de Baixo. Source 2 (TCJL emissions) did not appear to have any significant contribution to the measured PM_{10} concentrations, but it has a significant contribution to the concentration of metallic elements. The differences between the UNMIX and PMF results, especially for Ni and Pb concentrations, could not be explained within the context of this study.

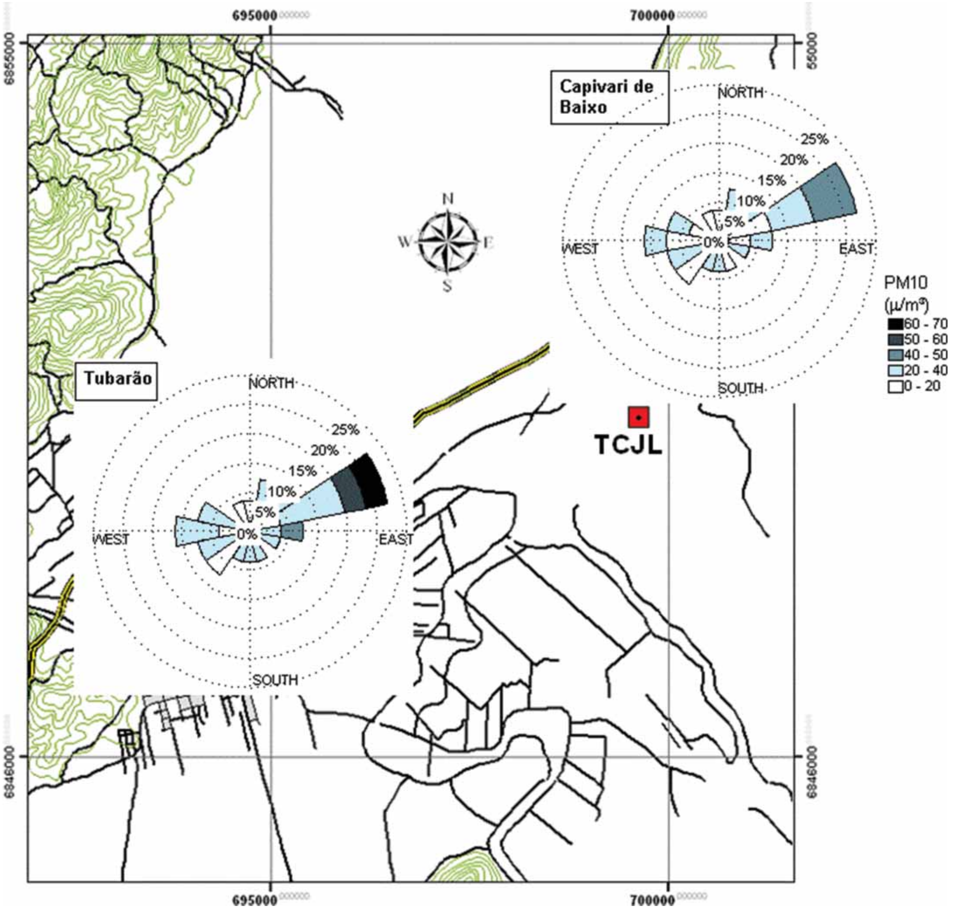

Figure 7. Tubarão and Capivari de Baixo Map, TCJL position related to the cities and pollution roses of respective cities located under the sampling points.

3.3. Application of pollution roses to receptor modelling results for source identification

The pollution roses (Figure 5) showed similar conclusions to the UNMIX and PMF modelling results. Under north or northeast winds, the air had already gained the majority portion of the PM_{10} before it reached the city of Capivari de Baixo and it is most likely that this PM_{10} (40–50 $\mu g/m^3$) originates from soil re-suspension and road dust emissions (source 1) since there is no other major urban area (only small cities) within 100 km. In his study, Hopke [11] also comment that airborne concentrations due to specific sources may display a sharp directional pattern with regard to wind direction.

The smallest representative component of the PM_{10} was predicted (by UNMIX and PMF) to originate from the TCJL emissions. After passing through the cities of Capivari de Baixo and Tubarão, the PM_{10} concentration in the air under North, Northeast or Easterly winds increases, according to the pollution roses (Figure 5). As the TCJL (Figure 7) is located between these cities, it is most probably the cause of the increase in the PM_{10} concentration (0–10 $\mu g/m^3$), indicated by the darkening on the Tubarão pollution rose under an ENE wind direction.

3.4. Comparison with previous study

In Figures 8 and 9, the soil re-suspension and TCJL profiles determined using the UNMIX and PMF models are compared with the results obtained by Godoy [22] in the 2001

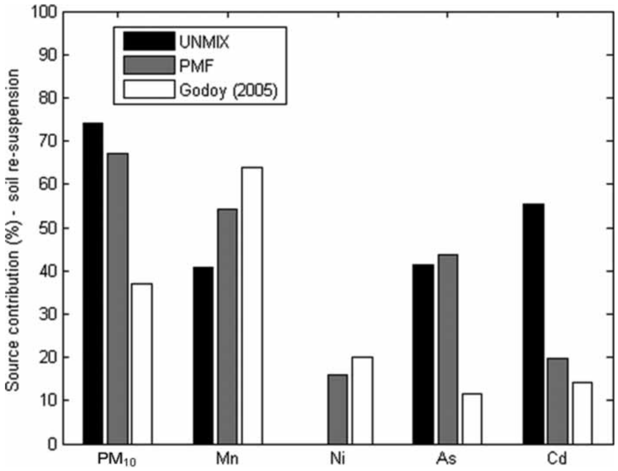

Figure 8. Soil re-suspension PM_{10} profile estimated by UNIMIX, PMF and by Godoy et al. [22]

study for the same sites. Godoy [22] used PCA modelling to identify and quantify the principal sources of PM around the TCJL, based on the analysis of up to 42 elements from up to 120 results per element. After applying the model, Godoy [22] identified four main sources; soil re-suspension; TCJL emissions; sea spray; biomass burning.

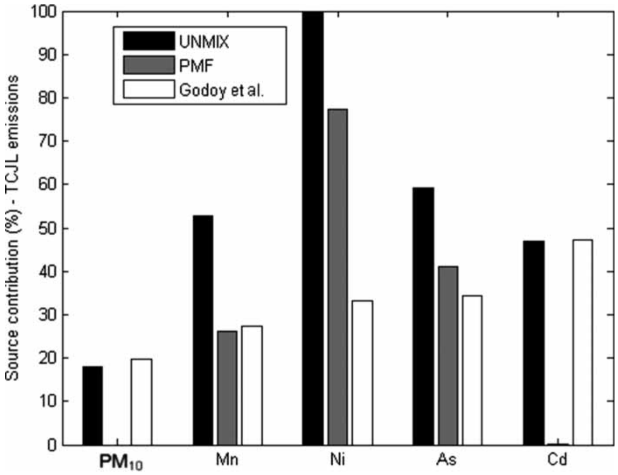

Figure 9. TCJL PM_{10} profile estimated by UNMIX, PMF and by Godoy et al. [22]

As the selected range of chemical elements did not include those representative of sea spray and biomass burning sources (aluminium (Al), Si, black carbon (BC), Na, chlorine, Fe, Mg) were not detected by the UNMIX and PMF modelling in this study due to the chemical elements not being determined. Organic, inorganic and metallic traces elements such as BC, Na, Si, Mg, Al, titanium (Ti) can represent a large mass amount of PM_{10} concentrations, and their inclusion would improve the UNMIX and PMF modelling results and allow identification of additional emission sources. In some cases, PMF and UNIMIX leaves some of the mass unexplained, as reported by Poirot et al. [13] and Polissar et al. [29].

The obtained source profiles were compared with equivalent source profiles for TCJL fly ash and local soil, [22] with the results presented in Figure 10. In general, a good agreement is observed, but the local soil profile hade lower levels of Pb than the profile generated by UNMIX and PMF. The TCJL fly ash profile was in good agreement with the corresponding profile generated using the PMF model.

4. Conclusion

A combination of modelling (UNMIX and PMF) and graphical interpretation methods were applied to

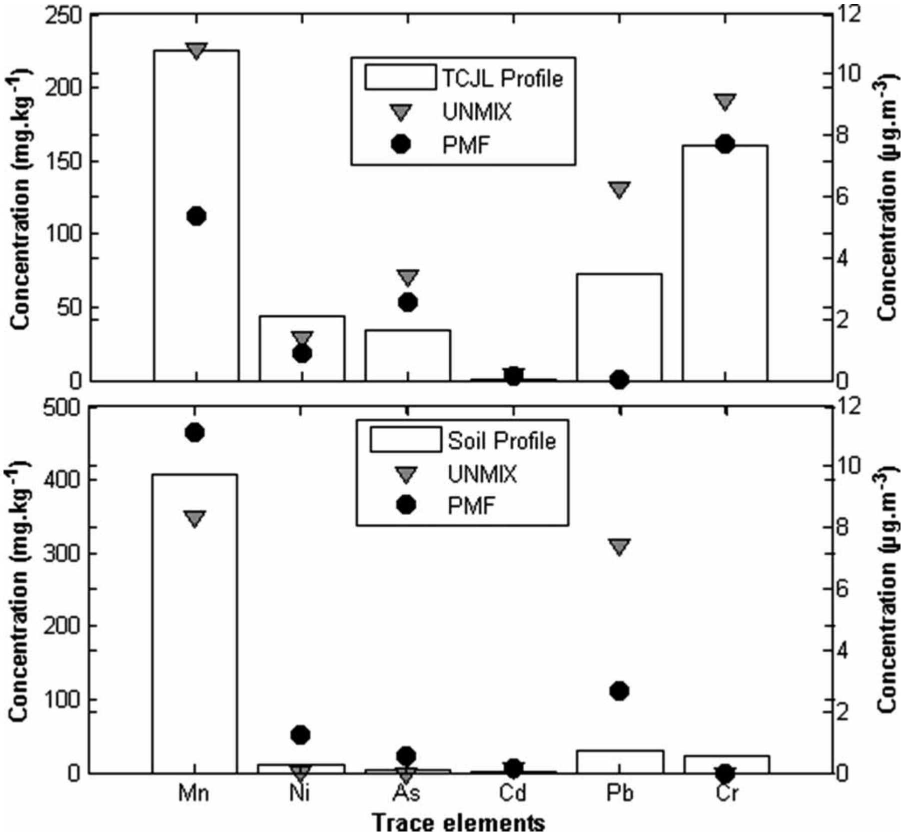

Figure 10. TCJL and soil profiles obtained by Godoy [22] compared to profiles reached by UNMIX and PMF. Profiles in mg/kg (left axis) and UNMIX and PMF results in µg/m^3 (right axis).

identify dominant sources for measured ambient PM_{10} concentrations at two sites located in Tubarão and Capivari de Baixo, Brazil. It should be noted that the total number of samples was relatively small (50 samples); however, the results of this study were in general agreement with those obtained by Godoy, [22] which was based on a much larger experimental study.

During the sampling periods, no exceedance of the Brazilian PM_{10} and the USEPA PM_{10} standards were observed. Based on the receptor models employed (UNMIX and PMF), the TCJL contributions to the ambient PM_{10} concentrations were estimated to be between 0 and 20% at the two monitoring sites, with the TCJL being identified as the main source of As and Cd in the PM_{10}. Soil re-suspension appeared to be the main source (60–75%) of the ambient PM_{10} at Tubarão and Capivari de Baixo. Additional analysis with a larger dataset and the use of an expanded speciation of the organic and inorganic PM_{10} components would improve the identification and quantification of PM_{10} sources.

Despite using a different methodology, models and a reduced number of field samples, the study results confirm those obtained by Godoy, [22] and demonstrate that regional PM_{10} sources can have a significant impact on local PM_{10} concentrations. The study demonstrated that the receptor-based techniques and statistical exploratory analysis are useful tools in identifying PM_{10} sources and evaluating their impact on ambient air quality, supporting the development of defective management strategies and reductions in environmental PM concentrations.

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