VARIABILITY OF PM10 IN INDUSTRIALIZED-URBAN AREAS. NEW COEFFICIENTS TO ESTABLISH SIGNIFICANT DIFFERENCES BETWEEN SAMPLING POINTS

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Abstract

One of the main problems that arise in the assessment of air quality in an area is to estimate the number of representative sampling points of each microenvironment within it. We present a new model that reduces the variability and increases the quality of the comparison of the sampling points. The study is based on the comparison between a city in eastern Spain, Vila-real, a macro city in México, Monterrey and the Piemonte region regarding the assessment of PM10 in microenvironments. Vila-real is located in the province of Castellón. This province is a strategic area in the framework of European Union (EU) pollution control. On the other hand, Monterrey in México, located in the northern state of Nuevo León, has several problems with particulate material in the atmosphere produced by the extraction of building materials in the hill that surround the city. Finally, the Piemonte region, which is located in the north of Italy, has to be in consideration due to higher concentrations of PM10 in the Po river basin. In the case of Vila-real the PM10 samples were collected by a medium volume sampler according to European regulations. Particle concentration levels were determined gravimetrically (EN 12341:1999). In the case of Monterrey the PM10 concentrations were determined by Beta Ray Attenuation according to US-EPA regulations. In the Piemonte region, the average concentration of PM10 was also obtained by means of the Beta Ray Attenuation as well as using gravimetric instruments. The methodology carried out in this paper is a useful tool for developing future Air Quality Plans in other industrialised areas.

Key Words: PM10, sampling points, statistical methodology, industrialized-urban areas

Capsule: Estimate de number of representative sampling points in a polluted area through a new statistical tool.

1. INTRODUCTION

Urban sprawl has been an important feature in the process of human development all throughout history. This trend is often associated with spatial mismatch from the countryside to the cities (Agrawal et al., 2003). This expansion has led to a deterioration of air quality in cities. Inhabitants of high dense cities are directly exposed to higher concentrations of different pollutants (Massoud et al., 2011).

Particulate matter (PM) is currently considered the best indicator for health effects of ambient air pollution (Burnett et al., 2014, WHO, 2014). Human exposure to PM in urban micro-environments is of particular interest because it has been demonstrated that their levels are particularly dangerous for health. (Pope and Dockey, 2006; Kampa and Castanas, 2008). Airborne particles are among the most important pollutants that adversely influence human health in urban areas due to their great potential of reaching the furthest part of the lungs (Unal et al., 2011). Some studies have shown a positive correlation between high concentrations of particles and deterioration in public health (Kappos et al., 2004; Neuberger et al., 2004; Le Tertre et al., 2005; Wilson et al., 2005). PM has been associated with causes of morbidity and mortality (Pope et al., 2002; Vedal et al., 2003; Guaderman et al., 2004).

PM results from emissions of diverse pollutants from different stationary and mobile sources, and from chemical reactions between primary pollutants that form secondary pollutants that, in turn, form again secondary pollutants. Due to chemical and physical processes that originate PM, vertical and/or horizontal transport condensation and photochemical reactions in addition to traffic intensity and the location of the buildings and another obstacles, concentrations of PM may vary spatially in urban sites. Accordingly, urban air quality is characterized by high spatial and temporal variability (Moore et al., 2009; Moltchanov et al., 2015). Therefore, studies on the intra-city spatial variation become crucial from the point of view of habitability and health risk. One of the main problems that arises in the assessment of air quality in an area is to estimate the number of representative sampling points of each microenvironment in it. A common approach to tackle this task has been based on Geostatistical analysis (McBratney and Webster, 1983; Yfantis, et al., 19987;Wester and Oliver, 1990 and van Groeningen, 2000). However, these methods rely on a good estimation of the spatial covariances which requires several operative monitoring sites beforehand. In this study, we present a method to decide which sampling points are similar and from this information decide the minimum distance between them using the distance between monitoring stations as a covariate. We also present a new statistical methodology to facilitate this decision as a well as an algorithm to obtain a monitoring network. The corresponding schedule step by step is presented in Figure 4. Through this schedule the researchers can follow the procedure to develop the statistical analysis.

The study is based on the assessment of PM10 (particulate matter with aerodynamic equivalent diameter lower than 10µm) in microenvironments within a city in eastern Spain, Vila-real, in a macro city in México, Monterrey and in Piemonte region in Italy. First of all, we start with the assessment of the spatial PM10 variability and test if the normality assumption for the data is valid, and the proceed with the subsequent statistical analysis, which includes ANOVA analysis and the study of the coefficients presented for other authors, Pearson's Coefficient of Correlation and Coefficient of Divergence (COD) (Wilson et al., 2005). Second, a new Coefficient of Diversity and Redundancy (CODR) is presented and also the CODRcv with the inclusion of the variability in it, more specifically, the variation coefficient of Person. Finally, these new CODR and CODRcv are applied to the PM10 data.

2. DESCRIPTION OF THE STUDY AREAS

The study is carried out in three industrial areas, Vila-real in Spain, Monterrey in Mexico and in Piemonte region in Italy.

Vila-real is a city in an industrial zone, in the eastern of the Castellón Province (Spain), situated 46 meters above the sea level and between two fluvial basins. The inhabitants that can be affected in this area by the pollution are approximately 51,000. This area has a complex atmospheric environment. The climate dominant is Mediterranean with low rainfall (400mm per year, Vicente et al., 2011). The lack of the rainfall together with the few vegetation that covers the soils and the frequent high particulate Sahara air-mass intrusions (Rodríguez et al., 2002) contribute to increase the background of air pollutants concentrations in this area. In addition, it must be considered that a system of local breezes occurs in the study area. These periodic winds, land-sea type, have been extensively studied by several authors (Martín et al., 1991; Boix et al. 1995, and Millán et al., 2001). This system of breezes does that the concentrations of pollutants can be affected by emission sources located outside the Vila-real city.

As mentioned, this city is in the industrial province of Castellón. This area has one of the most important industrial clusters in the world, many ceramic tiles and ceramic frit manufactures are concentrated there. For this reason, this industrial zone is a strategic area in the framework of European Union (EU) pollution control.

The main natural sources of pollutants in this area are the resuspension of mineral materials from the surrounding mountains with few vegetation coverage (Gómez et al. 2004) and from the long-range transport of particles from North Africa (Rodríguez et al. 2001, Pérez et al. 2006). The influence of the dust intrusions from North Africa to ambient PM10 levels in the study area reaches around 2µg/m^3 on annual basis (Minguillón et al, 2009).

The anthropogenic pollution sources are the traffic and the industries. The main industrial activity is based on producing ceramic tiles (Vicente et al. 2007). In addition, at the East of Vila-real there are a power station, a refinery and several chemical industries (Boix et al., 2001). Finally, important sources of secondary PM include the precursor emissions of VOC's NO_x and SO_2 from the high temperature ceramic processes, power generation, petrochemistry and biomass combustion (Minguillón et al., 2007).

On the other hand, Monterrey is the third largest city in Mexico (Martínez et al., 2012), housing a high proportion of the industries in the country. It is located in the Northeast of the country, and is the capital city of the state of Nuevo León. The city is located in a semi desertic plain, with an average altitude of 540 meters, crossed by the Santa Catarina river and the mountain range of the Sierra Madre Oriental in the southern part, which acts as a geographic barrier for winds (Menchaca-Torre et al., 2015). Rains generally do not exceed 60 mm annually, with average temperature of 22,3ºC (INEGI, 2014). The name Monterrey stands for the metropolitan area of a city with about 2 million inhabitants in 4 municipalities: Monterrey, San Pedro Garza, Apodaca and San Nicolás de los Garza (INEGI, 2014). Monterrey has a semiarid climate BSh, according to García (1988).

During spring, Summer and Fall dominant winds blow from the northwest, with an average heading of 105 degrees and during winter months winds from the southeast prevail, with an average heading of 285 degrees. During winter sometimes cold winds from the north blow, with an average heading of 190 degrees (Ramírez Lara, 2007).

Monterrey has a variety of industrial complexes including production of glass, steel, cement and paper, among others. Due to the large demographic explosion, there are 1,8 million vehicles (Menchala-Torres et al, 2015). Despite the high industrial activity, the government has not given industries specific guidelines or regulations to promote the investment and industrial development without damaging the environment. On a report done by the Clean Air Institute on the particle pollution in Latin America, Monterrey is ranked the most polluted city in PM10 concentration with an annual 24 hr average of 85.9 $\mu g/m^3$ (INEGI 2014), above Mexico City, which has a 24-hour average of 57.0 $\mu g/m^3$. The Mexican Official Norm for PM10 concentration is 40 $\mu g/m^3$ as a 24-hour annual average. There are no documents reporting the proportion of PM10 from natural sources, but given the extent of the urban area, we may consider that almost all of the PM10 pollution in Monterrey comes from anthropogenic activities and sources (INECC-SEMARNAT, 2015). Research conducted by the Clean Air Institute on particle pollution in Latin America, declared Monterrey as the second Latin American city with most deaths by air pollution, after Santa Gertrudez, Brasil (Green and Sánchez, 2010).

In addition, air pollution has been increasing in Monterrey because of the lack of public policies to regulate the growth of industries in the neighbourhood of residential sectors. Also, the government has set no public policy for the reduction of carbon emissions from factories. The official Mexican norm of PM, exceed the international standard set by the

World Health Organization in particle pollution. Mexico's official 'safe levels' of ozone, PM10 and PM2.5 (Pollution concentration by area) are all significantly higher than the levels recommended by international environmental and health organizations, which allows the government to cheat by declaring that the pollution is not at dangerous levels (INE, 2003).

The study was also carried out in the Piemonte region, located in the North of Italy and more precisely in the western part of the Po valley. Although larger than the two previously described areas considered in this study, the Piemonte region is an area of interest due to its air pollution problems, mainly because it includes the largest industrial, trading and agricultural area with high population density in Italy. (Mélin and Zibori, 2005; Bigi, et al., 2012; Arvani et al., 2016)

The Po river basin is a critical area since it exceeds the annual and daily limit values fixed by the European Union for human health protection (see EU Council Directive 2008/50/EC) (Carnevale et al., 2008; Padoan et al., 2016). Consequently, the population is exposed to hazardous pollution levels. For this reason, researchers have a special interest in analysing concentrations of PM10 in this area to try to avoid multitude of harmful consequences, ranging from minor effects on the cardio-respiratory system to premature mortality (Samet et al., 2000; Samoli et al., 2008).

The Po river basin, located between the Alps and the Appeninesis is characterised by a complex orography, which determine a singular meteorology. The climate is not uniform throughout this area and shows significant temperature variations caused by its complex relief. This heterogeneity causes climate variability, especially in winter, when the mixing layer height is low and thermal inversion is frequent (Padoan et al., 2015). For example, it may happen to have weak winds and stagnation conditions that result in accumulation of pollutants in the central part of the region, at the same time, breezes and foehn winds prevail in the mountains and valleys (Mazzola et al., 2010). Therefore, lower PM10 concentration is usually observed near the Alps and higher pollution levels are detected in plains closer to urban areas (Pernigotti et al., 2012).

The Po plain is characterized by urbanized areas where the most important emission sources of primary PM10 and secondary precursor pollutants, such as industrial sites and main roads with high levels of traffic, are located. Even if, the 40% of the PM10 emissions are caused by automotive circulation (Shilirò et al., 2015), the number of manufacturing industries as well as the weather conditions, presumably aggravated by climate change, are also significantly contributing to higher levels of PM10 (Palatella, L., et al., 2010; Mercuri et al., 2012). In addition, anthropogenic activities like fossil-fuel usage and biomass burning as well as natural processes such as plant abrasion (processes caused by water, ice and wind) and secondary particle formation by atmospheric oxidation of biogenic precursors are also identified as important sources of the carbonaceous PM (Penner, 1995; Turpin and Lim, 2001; Bond et al., 2004). In particular, according to the emission inventory of the Lombardy Region the main sources of primary PM10 in the Alpine city of Sondrio are biomass burning (42%) and transport (36%) compared to 10% and 64% respectively of primary PM10 in the city of Milan (Belis et al., 2009). The concentrations of levoglucosan, a chemical marker for biomass burning (Simoneit et al., 1999), measured between 2005 and 2007 in 4 sites distributed across the Po Valley and the Alpine area strongly support the hypothesis of a higher contribution of this source to the PM in the Alpine valley floors.

3. METHODOLOGY

3.1. Sampling conditions

In the Vila-real area, two types of sampling stations had been established, one in a fixed point (EF), and another mobile (EMP). Figure 1 shows the location of the six sampling points in Vila-real. They were set up in accordance with the implementation guidelines of the European Council Directive 2008/50/EC. In this case, we used Jaume I University's equipment to do the sampling, while the location belongs to Vila-real local Council. The samplers used were PM10 medium volume, model INLD-LVSE, manufactured by Kleinfiltergerät. The sampling flow volume was 2.3m^3/h during 24-h periods. Particles were trapped on a permeable support, this being a 47mm diameter filter. The method used in order to know the concentrations of PM10 was gravimetric (UNE 12341). The sampling period was from 2001 to 2005.

In Monterey area, the sampling sites are part of a network made of two types of monitoring stations, operated by National Institute of Ecology and Climate Change since 1993. One type corresponds to fixed point monitoring stations (CE), and the other type corresponds to mobile (EMP). PM10 concentrations must follow norm NOM-0125-SSA1-1993, which establishes that a site follows the norm if its maximum daily average concentration is below 120 μg/m^3 and if the average of the daily averages is below 50 μg/m$^{3.}$ .Figure 2 shows the location of the sampling points in this area. PM10 medium volume samplers model BAM-1020, manufactured by Met One Instruments, were used. This device is considered as a reference according to US-EPA regulations for the continuous monitoring of PM10 particles. The BAM-1020 automatically measures and records airborne particulate concentration levels using the industry-proven principle of Beta Ray Attenuation. The data for the metropolitan area of Monterrey were correspond to PM10 measurements taken in the way described previously, form 2008 to 2013.

In the Piemonte region, the air quality-monitoring network is managed by ARPA (The Regional Agency for the Protection of the Environment) and it is composed of 20 fixed points of public property (FIXED), 3 private owned stations (FIXED) and one mobile unit (EMP). After analysing the information available in the database (http://www.r-inla.org/examples/case-studies/cameletti-et-al), we realized that five of monitoring sites included missing values and we decided to eliminated them focusing only on those with the whole information. Thus, for this study we have considered 18 fixed points and one mobile station. Figure 3 shows the location of the sampling points in this area. All fixed stations are connected via telephone lines to the central data acquisition and transmit hourly results of the measurements, allowing a constant control of the main factors that affect air quality. For the three areas considered in this study we have chosen the most centric monitoring site as the reference. Two types of measurement methods are used: beta (ß) and gravimetric instruments. PM10 and PM2.5 were measured continuously at the ARPA station at Lingotto, using devices that are checked daily. PM10was measured using a beta attenuation SM200instrument (Opsis, Furulund, Sweden) operating in mass mode (Tittarelli et al., 2008). The first one is characterized because it provides real-time data with short time resolution (<1h) that can be used for public information. In particular, the given average concentration of PM10 referred to a particular day of the year is available the day after. Nevertheless, due to its process of measurement (heated or unheated) some components can be lost and the final measurement may not be tight enough. The second method measures PM10 (reference method for PM10 specified in the Council Directive 2008/50/EC) using a set of appropriate filters and a subsequent gravimetric analysis. The sampling sites are located either in rural, urban or suburban areas and the emissions collected are classified among industrial, residential, commercial, natural or agricultural. For the Piemonte region the sampling periods were from 01/03/2006 to 31/03/2006 so it includes one month, March.

3.2. Statistical analysis

In this study, all the statistical data analysis aiming to compute the new Coefficient of Divergence and Redundancy (CODR or CODRcv) to obtain representative sampling points, have been made using the free *R* software (R Development Core Team, 2011).

As a first step, we explored the characteristics of the PM10 data, using univariate and bivariate exploratory data analysis techniques (Kara et al. 2007). This exploratory analysis allows detecting the main features reflected in the data sets. Exploratory techniques are distribution free in the statistical sense, so assumptions about normality are not needed. However, normality test will be needed in some posterior statistical analysis in order for them to be valid. The Bartlett (Stum et al. 1999) and the Shapiro goodness of fit tests (Shapiro and Wilk, 1965) have been proven to be useful tools for this task.

It is important to assess the differences between measurements of PM10 made at different monitoring sites and their variation in space. The comparison between the results at different sampling points was done using ANOVA. This step allows deciding whether there are statistically significant differences between the sampling points. The work of

Oliva and Espinosa (2007) is an example of this step. We used ANOVA for paired data for this part of our study (same time, same pollutant with same conditions).

After checking the normality assumption for the data, and in order to determine the existence of significant differences between sampling points the methodologies and coefficients used by others authors (summarized in review of Wilson et al., 2005) were applied. For instance, the correlation coefficient, the percentiles or the COD (formula 1).

$$COD = \sqrt{\frac{1}{p}\sum_{i=1}^{p}[(x_{ij} - x_{ik})/((x_{ij} + x_{ik})]^2} \quad (1)$$

Values observed for these coefficients showed that data may not provide enough information regarding the spatial variability of PM10 and that other covariates such as interurban distance should be included in the analysis. The introduction of covariates in statistical modelling, as it is shown in current works on spatiotemporal statistical (Porter et al. 2014), improves the quality of the predictions for future studies and helps in the decisions making selects fixed sampling points.

In this study, the Variance Inflation Factor (VIF here after) was used as a measure of the association between the different variables analysed. The VIF is defined as (O'brien, 2007)

$$VIF = \frac{1}{1 - R^2} \quad (2)$$

Where R^2 is Pearson's correlation

As a new step, through the study of this association, we show a new Coefficient of Divergence and Redundancy (CODR, 3) in order to determine representative sampling points.

$$CODR(d) = \sqrt{\frac{1}{p}\sum_{i=1}^{p}[(x_{ij} - x_{ik})/((x_{ij} + x_{ik})]^2} * dist \quad (3)$$

In addition, a second variant of this coefficient is presented in the equation 4, which includes Pearson's variation coefficient (cv)

$$CODR_{CV}(d) = \sqrt{\frac{1}{p}\sum_{i=1}^{p}[(x_{ij} - x_{ik})/((x_{ij} + x_{ik})]^2} * dist * \mathrm{CV} \quad (4)$$

In equations (3) and (4) x_{ij} and x_{ik} represent the 24 h average particulate concentration ($\mu g/m^3$) for sampling day *i* at sampling sites *j* and *k*, and *p* is the number of observations. The covariate included, the distance, is presented as *dist* (km). It referes to distance between a fixed sampling station and monitoring site and a candidate sampling location. The values of these new coefficients are in different rank from COD used for other authors (Wilson et al., 2005) because the covariate distance is included.

Criteria values of the new coefficient also depend on process variability, so that the coefficient of variation of the data are included in the definition. Different graphics of this coefficient with the variation coefficient of Pearson and distance will be needed in order to decide the final number of sampling points.

The data analysis flowchart, step by step using R software that we propose for this issue is in Figure 4. The flowchart proposed is a possibility for the researchers to work following steps when it is necessary to assess different pollutants and it is not clear what the procedure to follow is.

4. RESULTS AND DISCUSSION

4.1 Univariate analysis

Table 1 shows the summary statistics of different monitoring sites along the whole study period. In the case of Vila-real, the range of standard deviation is 12.98 – 20.67µg/m^3 for the fixed station and 8.72 – 16.67 µg/m^3 in the mobile stations. This is a first point of the differences between the stations and the moments of the data.

The range of the standard deviation is 22.3 - 40.5 µg/m^3 in the mobile stations and 24.8 µg/m^3 for the fixed station in the case of Monterrey. The differences between stations are shown up as well as in the case of Vila-real area.

In the case of Piemonte region, the range of the standard deviation is 24.8 - 37.2 µg/m^3 in the mobile stations and 30.8 µg/m^3 for the fixed station. As in the two previous cases, significant differences between the stations are observed in this initial assessment.

It is possible to see, the variability of the data through by checking the percentiles of the values at the monitoring sites. It means that if we compare the results between the 90% and 99% of the data (Em1, 39.5 for 90% and 74.2% for 99% for Vila-real or NO_2, 11.8

for 90% and 263.36 for 99% for Monterrey or EST3, 105.0 for 90% and 127.2 for 99% in the case of Piemonte Region) there is a significant difference (see supplementary data). Many authors use the percentile as the main variable in the decision to include or not new sampling stations in a given area (Wilson et al., 2005). In this study new coefficients for this purpose, which include new covariates, are presented because these data do not give us enough information nor a reliable criterion for the choice of the number of monitoring sites.

The next step in our research was to determine if the data values satisfy the normality assumption in order to choose the statistical methodology that could be applied. It is discussed starting from histograms in which a normal distribution (Gaussiana) is shown; therefore ANOVA methodology can be applied (see supplementary data).

The variably of the compared data between fixed and mobile stations is assessed through box-plot of paired sampling in the three study areas (see supplementary data).

4.2. Bivariate analysis

Having described the data by univariate pattern, without paired relationship, the statistic methodology is applied in order to know their correlation and if there are significant differences between the stations and, in addition, to reach a criteria or useful coefficient.

Firstly, the criteria used for other authors in the review of Wilson et al., 2005 is analysed in order to assess the own data with the ultimate goal of determining the number of representative stations in the study area (Table 2).

1) *Data correlation and relationship between stations*. A poor correlation is observed, far from the fixed station. There is a poor relationship between the values in the case of Vila-real. There is any value equal or superior to 0.9 then the inclusion of new stations is necessary. In the Monterrey area and Piemonte region, the correlation of the some values is close to 0.9 so some stations are redundant.

2) *Coefficient of Divergence (COD).* According to other authors a COD's values greater than 0.20 (Wilson et al., 2005) means that there are significant differences between the sampling points. Table 2 accomplishes this criterion in two areas of our study areas. Thus, apart from the fixed station, at least two sampling stations are needed in the case of Vila-real, six in the case of Monterrey and seventeen in Piemonte region.

3) *Difference between absolute concentrations.* The criterion of other authors is that there are significant differences in PM10 concentrations if there exist a difference greather than 10%, this is, if the proportion of the higher to lower concentration is above 1.10 . It is observed that the PM10 concentrations in all the sampling stations compared with the station with the lowest observed concentration were above 10% in the three study areas. Therefore according to this criterion all studied stations are necessary in the three cases.

The normality assumption has been tested by Shapiro test and Homogeneity of the variances by Ratio test and Barlett test. Then applying ANOVA paired of one factor can be analysed in order to see if the choice of sampling stations affects the results. In this step equality of means between sampling stations are assessed. In the same way, we can see with the t-Test the differences between fixed and moving stations. These results show the differences and the necessity of include all the stations. Given the different results obtained by different criteria, it is necessary to consider other covariate in order to know the number of sampling stations that are required. For instance, distance to the reference monitoring station, topography or climate related covariates.

4.3. New Coefficients

It was found from the univariate and bivariate description that there are significant differences between stations, so more sampling stations are needed, and it could be improved using covariates as distances between stations.

This first possibility gives us the information of relation between stations with a very simple formula (VIF formula). In order to understand VIF, we need to know the possibilities. If it is close to 1 there is a little correlation and if it is near to infinite there is not correlation. In many studies, the value 10 is the beginning of correlation (Marquardt, 1970). Table 2 shows the VIF values for the three study areas. In the case of Vila-real the VIF values are close to 1 and in the case of Monterrey and Piemonte region the values are not so close to 1 but in all cases are not very big, as no one is near 10.

In Figure 5 the values of the new coefficient CODR regarding the distance are presented for the three study areas. In this coefficient, distance has been included. The next step is to define the criterion that determines the number of sampling stations needed. We can see that a variability is presented and this is why it is necessary to introduce the variability in the coefficient. One possibility is the inclusion of Coefficient of Variability (CV), because it do not depend on the units we are using. It will be called CODRcv. It is noted that when distance increases, variability is lower and CODR coefficient increases.

In Figure 6 the variability of this new coefficient is shown. CODRcv is influenced by distance and standardised variation of the values. It is noted that CODRcv is influenced by the distance in the case of Vila-real. Likewise, Monterrey shows the same trend and there is relation between the variability, relationship and distance of stations. This gives us the idea that, considering the distance ,where we have to introduce the stations. In this case, the stabilisation of the data is when the distance is bigger than 2000 meters in the case of Vila-real. In Monterrey, the scale is different but the results are in the same way. In relation of Piemonte region, the change is around 80 meters.

The criteria for CODRcv could be the necessity of introducing the stations when the value is higher than 0.08 in the case of Vila-real and Monterrey, 10 in the case of Piemonte region. This is an important idea for introducing covariates, as the distance, in the station studies and so that, it could be an important criterion for the next studies. These studies should include spatial modelling using spatial varying covariates as well as construction of predictive maps. These studies can be made using modern spatial analysis techniques such as INLA (Rue et al., 2009).

5. CONCLUSIONS

A new statistical tool to estimate the number of representative sampling points in microenvironments is presented. Three study area are assessed, one in Spain (Vila-real) other in Mexico (Monterrey) and another in Piemonte region (Italy) with different environments.

An assessment of the coefficients used by other authors has been performed. The methodology used for estimating sampling points by other authors does not use the covariates (distances, meteorology…) and external elements (sources) that affect the concentrations of the pollutants. A new coefficient CODcv, formulated and developed by R software, in which the distance and variability is included is presented. This fact increases the quality of the comparison of the sampling points. As shown in Figure 6, different values of the CODRcv coefficient are observed depending on the analysed area. The stabilisation of the data is when the distance is bigger than 2000 meters in the case of Vila-real and Monterrey, and 80 meters in the case of Piemonte region. So, it is necessary to include the covariates in order to better characterized each study zone.

It is very important planning and optimizing the number of the sites because air quality monitors are expensive and/or because such monitors may not be placed anywhere. A well-designed sampling network also allows getting better estimates regarding the possible association between air pollution levels and the incidence of pollution-related diseases, as well to identify the location of possible pollution sources whose emissions are well beyond permissible levels. We have presented here a useful methodology to achieve those targets. By considering covariate information, the measures we propose allows to detect redundant sampling locations, minimizing the cost for obtaining the same amount of information.

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Table 1: Statistical data of stations.

Vila-real	Min	Max	1Q	Median	Mean	3Q	Var	Desv
Ef	3.0	101.0	34.0	45.0	45.3	55.3	292.6	17.1
Ef1	12.0	69.0	30.5	35.5	41.3	60.0	337.8	18.4
Ef2	24.0	79.0	39.0	47.0	50.3	65.0	266.4	16.3
Ef3	16.0	86.0	41.5	48.0	49.0	55.0	176.4	13.3
Ef4	3.0	101.0	32.0	42.5	45.2	63.0	427.4	20.7
Ef5	16.0	68.0	28.0	37.0	39.7	50.5	168.6	13.0
Em1	5.0	80.0	21.75	39.0	29.8	35.3	278.2	16.7
Em2	29.0	73.0	55.0	60.0	58.2	68.0	164.7	12.8
Em3	27.0	77.0	39.0	47.0	48.2	55.5	162.1	12.7
Em4	5.0	77.0	30.8	38.0	39.8	48.3	220.5	14.9
Em5	15.0	50.0	27.5	33.0	33.3	38.5	76.0	8.7
Monterrey								
CE	21.0	203.0	33.5	40.0	45.1	49.0	615.2	24.8
SE	25.0	193.0	41.5	50.0	59.6	69.0	852.0	29.2
NE	28.0	186.0	43.0	49.0	54.7	58.0	498.5	22.3
NO	31.0	207.0	49.5	64.0	66.6	76.5	692.1	26.3
SO	26.0	203.0	42.5	58.0	65.7	79.0	1025.4	32.0
NO2	42.0	268.0	65.5	79.0	86.5	95.0	1637.2	40.5
N	26.0	233.0	40.0	53.0	56.6	63.5	960.8	31.0
NE2	27.0	225.0	51.5	59.0	63.1	67.0	709.0	26.6
SE2	27.0	247.0	42.3	57.0	61.1	67.8	1092.8	33.1
Piemonte	**Min**	**Max**	**1Q**	**Median**	**Mean**	**3Q**	**Var**	**Desv**
FIXED	9.00	110.0	34.5	55.0	58.2	86.5	948.1	30.8
EST1	21.0	109.0	31.0	59.0	58.7	81.0	797.8	28.2
EST2	14.0	104.0	32.5	62.0	54.2	69.0	612.7	24.8
EST3	12.0	135.0	29.0	48.0	55.3	760.0	1084.6	32.9
EST4	14.0	105.0	24.5	37.0	44.8	64.0	627.7	25.1
EST5	11.0	110.0	28.0	47.0	51.7	67.5	759.1	27.6
EST6	6.0	143.0	35.0	63.0	64.1	91.5	1479.1	27.6
EST7	6.0	103.0	29.0	50.0	51.2	71.5	896.3	29.9
EST8	17.0	135.0	54.0	73.0	88.0	119.0	1295.1	36.0
EST9	19.0	116.0	39.0	55.0	60.1	84.5	808.2	28.4
EST10	8.0	97.0	32.0	49.0	50.0	68.5	650.3	25.5
EST11	13.0	121.0	40.0	47.0	61.0	86.5	1005.4	31.7
EST12	13.0	122.0	29.0	45.0	53.3	67.0	942.0	30.7
EST13	7.0	116.0	30.5	46.0	56.7	79.0	1110.1	33.3
EST14	9.0	109.0	35.5	52.0	54.0	79.5	847.6	29.1
EST15	15.0	103.0	33.0	50.0	53.0	72.0	650.7	25.5
EST16	10.0	140.0	41.0	65.0	68.4	94.50	1385.5	37.2
EST17	20.0	112.0	40.0	66.0	65.0	91.4	719.3	26.8
EST18	6.0	111.0	15.5	26.0	37.7	58.5	810.7	28.5

Table: 2 Coefficients relation between points.

Vila-real	Coef of correlation	COD	Absolute concentration	VIF
Ef1 – Em1	0.39	0.27	27.19 %	1.18
Ef2 – Em2	0.67	0.15	19.90 %	1.81
Ef3 – Em3	0.60	0.13	16.12 %	1.55
Ef4 - Em4	0.64	0.21	24.05 %	1.69
Ef5 - Em5	0.73	0.13	17.65 %	2.16
Monterrey				
CE-SE	0.54	0.21	24.93%	1.46
CE-NE	0.84	0.17	19.82%	3.35
CE-NO	0.86	0.23	31.84%	3.86
CE-SO	0.81	0.22	29.60%	2.92
CE-NO2	0.80	0.33	46.68%	2.78
CE-NE2	0.92	0.22	29.13%	6.15
CE-N	0.89	0.17	23.46%	4.88
CE-SE2	0.84	0.20	28.44%	3.39
Piemonte				
FIXED-EST1	0.92	0.16	17.63%	6.35
FIXED-EST2	0.75	0.22	28.11%	2.28
FIXED-EST3	0.85	0.21	24.74%	3.54
FIXED-EST4	0.79	0.26	30.57%	2.63
FIXED-EST5	0.74	0.33	26.70%	2.20
FIXED-EST6	0.84	0.26	27.31%	3.32
FIXED-EST7	0.89	0.24	24.63%	4.64
FIXED-EST8	0.92	0.25	32.00%	6.19
FIXED-EST9	0.85	0.21	21.62%	3.59
FIXED-EST10	0.74	0.28	29.91%	2.19
FIXED-EST11	0.76	0.23	27.36%	2.39
FIXED-EST12	0.81	0.23	26.85%	2.90
FIXED-EST13	0.84	0.26	30.32%	3.38
FIXED-EST14	0.78	0.26	26.63%	2.60
FIXED-EST15	0.83	0.21	22.33%	3.18
FIXED-EST16	0.87	0.24	26.22%	3.26
FIXED-EST17	0.87	0.21	19.90%	4.19
FIXED-EST18	0.73	0.37	43.09%	2.14

SUPPLEMENTARY DATA

1. Percentiles of the data

Many authors use the percentile as the main variable in the decision to include or not new sampling stations in an area. In the following tables the percentiles of the data (PM10,µg/m^3) are presented. These tables show the great difference between the results between 90% and 99%.

Table: Percentiles of the data. Vila.real.

	90%	95%	99%
Ef	68.0	74.6	84.0
Ef1	64.5	68.3	68.9
Ef2	72.4	76.6	78.5
Ef3	64.8	66.0	83.5
Ef4	72.0	78.8	91.8
Ef5	55.4	60.1	65.3
Em1	39.5	50.8	74.2
Em2	70.2	72.2	72.8
Em3	65.3	72.7	77.0
Em4	61.7	64.0	72.4
Em5	44.4	46.4	50.0

Table: Percentiles of the data. Monterrey

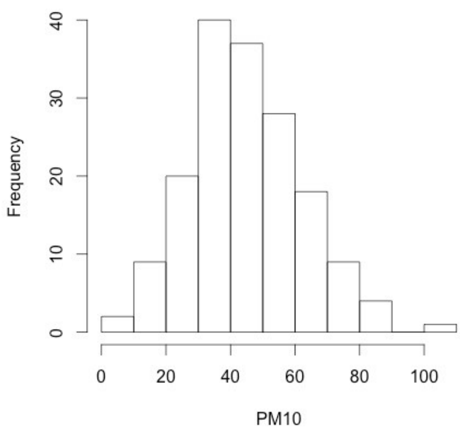
	90%	95%	99%
CE	57.80	71.20	140.36
SE	83.60	114.60	168.64
NE	73.40	80.10	136.12
NO	86.80	97.30	161.76
SO	101.40	117.90	162.98
NO2	111.80	144.70	263.36
N	72.40	88.40	175.67
NE2	74.80	91.60	165.26
SE2	82.90	115.90	180.88

Table: Percentiles of the data. Piemonte

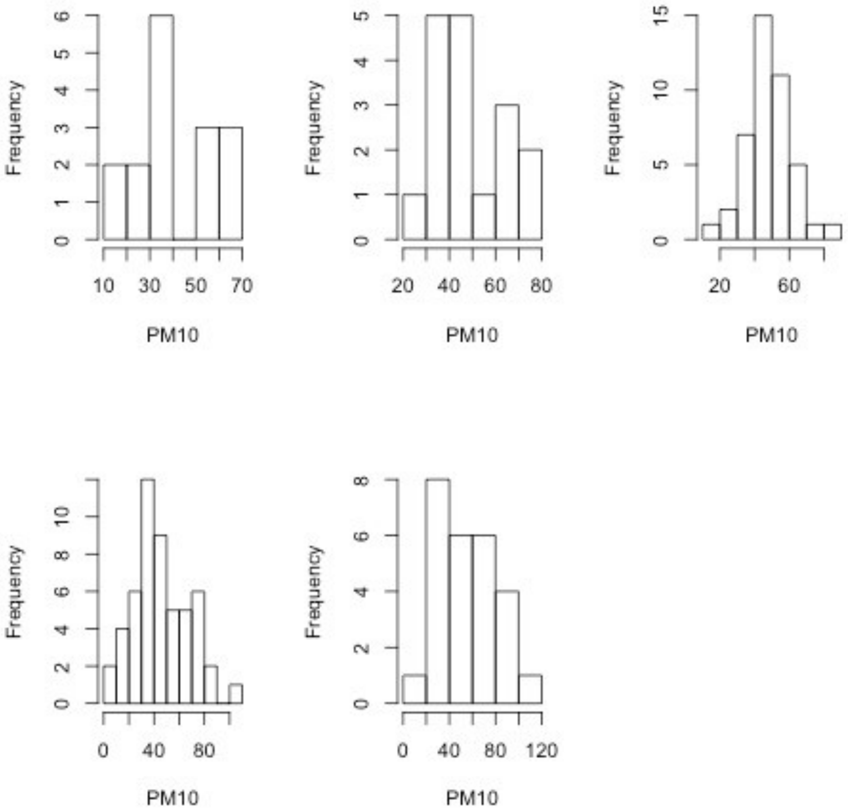
	90%	95%	99%
FIXED	100.0	102.5	108.2
EST1	96.0	99.5	106.3
EST2	83.0	90.0	99.8
EST3	105.0	108.5	127.2
EST4	81.0	86.5	99.9
EST5	93.0	98.5	107.3
EST6	118.0	126.5	138.5
EST7	99.0	100.5	102.5
EST8	129.0	130.0	133.5
EST9	98.0	105.0	113.6
EST10	82.0	89.0	96.1
EST11	105.0	116.5	120.4
EST12	102.0	107.5	118.7
EST13	110.0	113.5	115.4
EST14	93.0	101.0	106.9
EST15	86.0	90.0	100.0
EST16	124.0	129.0	137.6
EST17	97.0	99.0	108.7
EST18	83.0	86.5	104.1

2. Assessment of the Normality of the data

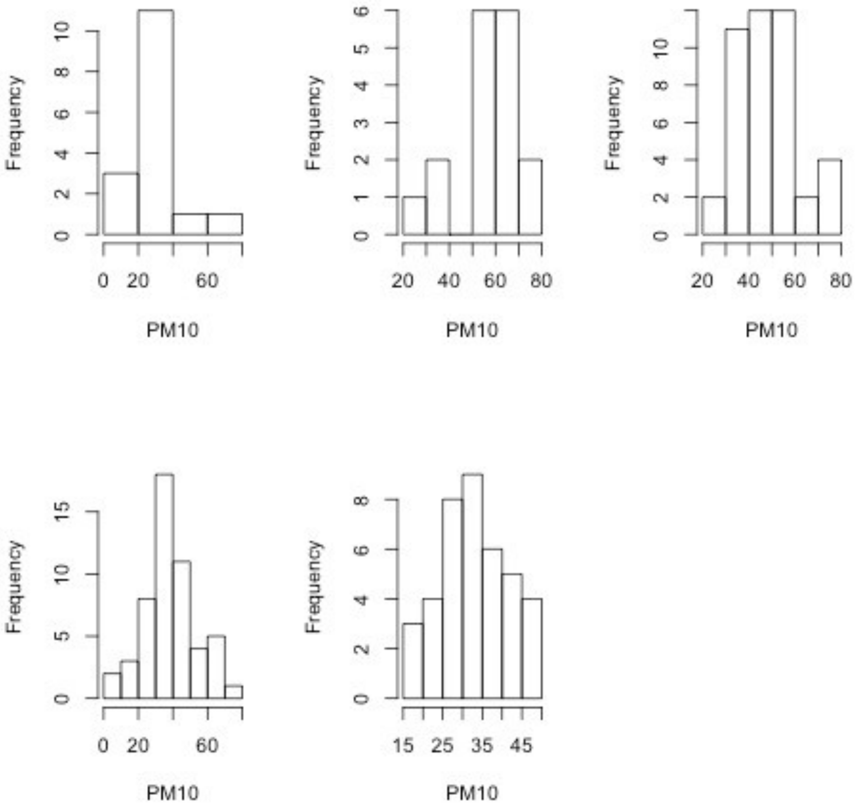
The study of the Normality of the data is very important in order to know what kind of statistical methodology can be applied. For this propose, the histograms of the PM10 of values are presented. In all the figures, a normal distribution (Gaussiana) can see, therefore ANOVA methodology can be applied.



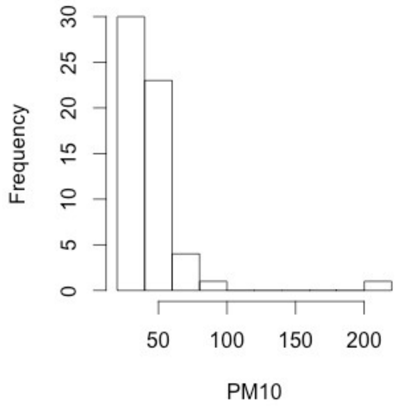
All PM10 values (μg/m^3) from fixed station. Vila-real



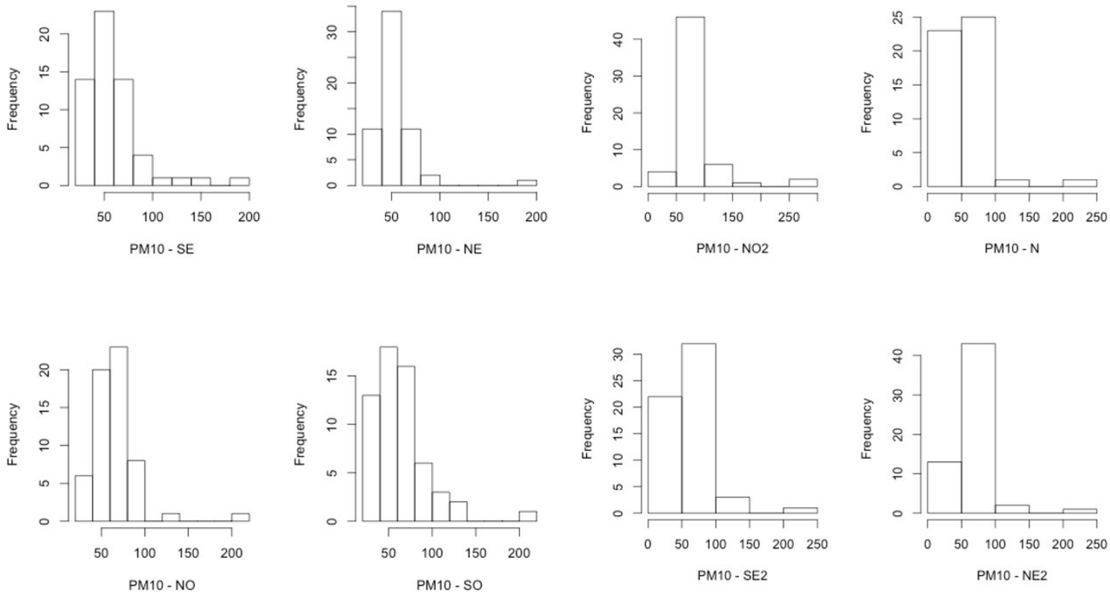
PM10 values (µg/m^3) from fixed station from Ef1 (topleft) to EF5 (bottonright). Vila-real.



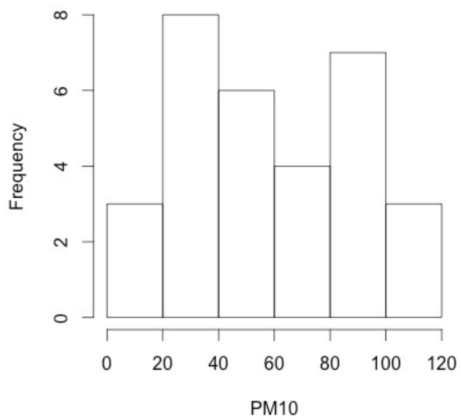
PM10 values (µg/m^3) of the five location of mobile station, from Em1 (topleft) to Em5 (bottonright). Vila-real



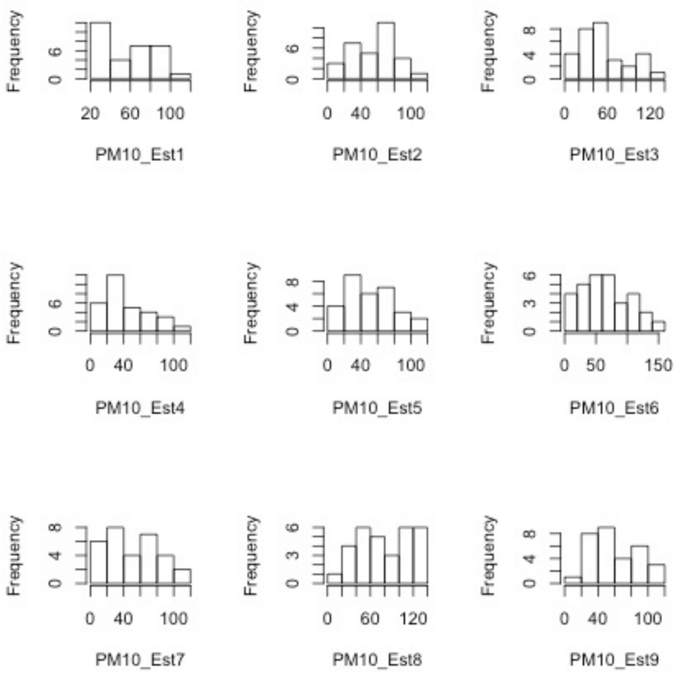
All PM10 values (µg/m^3) from fixed station. Monterrey.



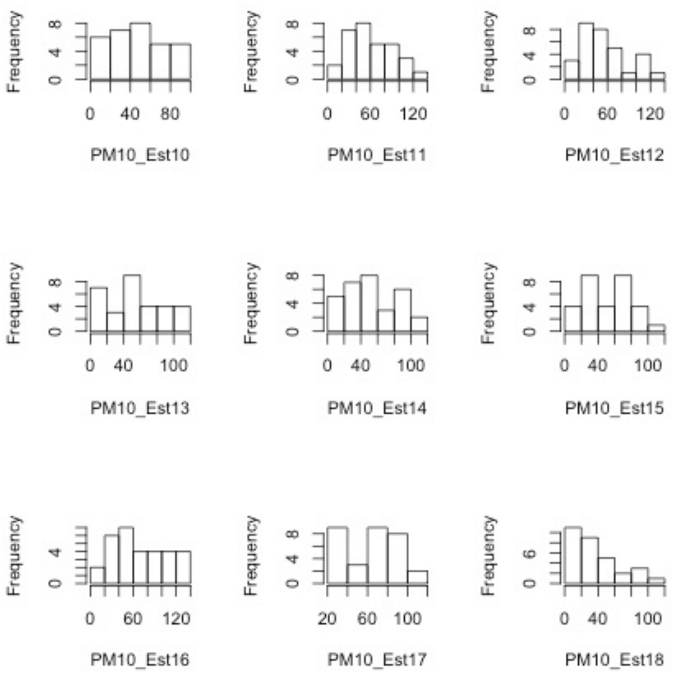
PM10 values (μg/m^3) of the eight location of mobile station. Monterrey.



All PM10 values (μg/m^3) from fixed station. Piemonte region.



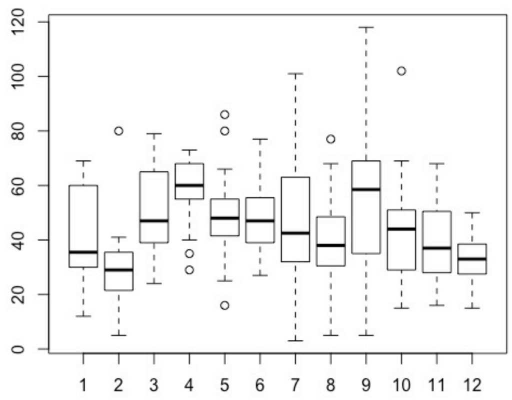
PM10 values (μg/m^3) of one to nine location of mobile station. Piemonte region.



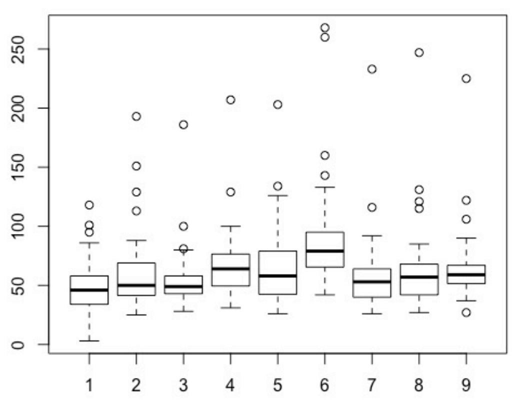
PM10 values (μg/m^3) of ten to eighteen location of mobile station. Piemonte region

__3. Assessment of the Variability of the data__

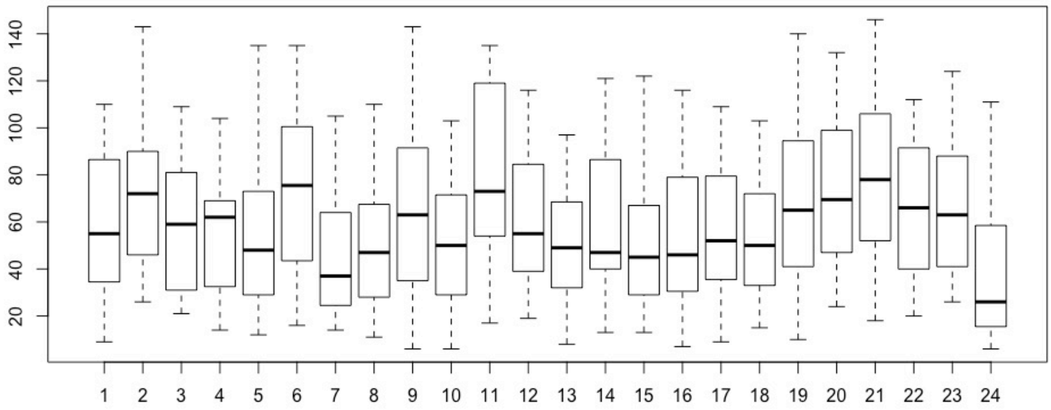
The variability of the data are assessed through the box-plot of paired sampling between fixed and mobiles stations.



Box-plot of paired data of fixed and mobile station (PM10, μ/m^3). Vila-real

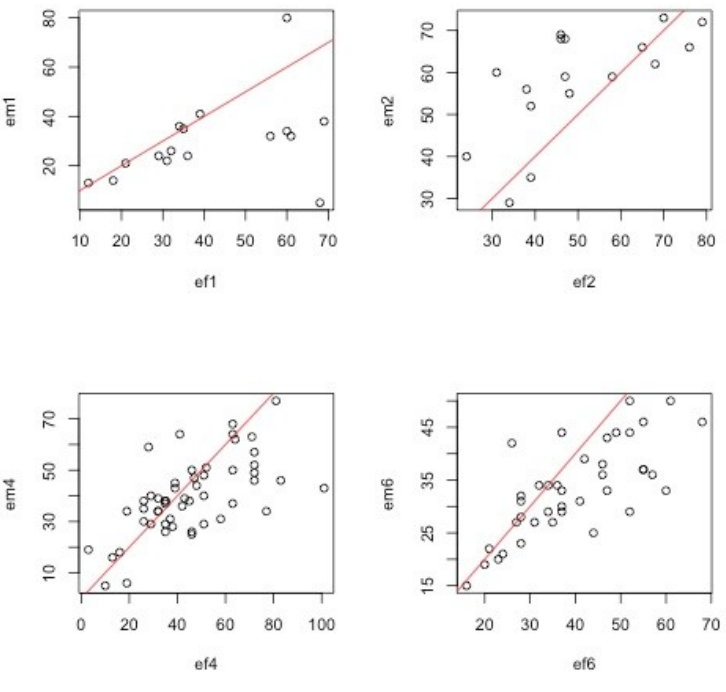


Box-plot of paired data of fixed and mobile station (PM10, μ/m^3). Monterrey

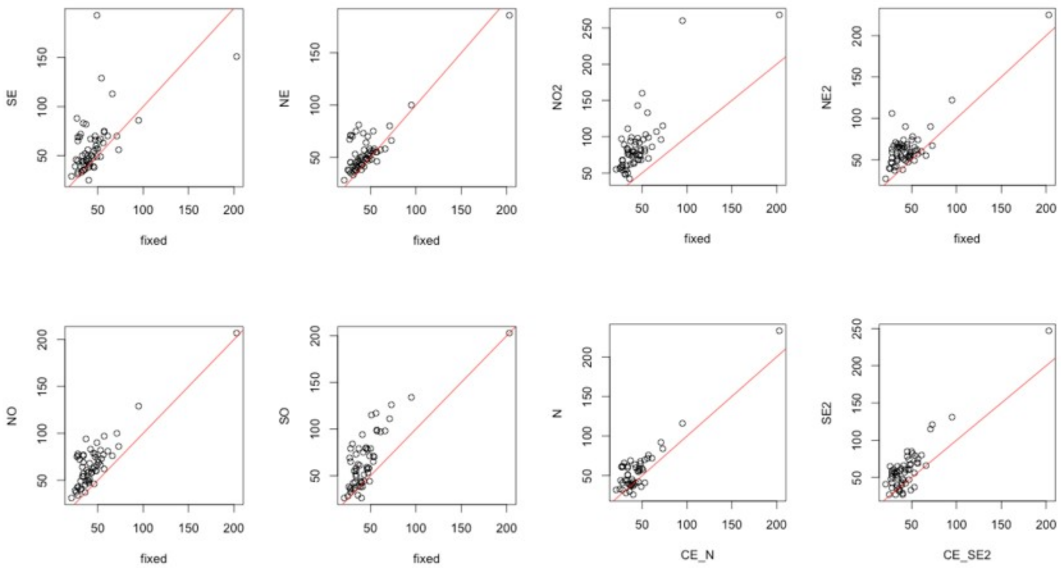


Box-plot of paired data of fixed and mobile station (PM10, μ/m^3). Piemonte region

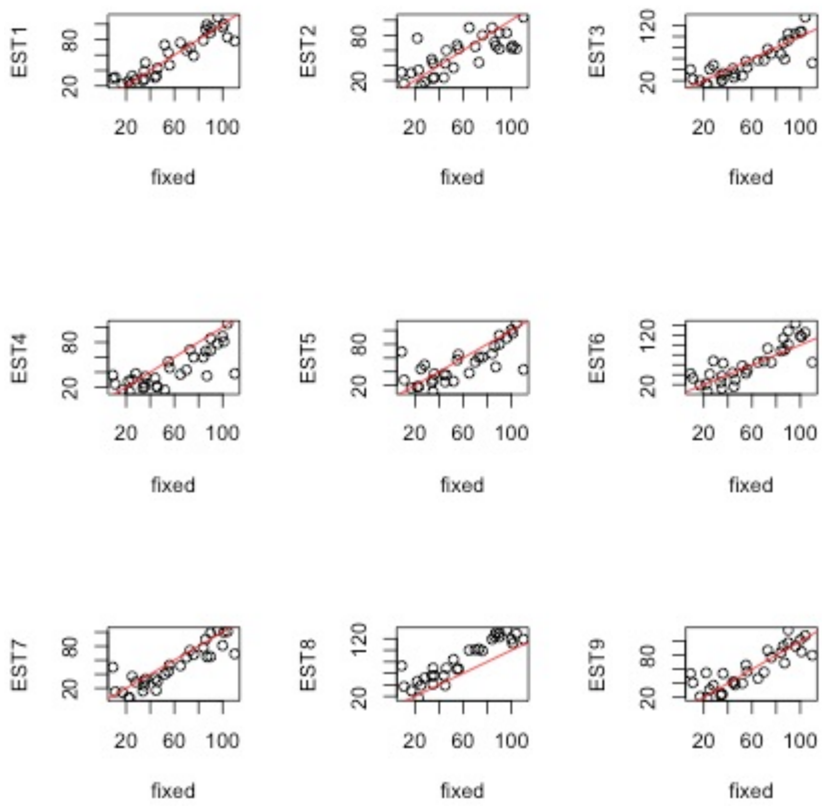
The relationship between the data of the fixed stations and the data of the different locations of the mobile stations are assessed through paired values figures'.



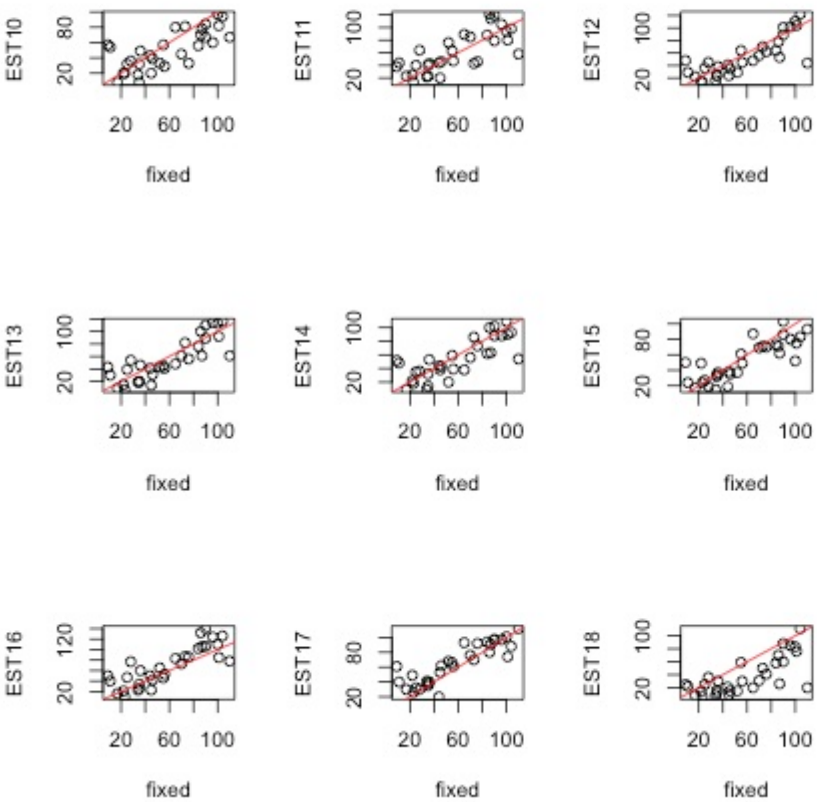
Paired values of PM10 (μg/m^3) of mobile station and fixed stations, Vila-real



Paired values of PM10 (μg/m^3) of mobile station and fixed stations, Monterrey



Paired values of PM10 (μg/m^3) of mobile station (1-9) and fixed stations, Piemonte region



Paired values of PM10 (μg/m^3) of mobile station (10-18) and fixed stations, Piemonte region

FIGURE CAPTIONS

Figure 1: Location map of monitoring sites in Vila-real, in the province of Castellón, Spain. Fixed station (EF), mobile stations (EM1-EM5). All the monitoring stations are located in a urban area.

Figure 2: Location map of monitoring sites, Monterrey, in the north eastern state of Nuevo León, México. Station CE was used as the reference monitoring station.

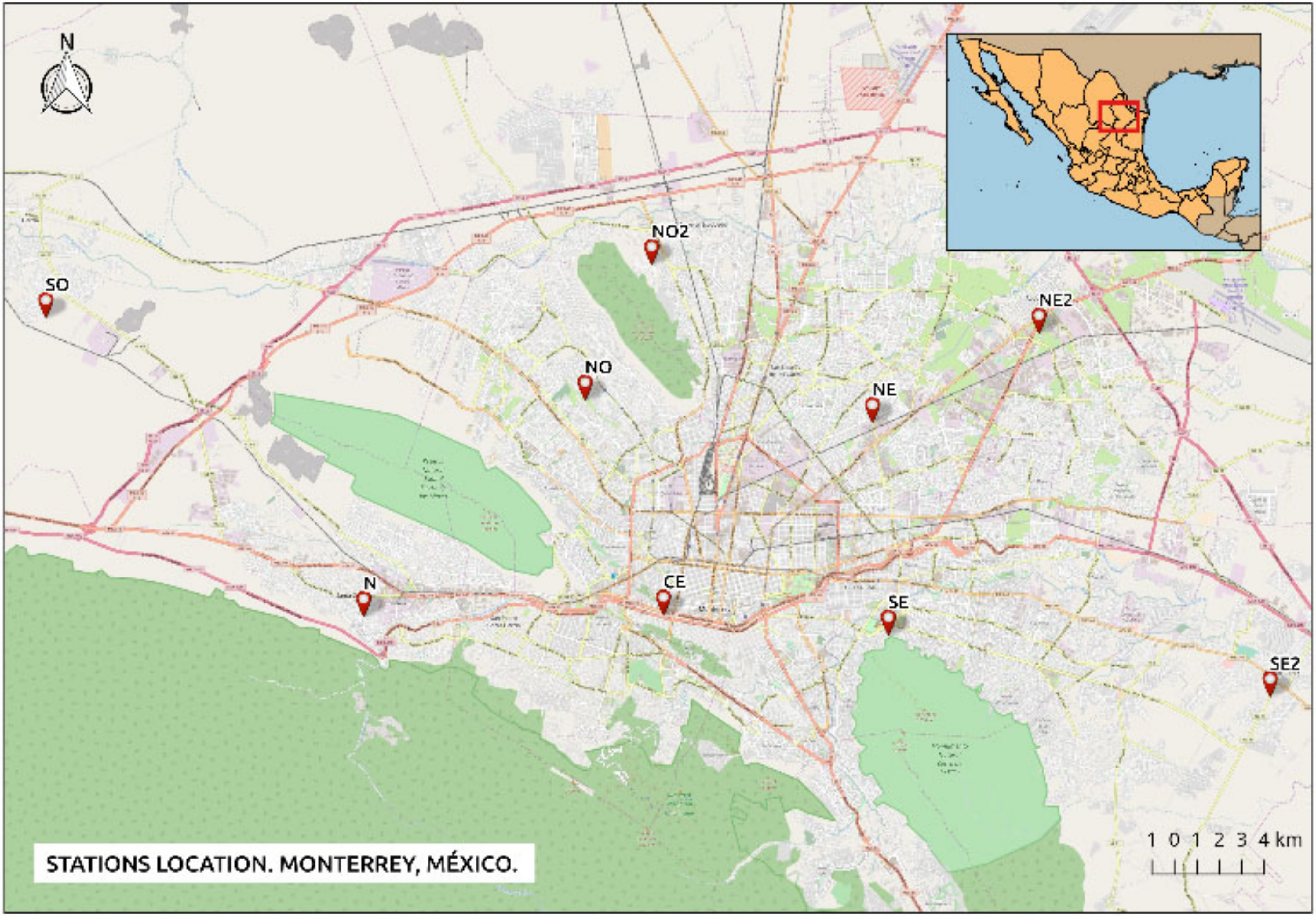
Figure 3: Location map of monitoring sites, in the Piedmont Region of Italy. The reference monitoring station was station 4.

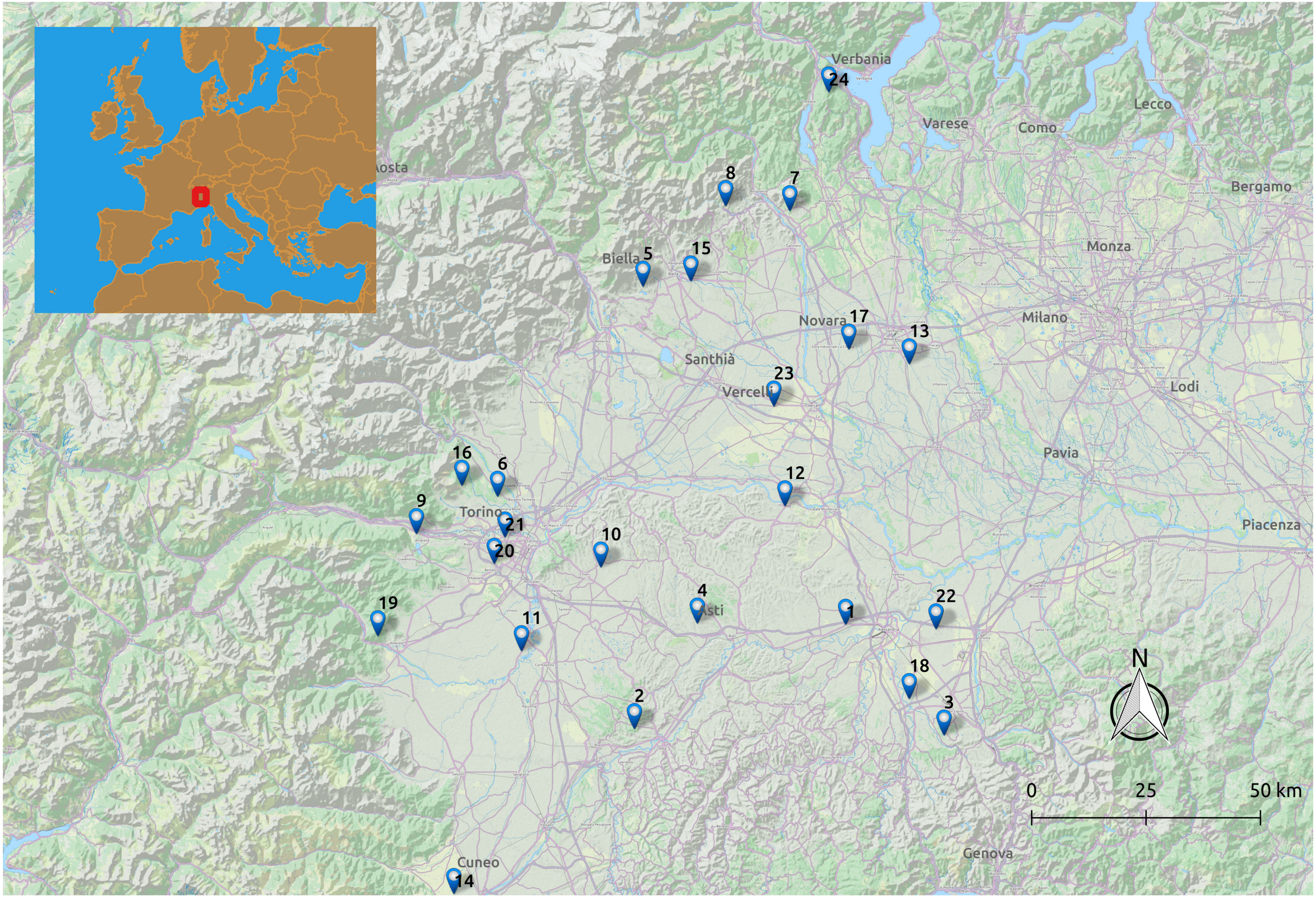
Figure 4: Data analysis flowchart.

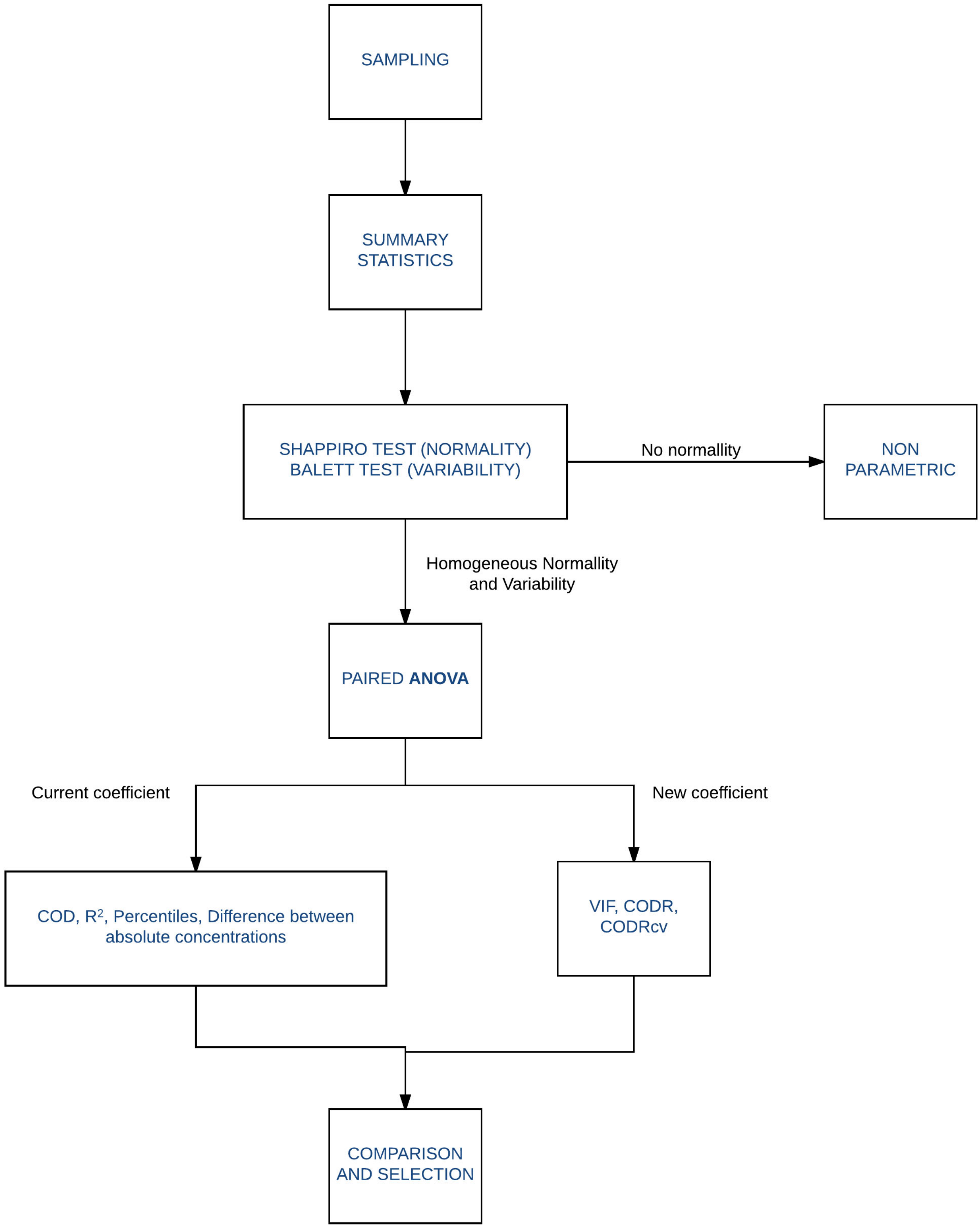
Figure 5: Coefficient of Divergence and Redundancy (CODR) versus distance (m) to the reference monitoring station for the three regions considered in the study

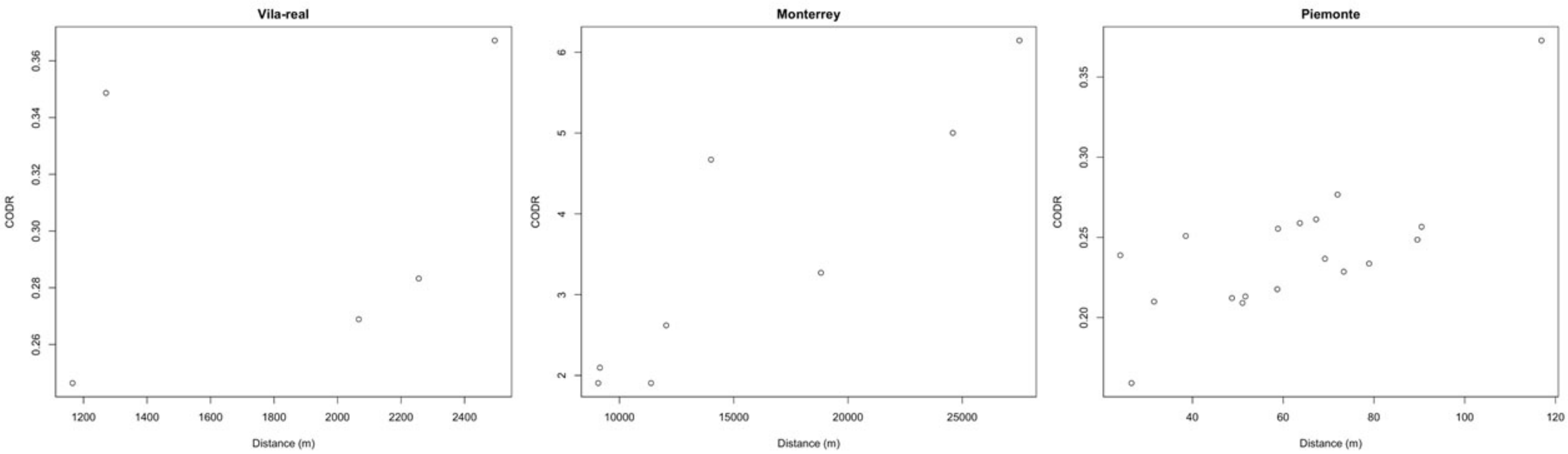
Figure 6: Coefficient of Divergence and Redundancy with coefficient of variation (CODRcv) versus distance (m) to the reference monitoring station for the three regions considered in the study.

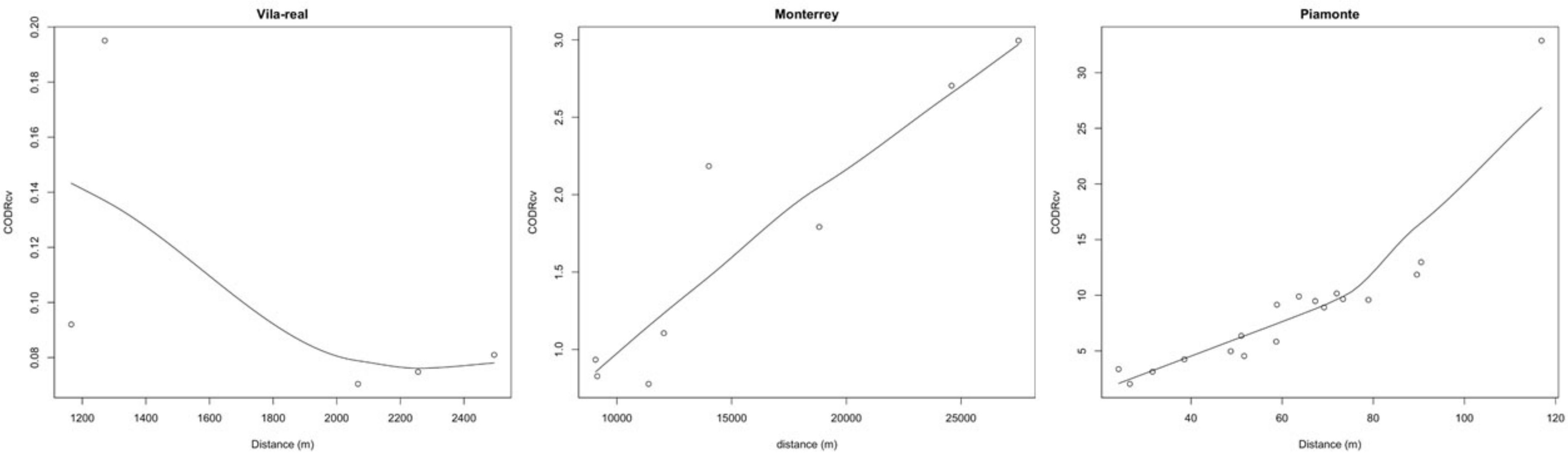

STATIONS LOCATION
VILA-REAL, SPAIN
EMP1
EMP2
EMP3
EMP4
EMP5
EF
N
250 0 250 500 m


N
SO
NO2
NO
NE2
NE
N
CE
SE
SE2
1 0 1 2 3 4 km
STATIONS LOCATION. MONTERREY, MÉXICO.


Verbania
24
Lecco
Varese
Como
Aosta
Bergamo
8
7
Monza
Biella
5
15
Novara
17
13
Milano
Santhià
23
Vercelli
Lodi
16
6
Pavia
12
9
Torino
21
10
20
Piacenza
4
Asti
22
1
19
11
N
18
2
3
0
25
50 km
Genova
Cuneo
14


SAMPLING
SUMMARY STATISTICS
SHAPPIRO TEST (NORMALITY)
BALETT TEST (VARIABILITY)
No normallity
NON PARAMETRIC
Homogeneous Normallity and Variability
PAIRED ANOVA
Current coefficient
New coefficient
COD, R2, Percentiles, Difference between absolute concentrations
VIF, CODR, CODRcv
COMPARISON AND SELECTION


Vila-real
CODR
0.26
0.28
0.30
0.32
0.34
0.36
1200
1400
1600
1800
2000
2200
2400
Distance (m)
Monterrey
CODR
2
3
4
5
6
10000
15000
20000
25000
Distance (m)
Piemonte
CODR
0.20
0.25
0.30
0.35
40
60
80
100
120
Distance (m)


Vila-real
CODRcv
0.08
0.10
0.12
0.14
0.16
0.18
0.20
1200
1400
1600
1800
2000
2200
2400
Distance (m)
Monterrey
CODRcv
1.0
1.5
2.0
2.5
3.0
10000
15000
20000
25000
distance (m)
Piamonte
CODRcv
5
10
15
20
25
30
40
60
80
100
120
Distance (m)