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MEASUREMENTS OF ATMOSPHERIC AEROSOL IN THE SALENTUM PENINSULA AND ITS CORRELATION WITH LOCAL METEOROLOGY

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Abstract

In this paper the results of measurements of Total Suspended Particles (TSP) and PM10 are presented. The samplings were carried out with a mobile laboratory in five locations on the Salentum peninsula situated in the southeastern part of Italy in Puglia. Measurements were taken discontinuously during the period 2002-2004. Some results of PM2.5 measurements taken only in 2004 are also presented. Up to now no systematic analyses of aerosol concentration in the Salentum peninsula have been presented in the scientific literature. This study is therefore a useful basis for assessing the local situation and for planning future monitoring and investigations. Measurements were performed using a standard European PM10 inlet (CEN-EN12341, 1998) and successive gravimetric detection of aerosol deposited over 24 hours periods on filters. The measurement sites can be considered representative of urban background for all the cases investigated. Results show concentrations in good agreement with lognormal distributions, indicating that the PM10 fraction is about 69% of TSP and PM2.5 is about 81% of PM10, which allows us to evaluate that the fraction of PM2.5 is about 56% of TSP. Concentration levels were correlated with local meteorological parameters, such as wind velocity, wind direction and precipitations. Results indicate that there is, on average, a substantial decrease of concentration levels in high wind conditions and on rainy days. They also suggest the possibility of a significant contribution

of African Dust to PM10 and TSP, especially in the summer season, which could be responsible for some periods of concentrations above the threshold imposed by the European legislation on PM10.

Keywords: PM2.5; PM10; TSP; Sahara Dust; Aerosol Concentration.

Introduction

The Salentum Peninsula is downwind of the Brindisi and Taranto industrial areas for most of the year. Brindisi is the site of a very large petroleum refinery and two oil combustion power plants, while Taranto hosts metallurgical industries with stainless steel production. Both areas have been classified by WHO as risk areas both for the population and the environment (Martuzzi et al, 2000). The meteorological conditions over Salentum are characterized by winds coming mainly from the northern sectors, which might expose it to industrial emissions situated to the NW (Taranto) and NNW (Brindisi). Such conditions and their implications for air quality is of great concern to the local administration (mainly the provincial government of Lecce). Moreover, a research study involving dispersion models (Mangia et al, 2000) has shown the probability, in the middle of the peninsula, of fallout of the pollution emitted by the power plants of Brindisi.

The local air quality control network consists of monitoring stations capable of measuring the standard gas indicators (as required by National and European legislations) and only the total suspended particulate (TSP) for the aerosol phase. In recent years, the provincial government of Lecce has been running, on its own, two monitoring stations also equipped with continuous PM10 and PM2.5 sensors, although the stations are still under calibration and maintenance conditions. As a general consequence, very little data on particulate matter (and specifically PM10) concentrations are available for the area. Several field campaigns have therefore been performed using a mobile laboratory, with the aim of monitoring the particulate matter concentration levels (TSP and PM10). The present work concerns the data obtained during the measurement campaigns carried out from January 2002 to December 2004 on urban background sites. Average values of PTS and PM10 concentrations, together with their statistical distributions, will be shown. The average value of PM2.5 concentration, even if with limited statistics, is also given. Correlation of aerosol concentrations with wind velocity and direction, at different heights above the ground, will also be analyzed. Results indicate, a decrease in both concentration peaks and average values for high wind velocities, confirming the efficacy of horizontal mixing in reducing aerosol concentrations. Also indicated is

the presence of a few days with peaks in PM10 concentrations above the limit of 50 μ g/m^3, set in the standards for PM10 mass concentration in the European legislation (Directive 1999/30/EC). Our analysis showed that several such cases occurred during Sahara Dust events.

It should be highlighted that there is a general lack in the scientific literature on aerosol concentration data in the Salentum Peninsula, and the present work is a preliminary assessment of the aerosol concentration levels which could be useful for planning the strategies for future monitoring.

Description of measurement sites and equipment

The measurements were performed using the mobile laboratory shown in figure 1. The laboratory is equipped with a BTX analyzer (Syntech Spectras GC855), a sequential PM10 sampler operating at the constant flow-rate of 2.3 m^3/h (Thermo ESM Andersen FQ95SEQ) with the inlet at 2.5m above the ground, capable of 16 days of continuous samplings, and a sequential TSP sampler (Zambelli, Explorer V53M) capable of 8 days of continuous monitoring when operated over 24h averages. The mobile laboratory is also equipped with a telescopic mast (10 m tall) that was sometimes used as a base for a micrometeorological station based on a Gill R3 ultrasonic anemometer and a Rotronic termo-igrometer directly operated by the anemometer itself. In this way, it was possible to obtain detailed information on local meteorology, allowing the correlation of aerosol concentration levels with local meteorological conditions. During the field campaigns some preliminary PM2.5 data were gathered by means of a manual sampling line according to the CEN TC 264/Working Group 15 indications. The line is equipped with a standard inlet (Zambelli) and is run by a vacuum pump (Tecora, Bravo H-Plus) operating at a constant flow-rate of 2.3 m^3/h.

The sampling sites were the University of Lecce Campus, hereinafter referred to as Ecotekne, (where the ISAC-CNR Research Institute is located), and the towns of Galatina, Maglie, Campi Salentina and Spongano. The locations, indicated in figure 2, all lie along the middle line of the peninsula. The measurement locations were chosen in order to be representative of the urban background, so that measurements were taken far away from the centre of the towns and far from roads with heavy traffic.

The most widely used filter supports were made of cellulose nitrate with 0.8 μm porosity (Millipore), although in some limited cases PVC (polyvynilcloruro) filters were employed, and, starting from the beginning of 2004, quartz fibers (Sartorius). The gravimetric assessment was performed by means of an analytical balance with 10 μg sensitivity (Sartorius). During each measurement campaign (10 days long on the average) some blank filters were used as a check of measurement quality. Experimental errors on gravimetric measurements of PM can arise from several independent sources, most notably moisture absorption, electrostatic effects, filter damage during handling and imprecision in weighing due to the balance used. A method of estimating the measurement accuracy is to use a statistical analysis of blank filters (Paik & Vincent, 2002). The procedure makes use of a certain number of blank filters which go through the same procedure as the exposed filters. In particular, blank filters follow the same conditioning, handling and weighing procedure as those employed for measurements, including expositions to the same environment and for the same time periods, the only difference being that the airflow through the blank filters is zero so that no particles are collected on them. The blank filters are weighed before and after exposition to the environment to determine the difference in mass P. In our experiments P was determined, for each filter, as the average of three successive and independent measurements. The variability of P furnished information on the experimental systematic and random uncertainty. The systematic uncertainty is largely due to differences in the relative humidity of the environment between the first weighing (before exposition) and the second one (after the exposition). It could be corrected, if necessary, using a certain number N_b of blanks filters for a specific measurement campaign. After correction for the systematic part of the uncertainty, it is possible to estimate the random part using the standard deviation (Paik & Vincent, 2002) as:

$$\hat{\sigma}_P = \sigma_P \left(1 + \frac{1}{N_b}\right)^{0.5} \qquad (1)$$

where σ_P is the standard deviation of the mass difference P of the blank filters. We performed the analysis of two sets of blank filters, the first referring to cellulose nitrate filters and the second to quartz fibre filters. The results indicate a value of σ_P equal to 41.3 μ g for the first set of 9 blanks and 26.6 μ g for the quartz fibre filters. The values of σ_P are smaller for the quartz fibre filters, reflecting their lower sensitivity to

environment humidity. Supposing that 3 blank filters are used in a measurement campaign, the uncertainty on mass-detection, in terms of one standard deviation, can be considered equal to $\hat{\sigma}_P = 47.6 \mu g$ for cellulose nitrate filters, and 30.8 μ g for quartz filters. The Limit Of Detection (LOD) is usually evaluated as 3 times $\hat{\sigma}_P$ and the Limit Of Quantification (LOQ) is generally defined as 10 times $\hat{\sigma}_P$. Therefore, in terms of concentration the values are: LOD=2.6 μ g/m^3 and LOQ=8.6 μ g/m^3 for the cellulose nitrate filters and LOD=1.7 μ g/m^3 and LOQ=5.6 μ g/m^3 for the quartz filters.

3. Discussion of results

3.1 Average concentration

All the measurements sites have similar characteristics, permitting the employment of all measured daily concentrations in evaluating average concentrations values of atmospheric aerosol. This means that 99 measurement days are available for PM10, 92 days for TSP, but only 24 days for PM2.5, all of them concentrated in the year 2004. Table 1 shows the arithmetic means and the standard deviations of PM10, TSP and PM2.5 measurements. The results indicate an average value of PM10 of 27 μ g/m^3, an average concentration of TSP equal to 46 μ g/m^3, and a PM2.5 concentration of 22 μ g/m^3. Table 1 also shows the averages disaggregated by years (2002, 2003 and 2004), together with their standard deviations and the number of available samples. It should be borne in mind that the 2002 statistics for PM10 are limited, plausibly explaining the relatively large average value. Table 2 shows the effects of the two main yearly seasons: the cold season, considered from October to March, and the hot one from April to September. The comparisons show a higher PM10 concentration during hot months with respect to the cold ones. This is the opposite of what occurs in the north of Italy, where meteorological conditions and domestic heating systems combine to give rise to generally higher particulate concentration in winter months (Marcazzan et al 2001). In the South of Italy, apart from the aforesaid difference, the opposite trend might be explained by the contribution of African Dust to aerosol concentration (see below). There is no PM10 data available in the scientific literature measured in the Salentum peninsula, however Bruno et al (2002) report an analysis of

atmospheric aerosol in Bari, which is the largest city in the Puglia region, located about 150km from Lecce. In a residential area with light traffic PM10 concentrations are found to range from 15 μ g/m^3 to 60 μ g/m^3.

The ratio between concentration levels in the different aerosol fractions is usually more comparable among the results obtained at different measurement sites. The average values reported in Table 1 cannot be used to evaluate the different ratios, because some of the measurements are not simultaneous. In our results, considering only the 47 simultaneous measurements of daily concentrations of PM10 and TSP, the PM10/PTS ratio was found to be 0.68. The PM2.5/PM10 ratio was 0.81. This allows us to infer the PM2.5/TSP ratio to be equal to 0.55. The value of PM10/TSP is in good agreement with the values reported in Querol et al (2001), for the city of Barcelona, for which a ratio of 0.65 is reported. In Berico et al (1997), for the town of Bologna, the PM10/TSP ratio is about 0.8. The PM2.5/PM10 ratio evaluated with our results is slightly larger than the values reported for Milan (Marcazzan et al 2001; Marcazzan et al (2002) where an average of 0.7 was reported. It should be noted that Marcazzan et al (2001; 2002) found seasonal differences in PM2.5/PM10 values, ranging from 0.63 in summer to 0.73 in winter, which showed a daily ratio as high as 0.9. The PM10/TSP ratio is also influenced by local meteorology, and could change at high wind speed (Marcazzan et al 2001, 2002). In Querol et al (2001) values of PM2.5/PM10 ranging from 0.6 to 0.85 are reported, with an average around 0.7. In the highly populated Mexico City values of the average PM2.5/PM10 ratio were found to vary from 0.44 to 0.60, according to different measurement locations (Vega et al 2002).

A comparison of average concentration values during rainy days (days on with at least 1mm of precipitation in the 24 hours) with concentration levels on non-rainy days shows that the effect of precipitations is to reduce the concentration values by about 35% for PM10 and 30% for TSP.

3.2 Statistical distribution

The sampled concentrations could be considered as a statistical sample, coming from the population made of all measurement days in the overall period of research. Since the samples could be considered representative of the population, a statistical comparison of the distribution of daily concentrations with theoretical

distributions was performed. The method followed was the same as the one described in Hadley et al (2003) for SO_2 concentrations. Two statistical distribution models were considered: normal distribution and the lognormal one. The correlation coefficient R^2 is better for the lognormal distribution for both PM10 and TSP, as is expected in general for pollutant concentrations (Ott, 1990). Figure 5 reports the distributions of PM10 and TSP (marks), together with the estimated lognormal distribution (continuous line). The abscissa is constructed from the order statistic medians or normal score, Z(p), calculated from the inverse of the cumulative distribution function of the standard normal distribution.

The agreement is reasonable with R^2=0.973 for PM10 and R^2=0.979 for TSP. For PM10, the calculated parameters of the distributions are the geometric mean concentration (23 $\mu g/m^3$) and the geometric standard deviation (1.7 $\mu g/m^3$). For TSP, the geometric mean concentration is 42 $\mu g/m^3$ and the geometric standard deviation is 1.5 $\mu g/m^3$.

Using the parameters of the obtained lognormal distribution, is it possible to estimate the probability of a PM10 concentration above the legislation threshold of 50 $\mu g/m^3$. If the calculated probability is referred to a complete year (365 days of data), it is estimated that there would be 27 days with values above this limit. This must be compared with the PM10 standards in the legislation, which imposes 50 $\mu g/m^3$ as a limit not to be exceeded for more than 35 days per year. Such results are valid for measurement locations similar to those investigated in this work, i.e. urban background sites, although the situations could be different for hotspots situated in major areas of urban traffic.

3.3 Correlation with meteorological data

Measured concentrations at the different sites were correlated to the wind directions and velocities measured at the level of 958 hPa. The altitude corresponding to 958 hPa is roughly 600 m, and can be associated with the plume rise of industrial emissions both from the Taranto and Brindisi areas. Wind data are obtained from radiosoundings in Brindisi, located about 35 km from Lecce. Results for PM10 are reported in figure 4 and for TSP in figure 5: it can be seen that most of the sampled concentrations are associated with winds blowing from the northern and southwestern sectors.

Figure 4(b) shows the correlation between PM10 concentration and wind velocity, considered as the arithmetic means of wind intensities without considering their directions. Clearly evident is a correlation with the wind intensity, which might be explained by an increase in the dilution due to a more effective mixing at high wind velocities. There are also some events that appear out of this trend, a possible indication of Sahara Dust episodes. In particular, a clear correlation can be seen for the data taken in Lecce, the ones for Maglie and Galatina are less evident. At the latter sites, transport from local industrial sources could explain the difference in the pattern. As matter of fact, if there are industrial plumes at relatively low altitudes, the ground level concentrations could increases at high wind speed when the plumes are advected directly towards the measuring instruments. Such an increase is particularly evident in figure 6, where the correlation between PM10 concentrations and local wind direction and speed measured at 10m above the ground is reported for all the data which include this meteorological information. The available data were analysed by separating the cases with local wind velocity U (at 10 m above the ground) lower than 3 m/s from the cases with U higher than 3 m/s. The results indicate that concentrations of TSP decrease, on average, by 24% for U>3m/s and PM10 decreases, on average, by 31%. A similar behaviour for PM10 was found in Marcazzan et al (2001) for the case of Milan.

3.4 Saharan dust events

Aeolian dust coming from Africa has been measured for several years at different European stations, including the Monte Cimone CNR station (Bonasoni et al, 2002) and the Jungfraujoch site (see for example Schwikowski et al, 1995). Frequent cases of Saharian Dust have also been identified in the area of Lecce (Blanco et al 2003). Such episodes may have an important impact on detected aerosol concentration levels. For example, a recent research in Spain has indicated the potential effect of such dust in the difficulties of meeting the new standards concerning PM10 in Europe (Rodriguez et al, 2000). In Italy too, Saharan dust might have an important potential impact, at least in the southern and central regions of the country, especially at rural or urban background stations. Our data highlight five cases where PM10 concentrations exceed the legislation limit (50 μ g/m^3 daily average), details of whichs are reported in Table 3, classified as either Sahara Dust events (SD) and Non Sahara Dust events (NSD). Saharan dust was identified by the

lidar station at the Lecce University (De Tomasi & Perrone, 2003), as well as the TOMS satellite aerosol optical index, combined with information provided by from meteorological simulations at high altitude. Basically, 3 days out of 5 above 50 μ g/m^3 are identified as SD events, which are quite frequent in the Salentum peninsula, especially in the summer period (Blanco et al 2003). The SD days between 9/5/2003 and 10/5/2003 are probably a single episode of long range transport of African dust over the Italy. The episode is mainly a transport of aerosol in the central part of Italy that extends down as far as the Salentum peninsula. The results indicate that the PM10/TSP ratio during SD events is essentially equal to the average ratio evaluated over the entire measurement period. During the SD event in July 2002, the monitoring station of the town of Lecce was operating, and the PM2.5/TSP ratio decreased to 0.24. It could be argued that most of the particles contained in Saharan dust are larger than 2.5 μ m (coarse fraction). Therefore a simultaneous measurement of PM10 and PM2.5 could give a discrimination between anthropogenic particulate and natural sources. Such behavior is in agreement with the analysis reported in Blanco et al (2003), which shows how SD events in 2002 in Lecce are characterized by aerosol with a number distribution peaked at about 2 μ m. This means that the mass-distribution is peaked to diameters larger than 2 μ m. Measurements taken in Spain (Querol et al 2001) have also indicated that the PM2.5/PM10 ratio decreases during SD events.

Conclusions

The present work reports an analysis of concentration levels of PM10, TSP and PM2.5 measured in different sites in the Salentum Peninsula. Measurements were carried out discontinuously in the years 2002, 2003 and 2004 with gravimetric detection of aerosol deposited on filters over 24 hours. Due to the general lack of data in the scientific literature relating to aerosol concentrations in this area of the southeastern Italy, this work constitutes a preliminary assessment of the situation, which could be a basis for planning future monitoring and investigations. Measurements sites were representative of the urban background. Results indicate that the average value of PM2.5 is 22 μ g/m^3, average PM10 is 27 μ g/m^3 and average TSP is 46

μ g/m^3. Daily concentrations of TSP and PM10 follow distributions in good agreement with lognormal statistical distributions.

On rainy days PM10 concentrations decrease on average by about 30%, and TSP concentration by about 35%. Daily concentrations are usually related to wind speed, with the exception of high peaks due to the contribution of transport of African Dust, or to the situation of measurements sites near local industrial sources presenting plumes which could arrive at the sensors more concentrated at high wind speed. Our results show that, on average, PM10 concentrations are about 31% lower for wind speed (detected at 10m above the ground) greater than 3 m/s, as compared to concentrations measured at wind speed lower than 3 m/s, while TSP is reduced by 24%. During the research period, 5 days occurred on which the daily concentration of PM10 was above the legislation limit of 50 μ g/m^3 . Our analysis showed that on 3 days out of 5, the occurrence of a long range transport of aerosol from Africa. The results indicate, although with limited statistics, that during these episode of Sahara Dust the PM10/TSP ratio does not change markedly, while the the PM2.5/TSP ratio seems to decrease (although only one day of comparison was available from the monitoring network for PM2.5/TSP ratio).

Such results could be explained in terms of the transport of particles mainly in the coarse fraction between 2.5 μ m and 10 μ m. This is compatible with the data obtained in the Salentum peninsula during other research studies (Blanco et al 2003).

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Table Captions

Table 1) Average concentrations and standard deviations of TSP and PM10 for the different years. The last row is the combination of all data.

Table 2) Average concentrations and standard deviations of TSP and PM10 for the different seasons.

Table 3) Daily average concentrations of TSP and PM10 during days with Pm10 concentration above the limit of 50 μ g/m^3. Also reported the is the PM10/TSP ratio and the classification as Sahara Dust event (SD) and Non Sahara Dust event (NSD).

Figure Captions

Fig. 1) Mobile laboratory with telescopic mast (in the rear) retracted.

Fig. 2) Map of the Salentum peninsula indicating (black points) the measurement sites located near the main town of Lecce.

Fig. 3) Comparison of distribution of measured concentrations (marks) with lognormal distribution (continuous line). (a) PM10 data. (b) TSP data.

Fig. 4) (a) PM10 concentrations against prevalent wind direction for different measurement sites. (b) PM10 concentrations against average wind velocity for different measurement sites. Meteorological data are measured at 600m (985 hPa) above the ground.

Fig. 5) (a) TSP concentrations against prevalent wind direction for different measurement sites. (b) TSP concentrations against average wind velocity for different measurement sites. Meteorological data are measured at 600m (985 hPa) above the ground.

Fig. 6) (a) PM10 concentrations against prevalent wind direction for different measurement sites. (b) PM10 concentrations against average wind velocity for different measurement sites. Meteorological data are measured at 10m above the ground in the concentration measurement site.

TABLE 1

Year	TSP			PM10			PM2.5		
	Conc. ($\mu g/m^3$)	Std. Dev ($\mu g/m^3$)	n. Samples	Conc. ($\mu g/m^3$)	Std. Dev. ($\mu g/m^3$)	n. Samples	Conc. ($\mu g/m^3$)	Std. Dev. ($\mu g/m^3$)	n. Samples
2002	52	25	31	41	18	7	--	--	
2003	44	16	37	25	15	49	--	--	
2004	40	16	24	27	9	43	22	8	24
Averages	46	20	92	27	14	99	22	8	24

Table 1) Average concentrations and standard deviations of TSP and PM10 for the different years. The last row is the combination of all data.

TABLE 2

Season	TSP			PM10		
	Conc. ($\mu g/m^3$)	Std.Dev. ($\mu g/m^3$)	n. Samples	Conc. ($\mu g/m^3$)	Std.Dev. ($\mu g/m^3$)	n. Samples
Winter	44	21	45	23	12	58
Summer	47	18	47	32	14	41

Table 2) Average concentrations and standard deviations of TSP and PM10 for the different seasons.

TABLE 3

DATE	PM10 $\mu g/m^3$	PTS $\mu g/m^3$	PM10/PTS	TOMS, LIDAR, MAPS CLASSIFICATION
24/07/2002	80	113	0.71	SD
03/02/2003	77	Not available	Not available	NSD
07/05/2003	53	Not available	Not available	NSD
09/05/2003	57	77	0.74	SD
10/05/2003	54	62	0.87	SD

Table 3) Daily average concentrations for TSP and PM10 during days with Pm10 concentration above the limit of 50 μ g/m^3. Also reported is the PM10/TSP ratio and the classification as Sahara Dust event (SD) and Non Sahara Dust event (NSD).

FIGURE 1

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FIGURE 2

Fig. 2) Map of the Salentum peninsula indicating (black points) the measurement sites located near the main town of Lecce.

FIGURE 3

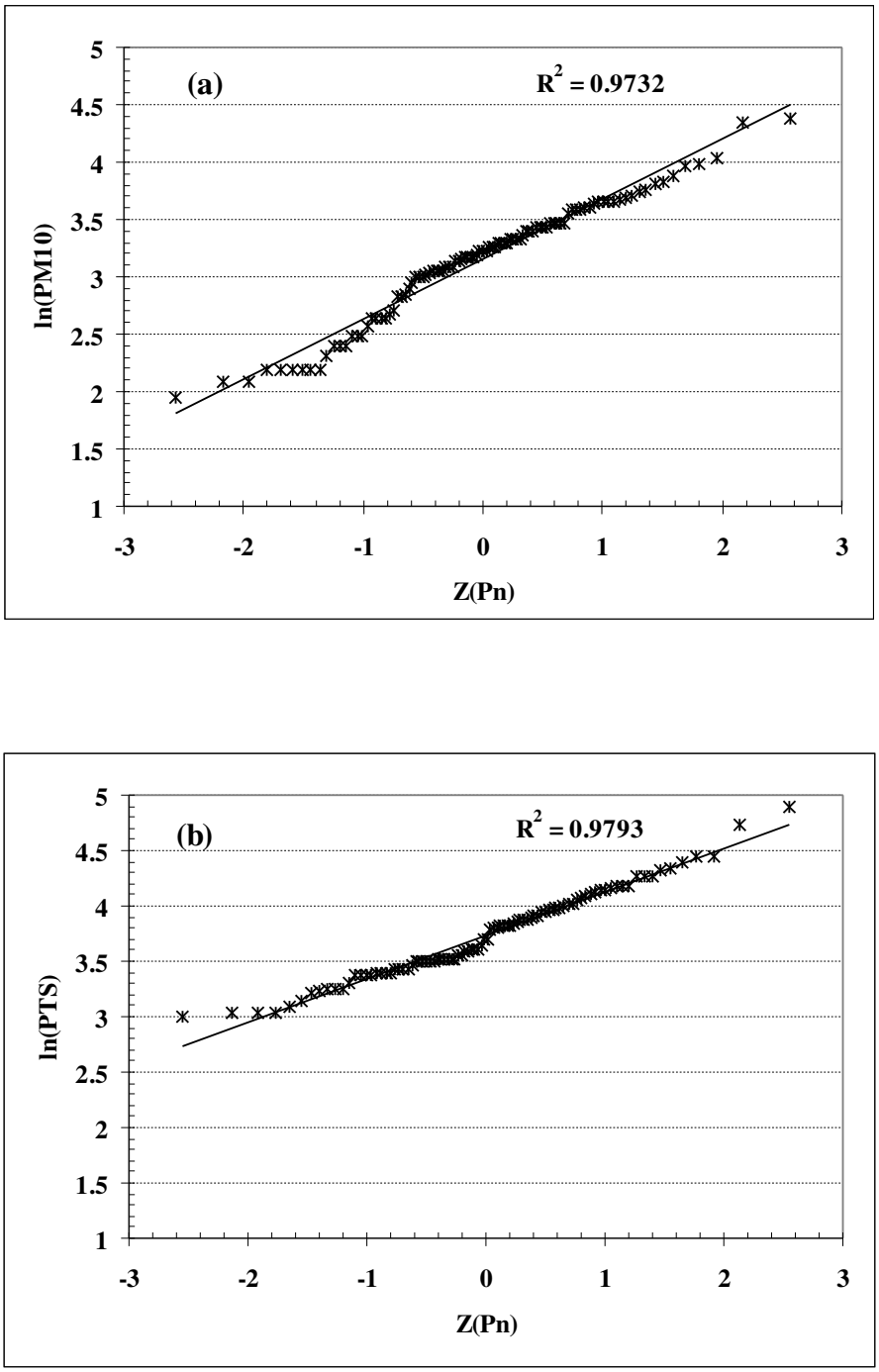

Fig. 3) Comparison of distribution of measured concentrations (marks) with lognormal distribution (continuous line): (a) PM10 data. (b) TSP data.

FIGURE 4

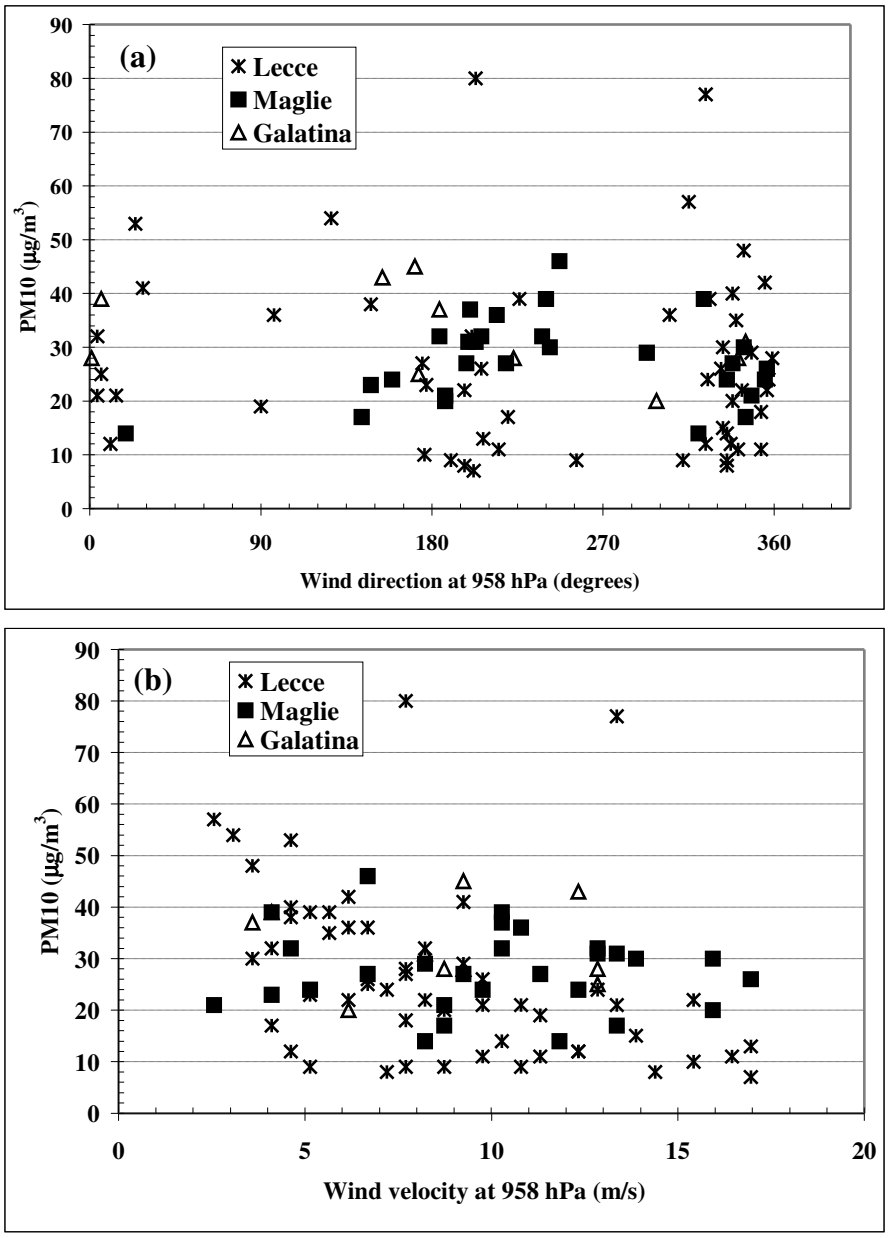

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FIGURE 5

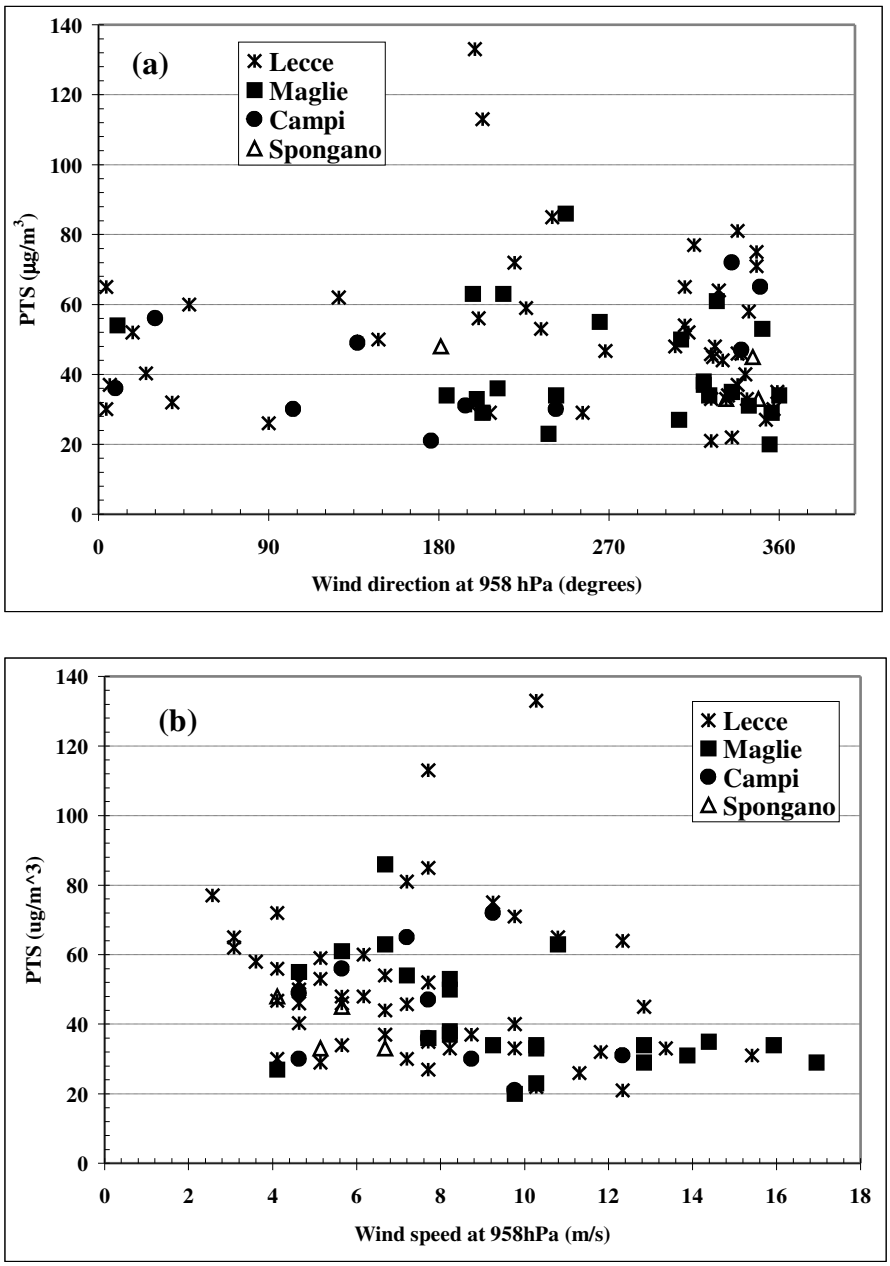
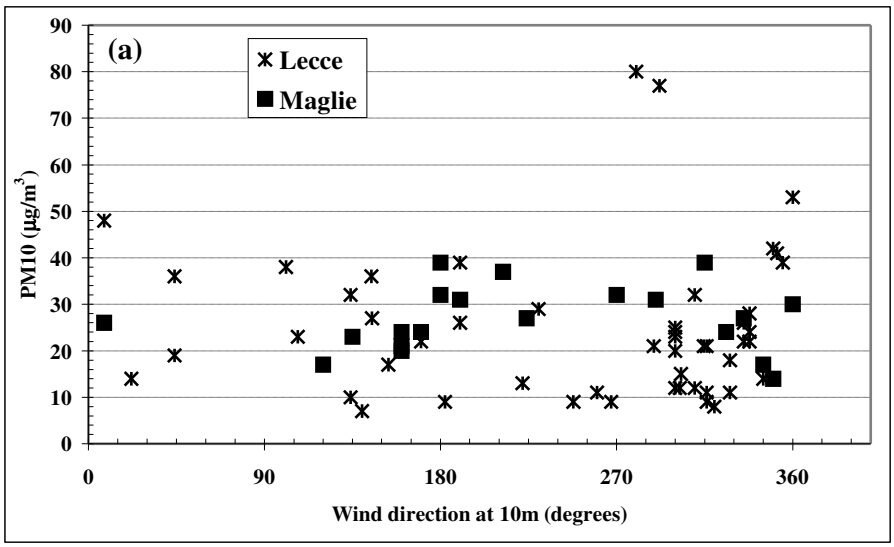


Fig. 5) (a) TSP concentrations against prevalent wind direction for different measurement sites. (b) TSP concentrations against average wind velocity for different measurement sites. Meteorological data are measured at 600m (985 hPa) above the ground.

FIGURE 6



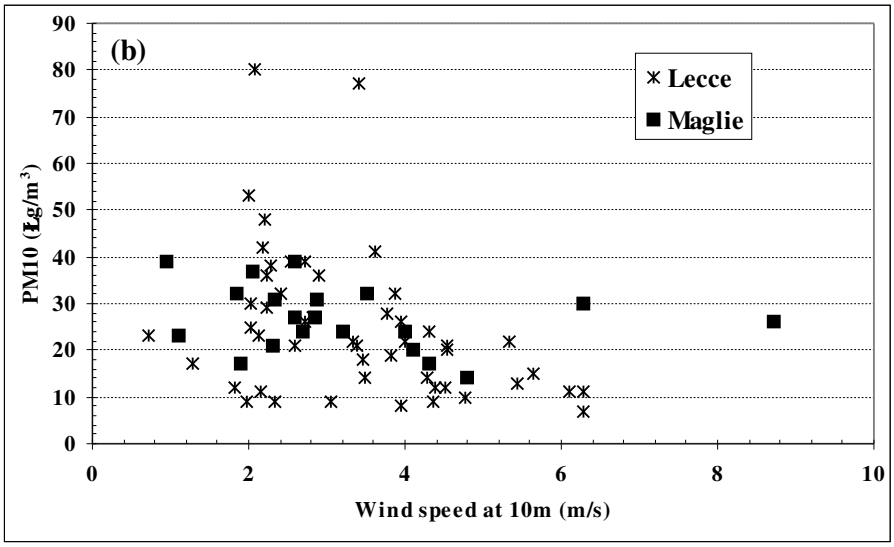


Fig. 6) (a) PM10 concentrations against prevalent wind direction for different measurement sites. (b) PM10 concentrations against average wind velocity for different measurement sites. Meteorological data are measured at 10m above the ground.