PM_{10} and $PM_{2,5}$ evaluation to support air quality plans and programs in Lombardy region

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Lombardy Region is one of the most populated areas of Italy: its economy represents one-fifth of the total Italian economy, and generates about the 21% of the national GDP; enterprises account approximately for 15% of the national total, with a productive sector in which co-exist a number of great, medium and small enterprises.
Most of the people and the activities are located in the plain area, where, as in the rest of the Po Valley, the dispersion conditions are critical due to the particular orography; in Lombardy, therefore, concentrations of particulate matter (PM_{10}) overrun the European thresholds for daily and annual mean concentrations.
In accordance with the legal framework, the PM_{10} monitoring in Lombardy has been integrated by the $PM_{2,5}$ one, with an increasing number of sensor units and with the identification, from year 2005, of a first automatic network. In the project, the main findings of $PM_{2,5}$ measures obtained till now will be shown, in relation to their temporal trend, spatial distribution and to the new European regulation's standard values. Moreover, the ratios between the $PM_{2,5}$ and PM_{10} concentrations will be considered and discussed.
The framework will be completed by particulate composition data obtained from specific research projects and by emission inventory.
All these information will be used as decisional support in the definition of remediation plans and programs.

1. Introduction

According with the legal framework (DM 60/2002 and directive 99/30) it's necessary to integrate the air quality monitoring network with $PM_{2,5}$ sensors. In Lombardy region the first $PM_{2,5}$ measures started in year 2000 with the sitting of an automatic sensor in the traffic station of Piazza Zavattari, Milan; in the following years, since 2004, some gravimetric sensors started measuring $PM_{2,5}$ within PARFIL Project, a three year long study about the distribution and the chemical composition of particulate matter in Lombardy, used as decisional support in the definition of remediation plans and programs. In the meantime, the automatic network has been developed with the addition of new $PM_{2,5}$ sensors, till the actual amount of a total of 14 units. Integrations are in progress in accordance with the new European air framework directive (2008/…/CE).
This paper analyses the main findings of this measurement period (mean values, statistical features, trends etc.); moreover, it shows some results of the regional

Emission Inventory (INEMAR), that estimates the single emission factors of the main anthropogenic activities.
Air quality network, emission inventory and specific projects are all important instruments for a complete framework of decisional support.

2. Data

The input data set is composed of both gravimetric and automatic measures, obtained from PM_{10} and $PM_{2,5}$ sensors located in each province of Lombardy region, Italy. Gravimetric instrument and some automatic monitors are characterized by daily temporal detail, while other automatic instruments use an hourly time step; where available, for specific elaborations, hourly data have been used in this work, although for PM_{10} and $PM_{2,5}$ only the daily ones are certified equivalent to the reference method.
The analyzed period is 2006 – 2007, since it's characterized by an interesting and complete data set; the temporal aggregations cover an entire year, summer (1 April – 30 September) and winter (1 October – 31 March). For each period, mean values and $PM_{2,5}$ to PM_{10} ratios were computed; moreover, only for some stations, a monthly analysis, a daily mean trend analysis, and a statistical distribution computation are presented.

2.1 Average values and $PM_{2,5}$ / PM_{10} ratios

Table 2.1 shows annual and seasonal mean values. Only data sets with an yield greater than 60% were considered.
In order to obtain a larger seasonal analysis, gravimetric series were integrated with data collected from October 2005 to December 2005, while automatic series were added with data registered in the period from January 2008 to March 2008.
As shown in table 2.1, annual $PM_{2,5}$ means exceeded the UE proposed threshold of 25 $\mu g/m^3$ in each station except for the pre-alpine site of Varese and the alpine remote site of San Colombano (SO); the highest values have been observed in the plain area, especially in the most populated and productive areas of the region, that are the metropolitan areas of Milan (MI), Brescia (BS) and Bergamo (BG) and the city of Saronno (VA), located into the plain, before the edge with the pre-alpine zone. Where available, the comparison between 2006 and 2007 means gives comparable results.
The collected datasets show the $PM_{2,5}$ seasonality, with a relevant difference between summer and winter means, due first of all to the different meteorological conditions (lower mixing height and worse diffusing situation in winter), and secondarily to the absence of emissions from the domestic heating systems.
The behavior is different for the remote station of San Colombano (2.250 m): during winter, this site is above the mixing height, with values near the instrument threshold; anyway, the concentrations in this site are all year round very low.
Monthly averages have also been computed and the results for four of the considered stations are presented in figure 2.1. These stations have been chosen as they have a quite complete dataset for the two considered years, 2006 and 2007.
The ratios between $PM_{2,5}$ and PM_{10} means (table 2.2) were computed to evaluate the fine matter contribution to total particulate amount, varying between 40% and 80%,

with exception of the remote site of San Colombano, where the high ratio values are probably influenced by the persistence of the fine fraction in the atmosphere.

Table 2.1: $PM_{2,5}$ annual and seasonal means

	Cantù (CO)*	Varese (VA)*	Cremona (CR)*	San Colombano (SO)*	Casirate d`Adda (BG)	Milano Juvara/ Pascal (MI)**	Saronno Santuario (VA)	Villaggio Sereno (BS)**	Cornale (PV)	Seriate (BG)	Calusco d' Adda (BG)	Merate (LC)	Ponti sul Mincio (MN)
	ANNUAL MEANS (μg/m^3)												
2006	28	18		5	36	39	32	31		28			
2007			30		30	35	34	36	28	34	34	31	27
	SEASONAL MEANS (μg/m^3)												
WINTER 0506	37	27		4									
SUMMER 06		12		6	20	20	13			17			
WINTER 0607	37	25	46	4	33	54	49	52	34	45	55	51	
SUMMER 07			16		17	18	17	19	17	20	20	17	16
WINTER 0708					50	48	45	49	35	43	42	40	37

* gravimetric sensor

** dataset obtained as an integration of automatic and gravimetric data

Table 2.2: $PM_{2,5}$ / PM_{10} annual and seasonal ratios

	Cantù (CO)*	Varese (VA)*	Cremona (CR)*	San Colombano (SO)*	Casirate d`Adda (BG)	Milano Juvara/ Pascal (MI)**	Saronno Santuario (VA)	Villaggio Sereno (BS)**	Calusco d' Adda (BG)	Merate (LC)	Ponti sul Mincio (MN)
PM2,5 / PM10 ANNUAL RATIOS											
2006	0,61	0,46	0,82	0,79	0,66	0,70	0,72	0,58			
2007	0,52	0,47	0,55	0,84	0,62	0,69	0,80	0,73	0,81	0,58	0,63
PM2,5 / PM10 SEASONAL RATIOS											
WINTER 0506	0,53	0,50		1,00							
SUMMER 06	0,46	0,58		0,87	0,60	0,68	0,50	0,58			
WINTER 0607	0,54	0,46	0,62	0,95	0,51	0,71	0,85	0,78			
SUMMER 07	0,50	0,42	0,50	0,75	0,62	0,59	0,65	0,63	0,70	0,50	0,51
WINTER 0708					0,75	0,74	0,79	0,75	0,80	0,60	0,67

* gravimetric sensor

** dataset obtained as an integration of automatic and gravimetric data

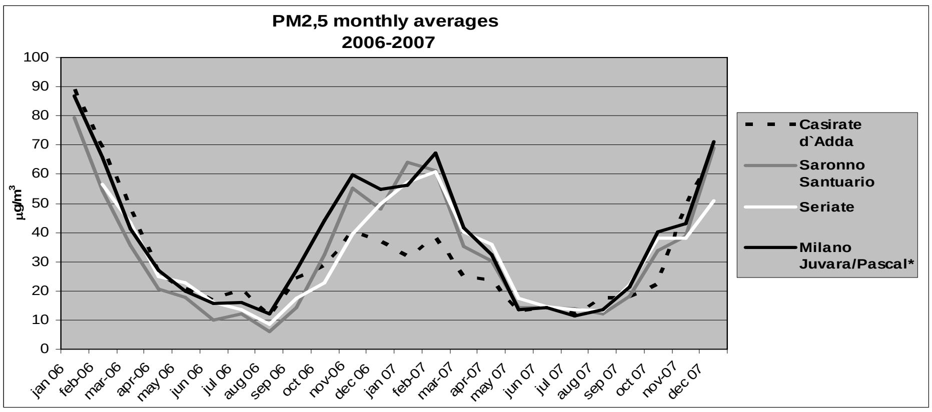


Figure 2.1: $PM_{2,5}$ monthly trends at Casirate d'Adda (BG), Milan Juvara/Pascal, Seriate (BG) and Saronno Santuario (VA) stations.

2.2 Statistical analysis

As it is well known from literature, both PM_{10} and $PM_{2,5}$ fit a lognormal distribution. In this study a size-frequency distribution analysis has been computed for $PM_{2,5}$ daily series collected at Juvara/Pascal station (Mi) and Casirate d'Adda station (Bg) in 2006 and 2007. The lognormal distribution has been used to analyze the two datasets. Considering μ and σ as the mean and standard deviation of the variable's logarithm, the following parameters were obtained: μ=3,3, σ=0,8 and geometric mean=27 for Juvara/Pascal station; μ=3,2, σ=0,7 and geometric mean=26 for Casirate d'Adda station. The results of the statistical and size-frequency analysis are shown in figures 2.2 and 2.3; the graphs confirm that both the distributions fit a lognormal trend.

Table 2.3 contains some statistic indexes calculated for the two considered datasets; if compared, the statistic parameters of the two series look rather similar, as both of them are located in the plain area of the region, although there are some differences due to the different station type: Juvara is a background station located in urban area, while Casirate is a background site too, but it's placed in a rural contest.

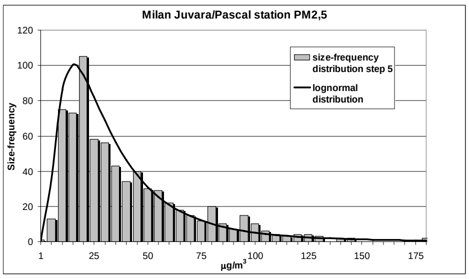


Figure 2.2: size-frequency distribution for $PM_{2,5}$, Juvara/Pascal station (Mi), 2006-2007

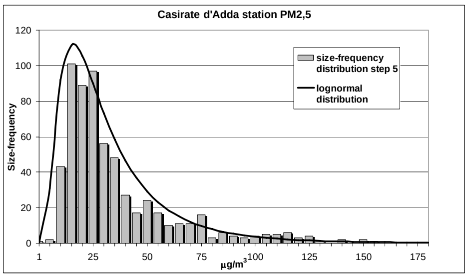


Figure 2.3: size-frequency distribution for $PM_{2,5}$, Casirate d'Adda station (Bg), 2006-2007

Table 2.3: statistic indexes for $PM_{2,5}$ dataset, Juvara/Pascal station (Mi) and Casirate d'Adda station (Bg)

	Minimum µg/m³	Maximum µg/m³	Arithmetic mean µg/m³	50% percentile µg/m³	95% percentile µg/m³	Yield, %
Milano via Juvara / Pascal	1	177	39	28	98	98%
Casirate d' Adda	4	155	33	24	96	85%

2.3 $PM_{2,5}$ historical trends

Piazza Zavattari station, located in the city of Milan, is characterized by a long-term $PM_{2,5}$ dataset, starting from October 2000 till the end of November 2006. In particular, for this whole period TEOM $PM_{2,5}$ measures are available, while only from December 2004 to November 2006 FDMS corrected data also exist. In order to obtain a consistent FDMS dataset, data have been rearranged by calculating $PM_{2,5}$ monthly mean values for the two instruments and computing the ratio between them, obtaining a mean ratio for each month; lately, these ratios have been applied to TEOM data in order to obtain simulated FDMS data where not available. In this way, the purpose of having an entire FDMS dataset from 2001 to 2006 has been achieved. The results of this procedure are shown in the following figures: the first one (2.4) illustrates Milan Zavattari annual trend compared to PM_{10} averages collected at Milan Juvara station. The patterns are quite similar, with a slight gradient. Zavattari trend shows a lower value in 2004, principally due to a data gap in the cold season of that year. The other figure (2.5) shows FDMS $PM_{2,5}$ monthly trend.

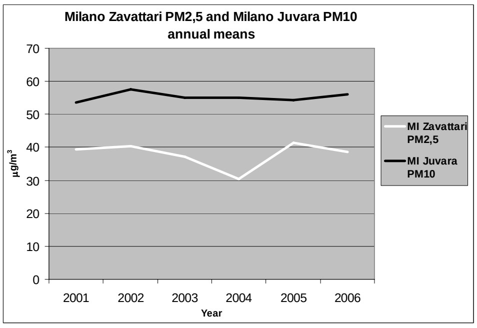


Figure 2.4: Annual trend for Piazza Zavattari PM2.5 and Juvara PM_{10} stations, Milan.

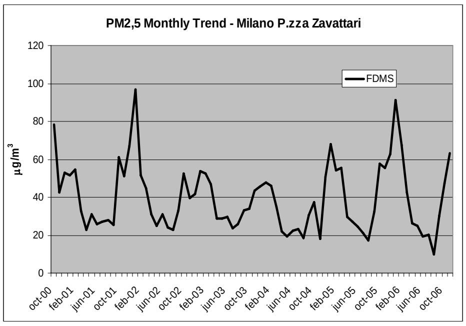
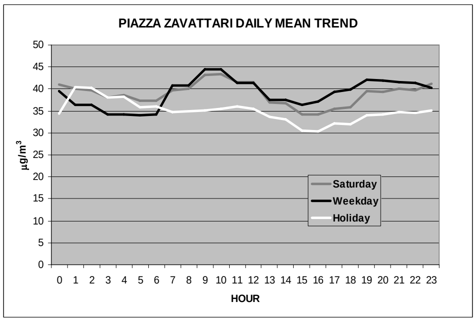


Figure 2.5: $PM_{2,5}$ monthly trend for FDMS dataset (2000-2006), Piazza Zavattari station, Milan.

2.4 PM2,5 daily mean trend

In this chapter are presented the results of the calculation of the $PM_{2,5}$ daily mean trend for the two stations of Piazza Zavattari (Mi), an example of an urban traffic station, and Casirate d'Adda station, that is a rural background one. With "daily mean trend" is here defined the sequence of mean hourly values calculated over a chosen time frame for each hour during a day; the considered periods are 2000-2006 for Zavattari station and 2006-2007 for Casirate d'Adda station, with distinction among weekdays, Saturdays and holidays.

In figures 2.6 and 2.7 are shown the results of this evaluation. It's clear that the distinction between the daily trend of the two graphs is due to their different location; the traffic station of Zavattari is influenced by urban traffic, therefore on weekdays and Saturdays there are peaks during rush hours (9 AM and 6 PM), while during Sundays and Holidays there's an increase of $PM_{2,5}$ concentration in the first hours of the day, caused by late night homecomings. The background station of Casirate d'Adda instead is characterized by a flatter trend, as it's not directly influenced by traffic emissions. However, also in this case, the transport of traffic pollution may be linked to the effect on Saturdays and holidays earliest hours, characterized by higher values, and on the small peak on 11 – 12 AM, that could be related to the early rush hour traffic, delayed by the transportation time. In this case, nights show higher values than daytime because the $PM_{2,5}$ concentration is directly influenced by the mixing layer height, that grows up during the day thanks to the warmer temperatures.



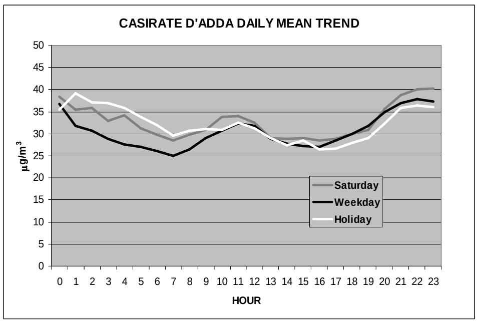


Figure 2.6: daily mean trend for $PM_{2,5}$ dataset, Piazza Zavattari station (Mi), urban traffic station

Figure 2.7: daily mean trend for $PM_{2,5}$ dataset, Casirate d'Adda station (Bg), rural background station

3. Emission inventory in Lombardy region

Emission inventory for Lombardy is based on the INEMAR database (EMission INventory in AiR, http://www.arpalombardia.it/inemar/inemarhome.htm), an archive that allows to estimate emissions to a municipality level for different pollutants, activities and fuels. This database contains all the variables needed to estimate emissions: activity data, emission factors and all statistical data required for spatial and temporal desegregation of emissions.

In figures 3.1 and 3.2 are shown the results for the 2005 inventory of PM_{10} and $PM_{2,5}$ emissions; the two graphs look similar in each considered macro sector. Therefore, it's clear that the same source pollution mitigation strategies are generally suitable for both PM_{10} and $PM_{2,5}$.

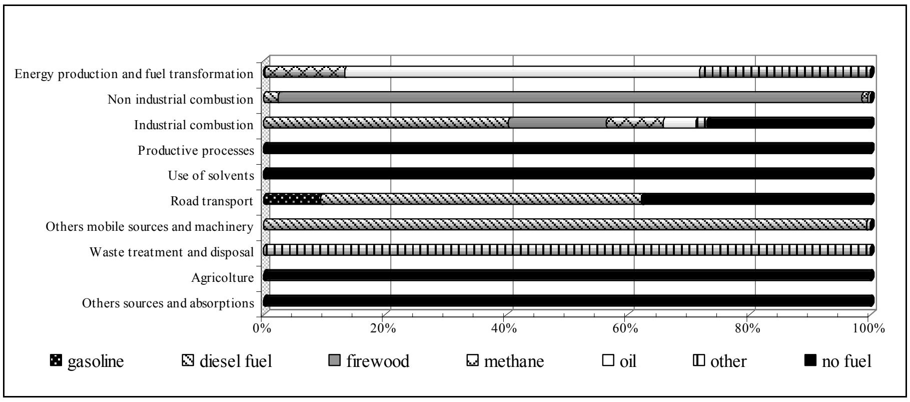


Figure 3.1 Lombardy region 2005 PM_{10} emissions (t/year), distinguished in macro sector and fuel.

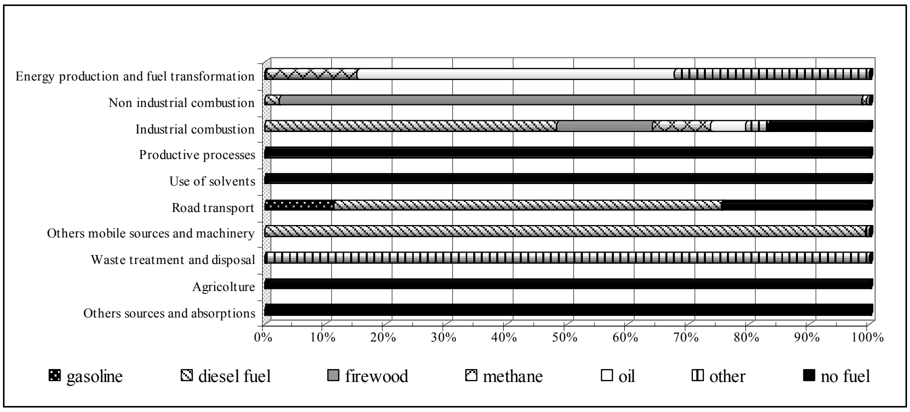


Figure 3.2 Lombardy region 2005 $PM_{2,5}$ emissions (t/year), distinguished in macro sector and fuel

4. PM_{10} and $PM_{2,5}$ composition

PM_{10} and $PM_{2,5}$ aerosol samples were collected in Milan and in other cities in Lombardy region in the frame of the ParFiL project (Particolato Fine in Lombardia, http://www.arpalombardia.it/qaria/doc_ProgettiInCorso.asp#PARFIL). The sampling sites have been chosen according to the different contributions estimated for combustion and traffic to particulate matter emissions. One of the project's outputs was the chemical and physical characterization of the collected samples with the identification of their typical composition profiles in relation to their spatial, temporal and dimensional distribution. Figures 4.1 and 4.2 illustrate the comparison between PM_{10} and $PM_{2,5}$ average composition in different 7 regional sites in 2005.

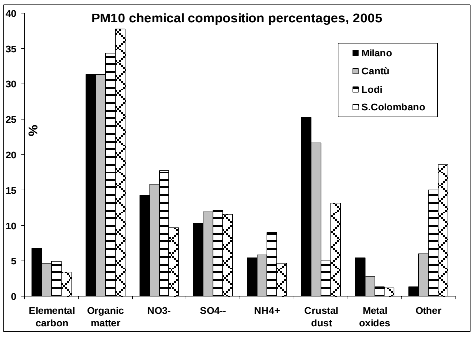


Figure 4.1: PM_{10} chemical composition percentages. Parfil project, 2005

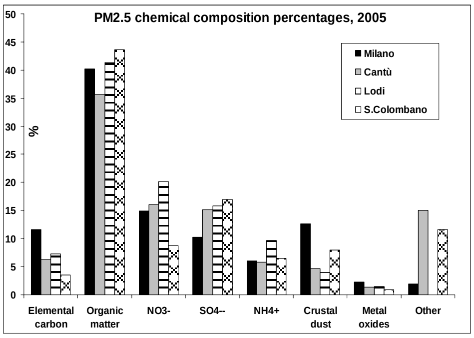


Figure 4.2: $PM_{2,5}$ chemical composition percentages. Parfil project, 2005

The two compositions look comparable; obviously, for $PM_{2,5}$ the contribute of crustal dust is less significant because it represents the coarser fraction of particulate getting re-lifted by the wind. Moreover, other studies have demonstrated that PM_{10} and even more $PM_{2,5}$ are constituted by a secondary organic and inorganic fraction for more than 50% (Cazzuli O., Padova, 2008).

5. Conclusions

The national legal framework (DM 60/2002 and directive 99/30) states the necessity of $PM_{2,5}$ levels measurements. According to this, Lombardy region counts 14 PM2,5 automatic sensors, widely spread all around the territory.

In agreement with the new European air framework directive (2008/…/CE) the annual $PM_{2,5}$ mean values will have to respect the new limit of 25 $\mu g/m^3$. The analysis on $PM_{2,5}$ data demonstrate that the annual $PM_{2,5}$ means exceed the UE proposed threshold of 25 $\mu g/m^3$ in most of the considered stations, so that, specific mitigation policies are necessary.

The emission inventory attests that PM_{10} and $PM_{2,5}$ are characterized by the same sources in a similar way; consequently, it can be thought that the same source mitigation policies adopted for PM_{10} may be useful also for $PM_{2,5}$.

Finally, the chemical composition analysis shows high percentages of secondary organic matter and inorganic compounds, such as nitrates, sulphates and ammonium, for both $PM_{2,5}$ and PM_{10}; therefore it's required a strict control on these kinds of precursors.

All these considerations indicate that $PM_{2,5}$ is not a "new" pollutant but it's inserted in the same context of PM_{10} control.

6. References

Cazzuli O., 2008, "Lo studio della composizione chimica del particolato a supporto dei piani di risanamento", Convegno Dionex "Particolato atmosferico dal campionamento all'analisi", Padova, 28 febbraio 2008, web site: http://www.coboldi.it/dionex/padova_280208/PD_280208.html.

Ministero dell'Ambiente e della Tutela del Territorio, 2002, Decreto Ministeriale n. 60 del 02 aprile 2002, Italy.

European Council, 1999, European Directive 1999/30/CE.

http://www.arpalombardia.it/qaria/doc_ProgettiInCorso.asp#PARFIL.

http://www.arpalombardia.it/inemar/inemarhome.htm.