DRAFT

PM atmospheric concentrations in France :

analysis and key findings

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1 Introduction

Since the year 2000, a significant effort has been provided in France for enhancing the air quality monitoring structure devoted to PM measurement Although PM_{10} was the priority, the assessment of the $PM_{2.5}$ concentrations has not been neglected because of their adverse effects for human health. Research partners together with operational national and local organisations in charge of air quality monitoring have developed skills and experience related to the behaviour of PM emissions and concentrations in the atmosphere. In 2007, it seems feasible to establish a first assessment of the PM situation in France, compiling data issued from all types of information :

- Emission inventories allow to analyse source apportionment, to assess the efficiency of control measures, and to feed air quality models. Development of PM (and their precursors) emission inventories is a crucial and difficult task, still hampered by the lack of knowledge related to the sources and the processes. However results considered as reasonably relevant are now published.
- In 2006, more than 300 PM_{10} measurement stations and 65 $PM_{2.5}$ stations were implemented in France. This is the result of more than 15 years devoted to the development of the PM monitoring strategy in France. Most of them are TEOM or beta gauge devices which allow to get high temporal and spatial resolutions. With such a dense network a database relevant for reporting on the PM situation and for trend analysis has been built. More information is provided by research projects, supersites, and fields campaigns devoted to a better characterisation of the aerosol composition and the origin of PM events.
- Although particulate modelling is still a science under development, a dynamic policy research in that field, allows the implementation of PM models dedicated to air quality forecasting and to a comprehensive analysis of the aerosol phenomenology. Thus modelling is considered as a part of the whole PM monitoring system implemented in the country .

The present report compiles the information currently available from the French PM monitoring system and provides an interpretation of the PM situation in the country. A first section is dedicated to the emissions status. The second part describes the PM measurement network and field campaigns . Results and trends issued from these data are

presented in a third section. Before concluding, an analysis of the PM phenomenology in France based on measurement and modelling is proposed.

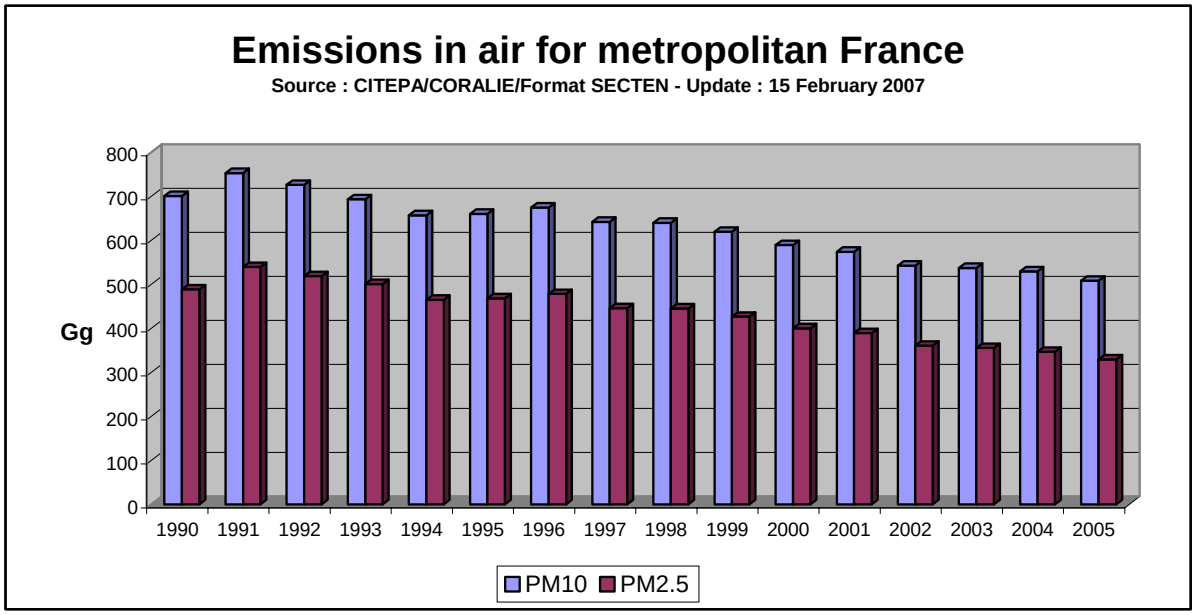
2 PM emission inventories

Emission data related to PM_{10} and $PM_{2.5}$ from 1990 to 2005 are given below. For the entire period, estimates provided in the previous inventories have been reviewed and corrected to take into account updated statistics, improved knowledge and possible changes in methodology.

PM EMISSIONS IN AIR FOR METROPOLITAN FRANCE (units Gg = kt)

Source CITEPA / CORALIE / format SECTEN — Update : 15 February 2007

Année	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005
PM10	**701**	**753**	**726**	**693**	**657**	**660**	**674**	**642**	**640**	**619**	**589**	**574**	**542**	**537**	**529**	**508**
PM2.5	**489**	**540**	**519**	**500**	**465**	**468**	**478**	**446**	**445**	**427**	**400**	**389**	**360**	**355**	**347**	**329**



For the year 2005, **emissions of PM_{10}** in metropolitan France are **508 Gg**. These emissions decreased by about 28% from 1990 to 2005, despite the high emission level in 1991. The main contributors to these emissions are, by order of importance:

- Agriculture and forestry (29%), especially ploughing
- Manufacturing industry (28%), especially quarrying and building sites
- Residential/tertiary (28%), especially wood, coal, and fuel combustion.

Emissions of $PM_{2.5}$ in metropolitan France are **329 Gg** in 2005. These emissions decreased by about 33% from 1990 to 2005, despite the high emission level in 1991.
The main contributors to these emissions are, by order of importance:

- Residential/tertiary (41%), especially wood, coal, and fuel combustion
- Manufacturing industry (26%), especially quarrying and building sites
- Agriculture and forestry (17%), especially ploughing.

For both PM_{10} and $PM_{2.5}$ emissions fell in all sectors except for transports (not including road transport). This fall is partially due to progress in dust removal techniques, but also comes from structural effects such as technological improvements in biomass combustion.

3 PM measurement networks description

Intensive work related to the measurement of PM concentrations has been realised these last years in France. A quite dense operational network devoted to reporting, monitoring and public information has been implemented, together with supersites devoted to research purposes and monitoring following the EMEP strategy. Field campaigns allow to get a complementary information, and to insure the equivalence of the automatic methods with the reference one (gravimetry HVS DA-80) described by the norm CEN/TC264, 2005.

3.1 Network implemented by the AASQA for monitoring purposes

Figure 1 presents the location of PM10 measurement stations in 2006. The stations are operated by local associations in charge of air quality monitoring (AASQA), with the agreement of the Ministry of Ecology. The observations are compiled in two databases implemented and maintained by the Agency in charge of the Environment and Energy (ADEME): near real time unvalidated data are stored in the BASTER database when validated data are compiled in a longer term process in the BDQA database.
All the PM monitoring stations are based on automatic devices (85% TEOM and 15% beta gauges) which allow high temporal resolution measurement of the PM mass.

PM10 stations are distributed all over the country in the following way : about 72% in urban and suburban areas, 2,5% in rural areas. Near emissions sources, about 13% of the number of sites are traffic ones and 12.5% are industrial stations.
PM2.5 stations are mainly located at urban and suburban sites (79 %), 19% at traffic sites and the difference concerns industrial sites

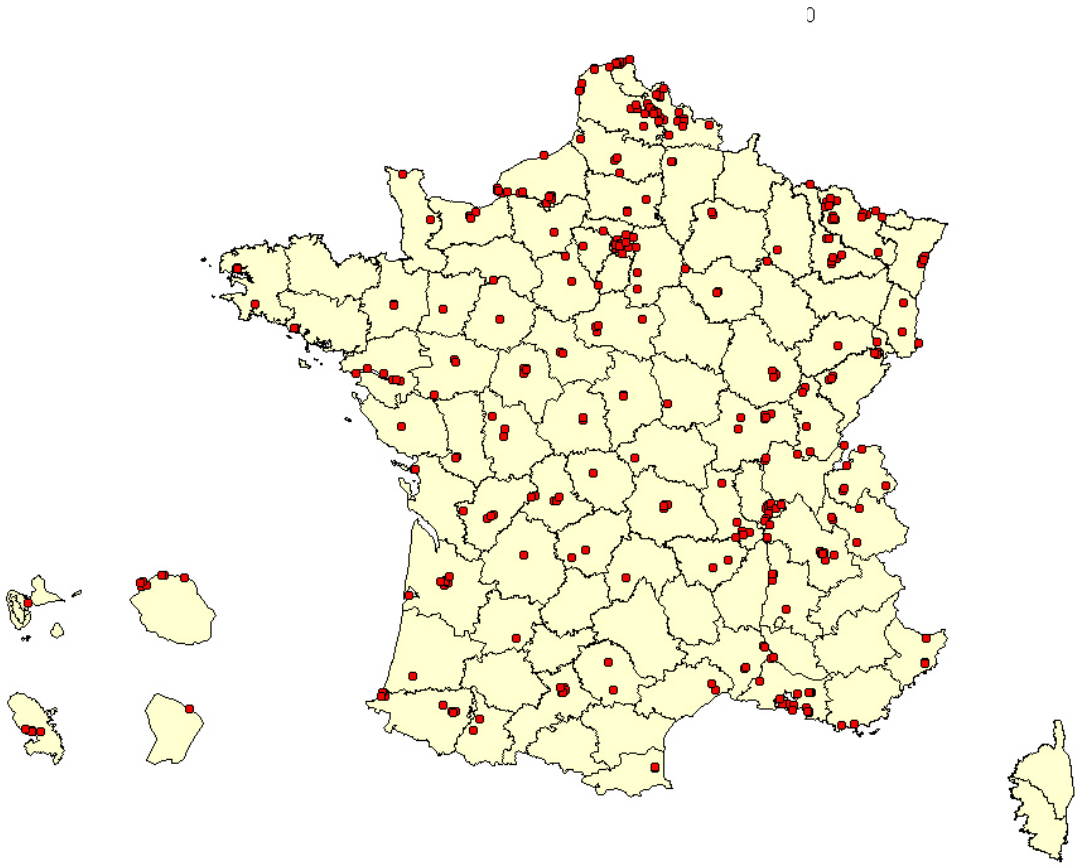


Figure 1 : PM10 measurement network in France (2006)

3.2 Field campaigns

As far as air quality monitoring is concerned, the main field campaigns from 2002 up to now are listed below:

Objectives	material	where	When	results	Ref
Demonstration of equivalence of the TEOM-FDMS PM_{10} and $PM_{2.5}$	Partisol 2025 TEOM-FDMS	Paris (Bobigny)	2005	TEOM-FDMS ok for PM_{10} and $PM_{2.5}$	[Le Bihan, 2005 and 2006] et mathe (2006)
		Marseille	2006		
Demonstration of equivalence of the Beta Gauge MP101M-RST PM_{10}	Partisol 2025, MP101M-RST	Paris (Bobigny), 2005	2005	MP101M-RST ok for PM_{10}	
		Marseille, 2006	2006		
		Italy (b)	2003		
		Belgium (a)	2006		
Preliminary test of the TEOM-FDMS PM_{10}	Partisol 2025, TEOM-FDMS	Paris	2003, 2004	High correlation between FDMS and reference method for PM_{10}	[Bessagnet, 2004 ?]
PM_{10} chemical composition	Partisol 2025	Paris, Rouen	2002	Confirmation of the TEOM 50°C under-estimation of PM_{10}	[Blanchard, 2002]

(a) : French participation to a programme leaded by Belgium.
(b) : Intercomparison test performed by CNR-IIA (Italy)

3.3 EMEP remote stations and research supersites (figure 2)

In the framework of the EMEP monitoring strategy, two french remote stations of the French Background Air Pollution Monitoring Network (MERA) have been recently equiped to measure PM10. The sampling method used is the automatic system TEOM. The sampling inlet is US EPA type and operate at a flow of 1m3/h. The mean concentration measured at Peyrusse Vieille (FR13 EMEP site) was 15.3 µg/m3 (Aug 06 to Feb 07, average daily data, 9h-9h TU)) with a maximum daily value at 60 µg/m^3 in October 2006 and a 98-percentile at 36 µg/m^3 (figure 3).

Before the decision of choice of monitor, a preliminary comparison had been organized during two months in 2005 with the objectives to define the type of automatic monitor (TEOM or TEOM-FDMS) which could be used in this rural station and to show the equivalence (CEN/TC264, 2005, cf 3.4) of automatic methods compared the reference method by gravimetry (HVS DA-80)(EN 12341, using weighing procedure requirements of EN 14907). The measurement uncertainties for the PM10 measurement have been also quantified from collocated parallel measurements to be ±2 µg/m^3 (13.6%) and ±4.4 µg/m^3 (29,4%) for TEOM-FDMS and TEOM respectively at the concentration 15 µg/m^3. The equivalence has been obtained for both equipments on the measurement period [Coddeville, 2006].

At present a 1-year measurement campaign, started in June 2006, is running in another EMEP-MERA site (FR09 Revin) with the same objective to evaluate the equivalence of the automatic method. The first preliminary results already show the necessity of TEOM FDMS. During the period of measurement, the mean concentration measured at Revin is 13.8 µg/m^3 (TEOM, Aug 06 to Feb 07, average daily data, 9h-9h TU) with a maximum value at 46 µg/m^3 and a 98-percentile 31 µg/m^3 in September 2006.
Nitrates and ammonium daily concentrations are also measured at the two sites and could help out with the interpretation.

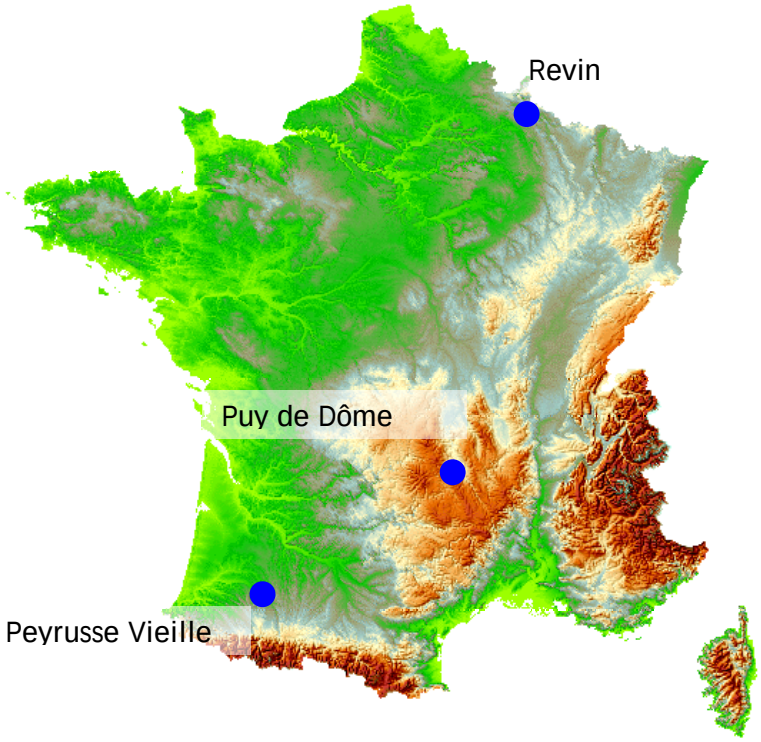


Figure 2 : location of EMEP supersites and Puy-de-Dome PM station

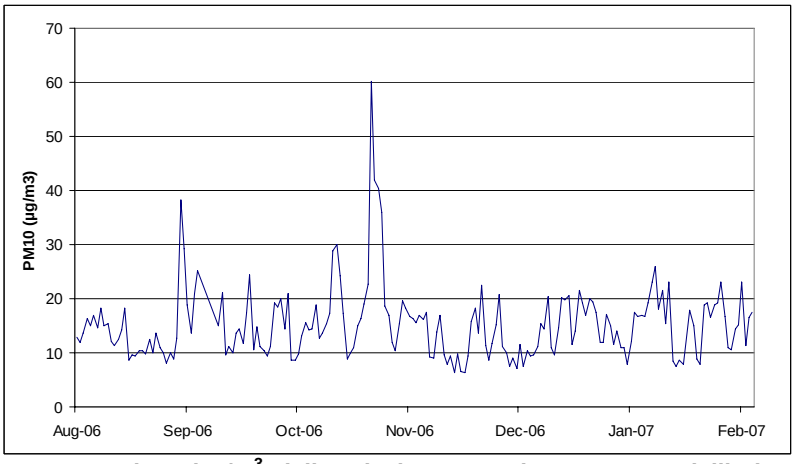


Figure 3 : PM10 concentrations (µg/m^3, daily value) measured at Peyrusse Vieille (FR13, EMEP station) from July 2006 to February 2007

The Puy de Dôme supersite is operated by the Observatoire de Physique du Globe de Clermont-Ferrand. The site is located in the Auvergne region (Central France) at 1465 m high, far away from emission sources. The sampling site is influenced by both long-range transport of gas and particles from the free troposphere and by the more regional boundary layer air. On average, the site is under the influence of free tropospheric air during the October/April period, and during night-time (8PM-10AM) during the remaining portion of the year.

The site is equipped with complete instrumentation to characterize aerosol properties:

- Aerosol Chemistry : low volume samplers for IC and OC/EC chemistry (PM10) since 2003
- Aerosol Chemistry : High volume sampler for IC and organic speciation (PM10-CARBOSOL program) since 2004
- Aerosol Size segregated Chemistry since 2006
- Size distribution since 2005

- Optical properties : scattering (nephelometer) since 2006 and absorption (aethalometer) coefficients since 2000
- Hygroscopic properties : HTDMA during research campaigns in 2006
- Aerosol Mass –(RH-Controlled gravimetry on aerosol filters PM10) since 2006 and number concentrations (CN counter) since 2003

Finally, it should be noted that puy de Dôme is now part of the EUSAAR network. The objective of the project EUSAAR is the integration of measurements of atmospheric aerosol properties performed in a distributed network of 20 high quality European ground-based stations (Supersites). This integration contributes to a sustainable and reliable operational service in support of policy issues on air quality, long-range transport of pollutants and climate change. The project is coordinated by CNRS in Clermont-Ferrand.

3.4 Issues related to the use of TEOM and beta gauge for the regulatory network

Why does the French networks only use automated methods for PM_{10} monitoring ?
The reference method for the sampling and measurement of PM_{10} as mentioned in the 1st Daughter Directive 99/30/EC is described in EN 12341:1999 standard. It is based on 24 hours sampling of PM_{10} on filters followed by filters weighting under controlled humidity and temperature conditions. However, this method is not sufficiently operational for the routine monitoring. The main difficulties encountered when using this method are:
- delicacy of filter handling procedures;
- impossibility to provide real-time data for public information (daily basis prescribed in Directive);
- high running costs (weighting room with controlled atmosphere, whole measurement process).

Automatic tools bring a solution to these difficulties.

<u>The PM automated instruments from the 90's do not measure correctly the PM semi-volatile compounds.</u>
In the 90's, the French PM_{10} monitoring network has been developed both on the TEOM (Tapered Element Oscillating Microbalance) and the Environnement SA MP101 Beta Gauge (β ray attenuation monitor).
However, the use of these techniques leads to an underestimation of PM levels compared with the reference method.
The main reason for that was clearly identified as the volatilisation of some PM_{10} compounds in the instrument. This is illustrated in the Figure 4 for the TEOM: TEOM measurements need to be corrected with volatilised compounds (ammonium nitrate in this case). This underestimation is an important point because it makes the TEOM measurement and beta gauge methods not equivalent to the reference method.

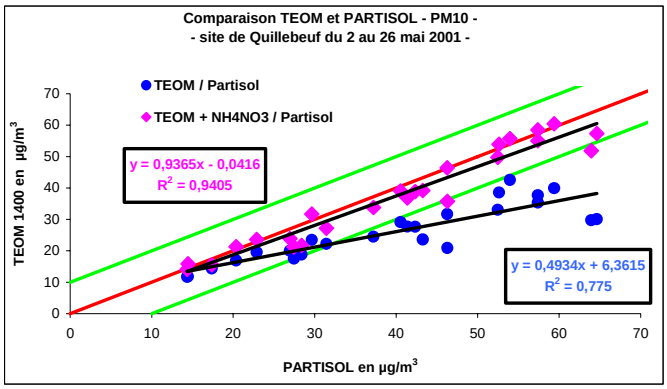


Figure 4 : Correlation between TEOM and reference method (blue), and TEOM corrected with NH_4NO_3 and reference method (pink), for PM_{10} measurements.

→ Mathematical correction can not tackle the problem

The first possibility examined to tackle this problem was to correct TEOM measurements with a conversion factor. However, field operations have demonstrated that this conversion factor is highly variable in time and space, even at the same location. Table 1 show that this factor varied between 1.0 and 1.5 on monthly averages, and can exceed 2 on daily values. It was then clear that the use of a conversion factor was not an appropriate solution to the problem.

Site	Typology	Period	n	PARTISOL/TEOM PM10 µg/m³	Mean ratio	Ratio [min - max]
Auteuil	Traffic	summer	30	43,6 / 43,8	1,0	0,9 – 1,1
		winter	30	40,5 / 36,4	1,1	0,9 – 1,3
Gennevilliers	Urban background	summer	23	24,0 / 22,0	1,1	1,0 – 1,3
		winter	30	24,5 / 18,7	1,3	0,8 – 1,7
PA 18	Traffic	summer	34	23,9 / 21,9	1,1	0,9 – 1,3
		winter	31	30,4 / 21,9	1,4	0,9 – 1,8
Vitry sur Seine	Urban background	summer	29	20,6 / 19,8	1,0	0,9 – 1,3
		winter	28	30,5 / 21,3	1,4	0,7 – 2,0
Quillebeuf	Industrial	summer	32	35,5 / 23,9	1,5	1,1 – 2,2
Le Havre	Urban background	summer	29	23,3 / 19,9	1,2	0,9 – 1,7

Table 1: Relation between TEOM and reference method for PM_{10} measurement for different sites and seasons. n is the number of daily samples.

→ Looking for an instrumental solution

The French approach has been to look for an instrumental solution.

Two technical solutions have been identified and tested: for the MP101M beta gauge, the RST module (Regulated Sampling Tube); for the TEOM, the FDMS (Filter Dynamics Measurement System).

These additional modules dry and master the particles in such a way that the measurements are comparable to the reference method.

→ Recognition as equivalent methods

As the preliminary tests shown a very good agreement with the reference method, the French community decided to validate the technology at two levels : technical and legal.

It has been decided to apply the “equivalence procedure” to these two instruments. By the end of 2004, field operations started to obtained necessary data with respect to the

European Commission "guidance on the demonstration of equivalence". These field campaigns have been carried out in France, or in Belgium in collaboration with European partners.
An example of intercomparison result is proposed in figure 5. Table 2 gives a summary of the all data.
Results for the two automatic monitoring methods show that the following meet the equivalence criteria set out: TEOM retrofitted with FDMS (for PM_{10} and $PM_{2.5}$); and beta gauge MP101M-RST (for PM_{10}). The equivalence criteria is respected without the application of correction for slope and/or intercept.
Due to the variability of test sites (in time and space) involving different composition of ambient air and meteorological conditions, it can be assumed that equivalence for equipment tested under the used configuration is valid anywhere else in France under ambient conditions.

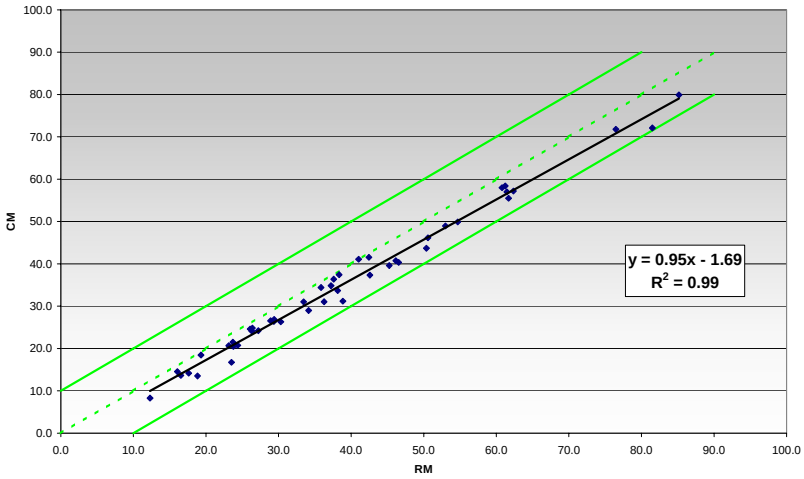


Figure 5 : Correlation between PM_{10} measurement with the reference method (RM) and FDMS (CM) in Paris (urban background site of Bobigny, winter 2005, 49 daily samples)

Candidate method	Trial site location	Time period	Equivalence criteria met?
Thermo R&P TEOM-FDMS PM_{10} series 8500 version b [a]	Bobigny (France)	25/01 to 17/04/2005	**Yes**
	Marseille (France	21/12/2005 to 13/04/2006	
Thermo R&P TEOM-FDMS $PM_{2.5}$ series 8500 version b [a]	Bobigny (France)	25/01 to 17/04/2005	**Yes**
	Marseille (France	21/12/2005 to 13/04/2006	
Environnement SA MP101M-RST PM_{10} [b]	Bobigny (France)	25/01 to 17/04/2005	**Yes**
	Marseille (France	21/12/2005 to 13/04/2006	
	Aarschot (Belgium)	10/05 to 24/06/2006	
	Monterotondo (Italy)	24/06 to 19/08/2003 & 10/12/2003 to 11/01/2004	

(a) 1h-step time measurement
(b) 24h-step time measurement

Table 2 : field campaigns devoted to the equivalence checking

→ Implementation in the PM_{10} monitoring network

As the equivalence procedure has been conclusive, 2006 spring has been dedicated to define an implementation strategy all over the French territory, to produce validated PM_{10} measurements, including the volatile fraction of PM_{10}. The system calibrated to produce

corrected data is based on a network of reference sites, where PM_{10} are measured simultaneously with usual TEOM and TEOM-FDMS. The difference between the two measurements is used on a scale defined by the user (regional in most cases) to adjust TEOM measurements on sites where FDMS are not yet installed.

→ First results

By now, about 50 reference sites are producing real-time corrections. The first results on the whole territory show that:

- a number of high PM_{10} episodes were not seen with TEOM (as expected)
- the major hypothesis done for this correction, that is the regional validity of the correction measured on a site, seems realistic when considering sites with the same typology (urban background for the moment).
- in particular, the regional validity of the correction is remarkable when high differences between TEOM and FDMS leading to high corrected PM_{10} levels are observed.

→ In progress

However, there are still some questions and works in progress. Thus, a number of actions have been started to reach the following objectives:

- to finish the demonstration of equivalence, that is to say to obtain the official recognition of the equivalence of FDMS and MP101M-RST PM_{10} measurements with reference method;
- to improve the QA/QC procedures of these new instruments
- to establish a strategy to produce corrected $PM_{2.5}$ measurements on the whole territory, - that could justify the validity and feasibility of a similar network for $PM_{2.5}$;
- to produce data helping in the choice for the implantation of future equipment (site position, typology, PM_{10} or $PM_{2.5}$)

4 Results and trends for PM_{10} and $PM_{2.5}$ concentrations

Note : the results presented below are issued from non corrected measurement data. The volatile fraction of the particles should not been taken into account.

4.1 Basic statistics

Figure 6and 7 show the trends of PM10 concentrations since 1998. Since 2004 concentrations keep the same level; a slight increase can even be noted in 2006. The exceptional events observed in summer 2003 with the heat wave are also visible on PM concentrations.

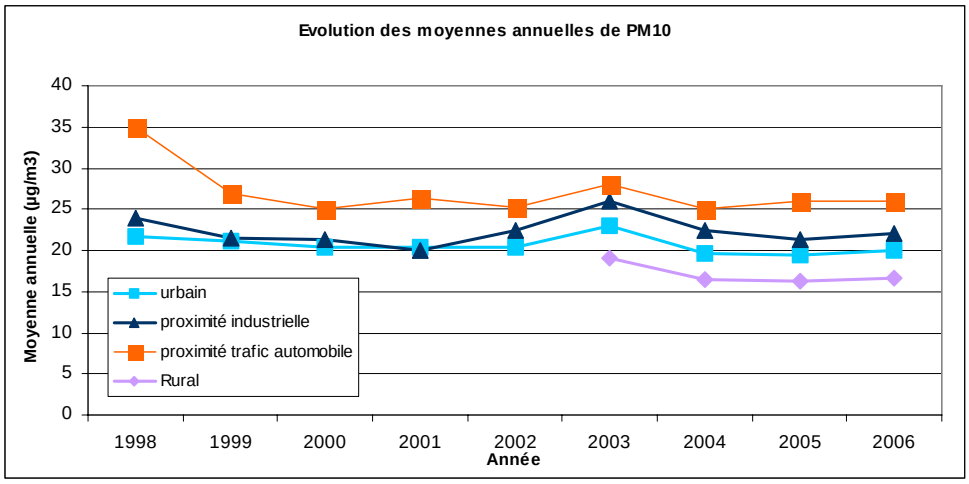


Figure 6 : PM10 concentrations in France since 1996

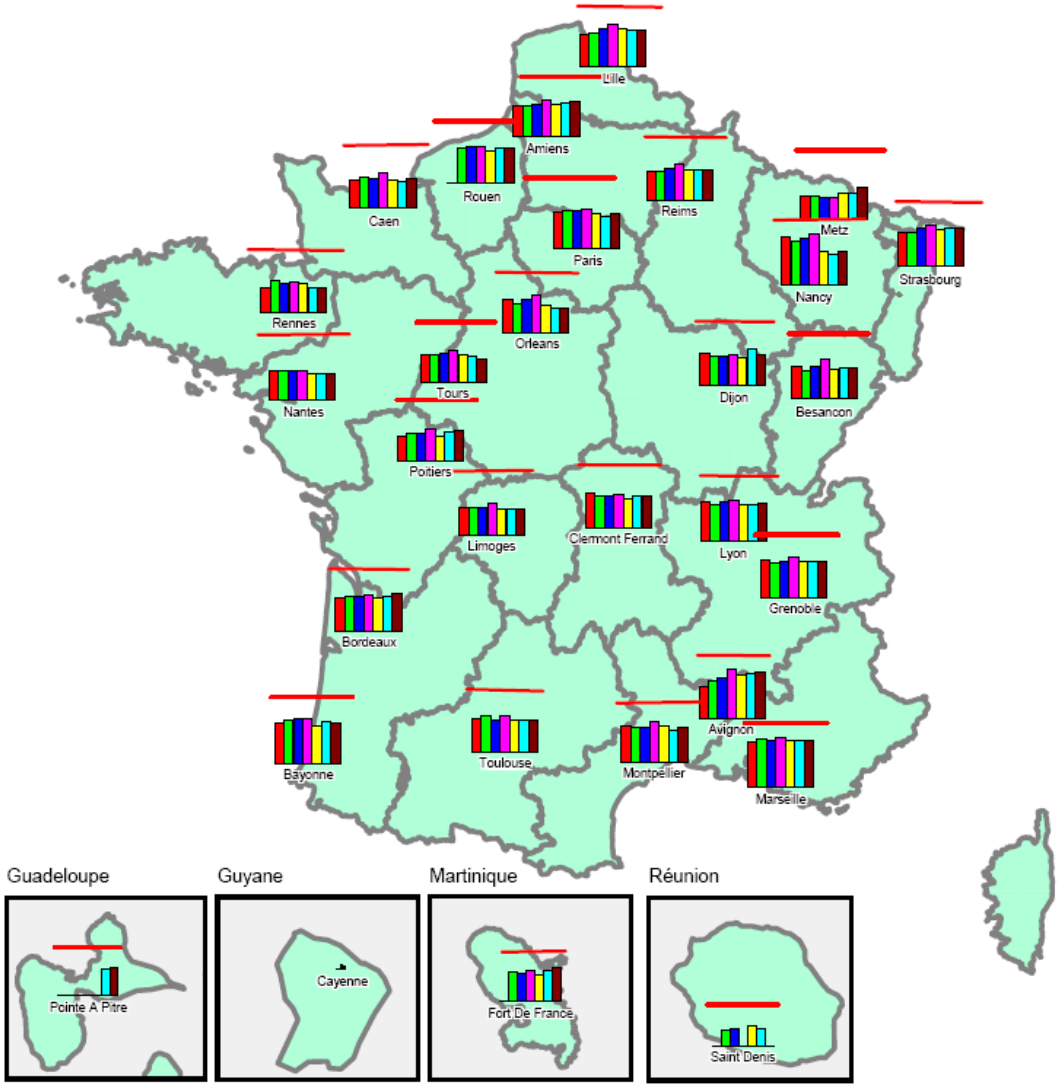


Figure 7 : PM10 concentrations in the French cities

The annual means of PM2.5 and PM10 concentrations averaged in different regions are presented in figure 8. Highest concentrations are measured at traffic stations for both pollutants.

In urban areas, highest PM2.5 concentrations (15 to 18 $\mu g/m^3$)are found in the south-East (Provence-Alpes Cote d'Azur), and in the Eastern side of the country (Alsace, Rhone-Alpes). The western part is not concerned by such concentration levels. Areas near the Atlantic side get PM2.5 concentrations 5 to 7 $\mu g/m^3$ lower than those measured at the eastern sites. The north, the Center of the country as well as Paris area get median concentrations levels (12 to 14 $\mu g/m^3$).

The East/west gradient noted for PM2.5 concentrations also holds for PM10 concentrations. Mediterranean area is still the most exposed and the Nantes areas, on the Antlantic coast the less one. Paris area and the North show quite high PM10 concentrations, generally higher than those observed in Alsace and Rhone-Alpes.

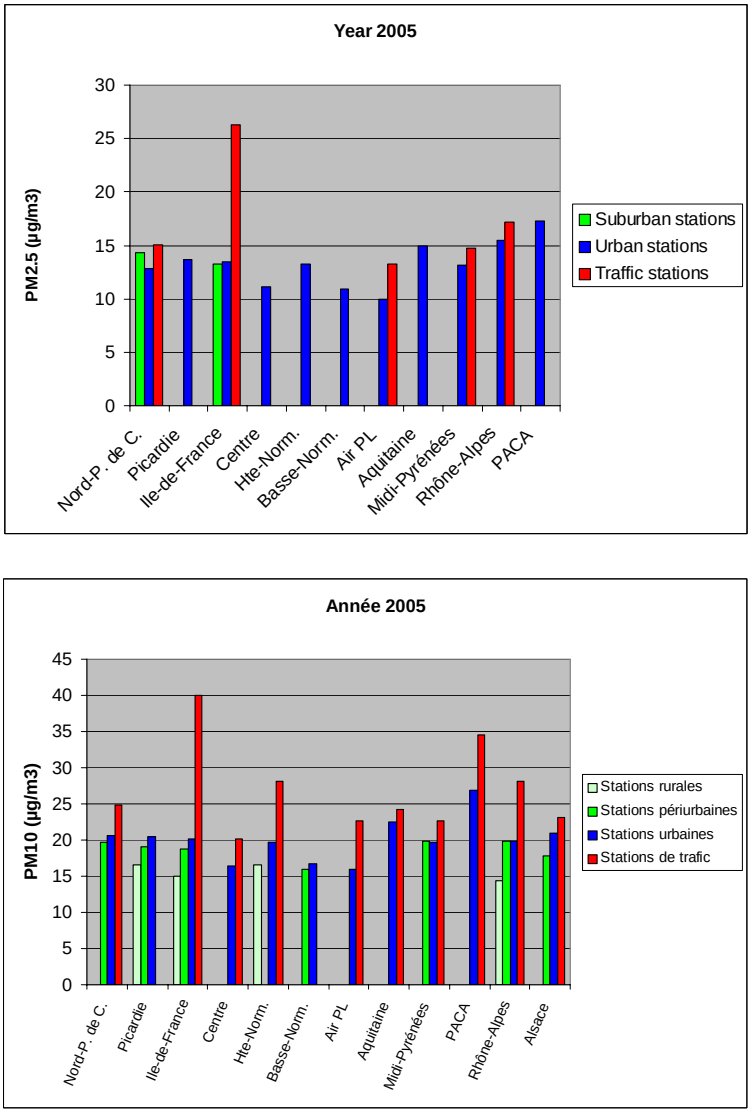


Figure 8 : PM2.5 (left) and PM10 (right) annual means in french regions

It should be noted (not shown) that PM2.5 and PM10 concentrations were particularly high everywhere during summer 2003, when the heat wave occurred in Europe. High photochemical activity could help to explain this phenomenon.

Except for this period, analysis of seasonal variability demonstrates the following statements:

- PM2.5 concentrations are higher in march/april and november/december than the rest of the year;
- They are the lowest in summer;
- Seasonal variability of urban PM2.5 is more or less sharpen depending on the cities considered (figure 9with a clear difference between summer and winter at Grenoble and Strasbourg).
- No seasonal trend is observed for PM10 concentrations. In some cases, they are higher in winter than in summer, in other situations the contrary is true. In most cases concentrations averaged over both periods are rather comparable.

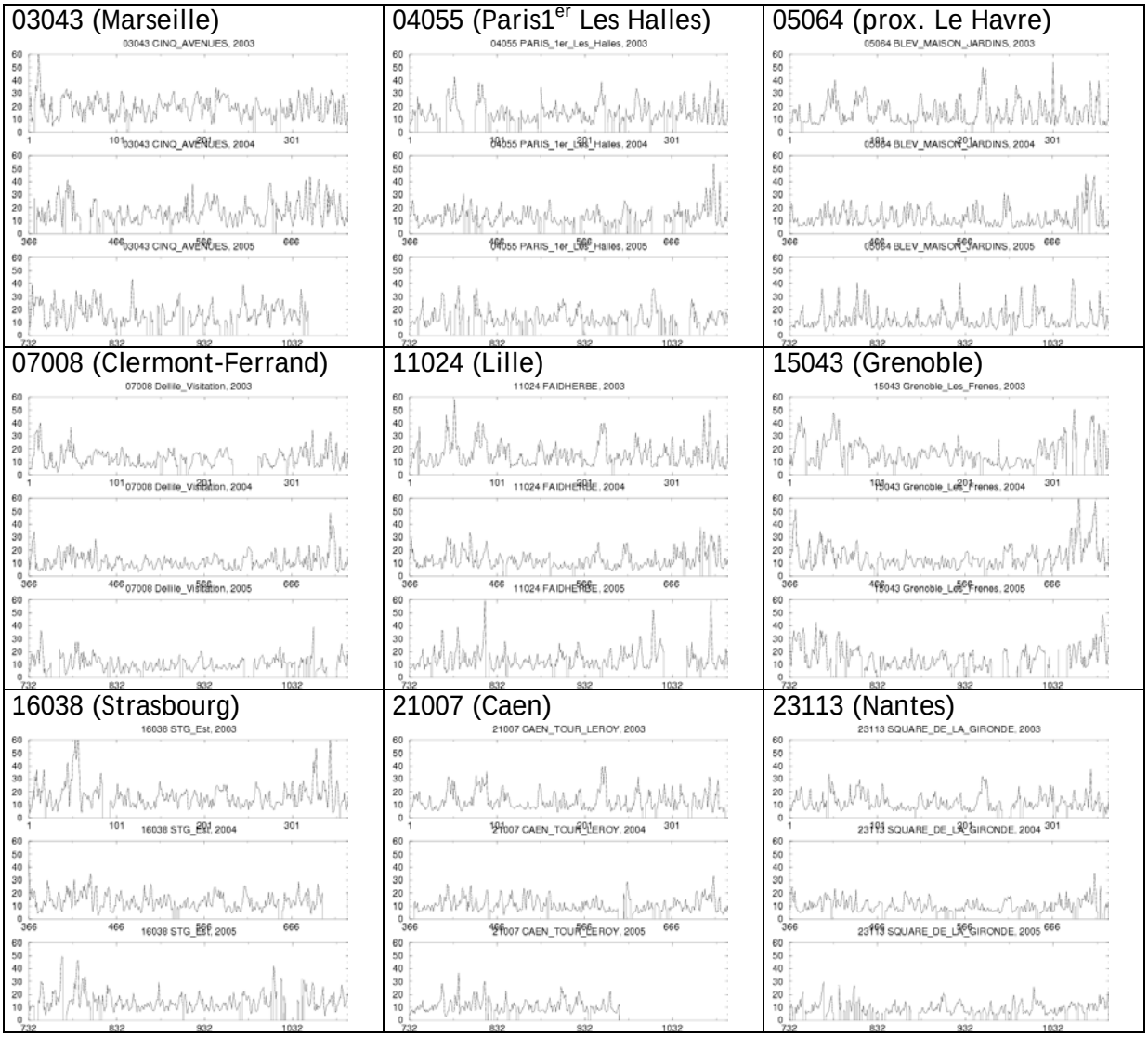


Figure 9 : PM2.5 concentrations time series for few french cities

4.2 PM characteristics

Characteristics of PM are thoroughly investigated at the Puy-de-Dome supersite (belonging to the EUSAAR network). A large panel of indicators is evaluated helping in a better understanding of the rural background composition. PM mass composition at urban and suburban sites has been analysed during the equivalence campaigns which held in Paris and in the Normandy region (see section 3.2).

4.2.1 Puy-de-Dome situation

Concentrations measured at the site show that the site is typical of the natural background. Average PM10 mass is slightly lower than 5 µg/m^3 (daily average) with minimum values lower than 1 µg m-3 (free tropospheric background) and maximum values close to 30 µg/m^3 (Saharan dust episodes).

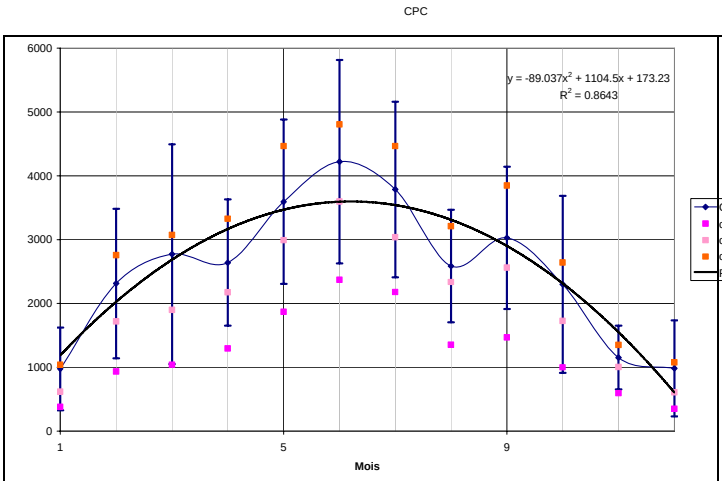


Figure 10: Max, Min and percentile (25%, 50% and 75%) of the particle number concentration at Puy de Dôme for each month

The number concentration also shows high variability, mostly linked to seasonal variability (figure 10) with highest values in summer (4000 part. cm-3 – hourly average) and lowest values in winter (1000 part cm-3 – hourly average). Such variability is obviously linked to both change in boundary layer height and in the origin of air masses advected to the site. This signal is also influenced by nucleation episodes as discussed thereafter.

The evolution of the particle size distribution is seen on a daily basis at Puy de Dôme. The mode of the size distribution lies around 50nm during background periods regardless of seasons and the distribution is mono-modal. During summer months, an additional source of particle is present, due to mixing with boundary layer air, leading to bimodal size distribution with both Aitken (30nm) and accumulation (80 nm) modes. Figure 11 shows typical daily variation during different seasons at Puy de Dôme.

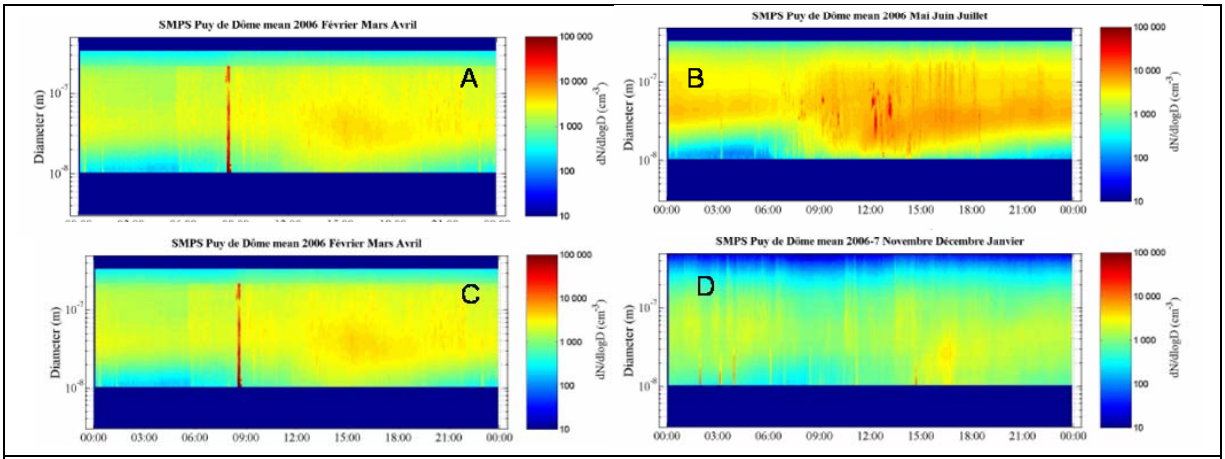


Figure 11: Average daily size distribution of aerosol particles measured at puy de Dôme for Spring (A), Summer (B) Autumn (C) and Winter (D) periods.

As mentioned earlier, the variability of the size distribution signal is also linked to nucleation episodes. The occurrence of these episodes is higher during spring time and early summer and usually takes place during clear-sky conditions, at the interface between free-tropospheric and boundary layer air masses.
The mechanisms of particle nucleation at Puy de Dôme has been reviewed by Venzac et al.(2007) showing that most of the episodes are connected with increases in the ion background in the atmosphere. Nucleation events are likely to be one of the most efficient processes controlling particle concentrations in the free troposphere over Europe.

The chemical composition of the aerosol at Puy de Dôme has been extensively studied in the framework of CARBOSOL (FP5-EU) completing the routine measurements of Puy de Dôme. Routine filters are collected since spring 2003 and analysed by ion chromatography

for their inorganic fraction and by thermo-optical methods for their OC and EC fractions. Note that sampling is not performed controlling positive or negative artefacts. Work is under way to estimate these artefacts (work performed in the framework of EUSAAR – FP6). Table 3 summarises the measurements performed at the Puy de Dôme (weekly averages).

	Cl-	NO3-	SO4--	oxalate	Na+	NH4+	K+	Mg++	Ca++	OC	EC
Number of samples	156	96	159	159	154	159	159	126	129	147	146
minimum	<dl	0,0006	0,0734	0,0022	0,0009	0,0128	<dl	<dl	<dl	0,1199	<dl
quartile 25	0,0010	0,0395	0,5713	0,0168	0,0110	0,1190	0,0088	0,0015	0,0053	0,3320	0,0401
median	0,0018	0,1251	1,0107	0,0294	0,0191	0,2029	0,0133	0,0034	0,0120	0,5328	0,0561
quartile 75	0,0037	1,0582	1,4764	0,0487	0,0363	0,2952	0,0203	0,0072	0,0332	0,7831	0,0862
maximum	0,0593	15,7277	4,4532	0,2020	0,2635	1,3201	1,5855	0,0984	0,6209	1,5582	0,5531
Mean	0,0036	0,8129	1,1275	0,0362	0,0282	0,2267	0,0274	0,0081	0,0397	0,5795	0,0831
variance	0,0063	1,8451	0,7646	0,0280	0,0291	0,1593	0,1263	0,0139	0,0836	0,2973	0,0868

Table 3: weekly averaged particle composition for inorganic and organic components of the aerosol at puy de Dôme. Measurements started in spring of 2003.

The weekly averaged concentration is close to 3 µg/m^3, slightly lower than PM10 measurements performed with cascade impactors. The difference is due to low efficiency of supermicron particles with low-volume collectors and to loss of the volatile fraction (both organic and inorganic – NO3NH4 -). On average, the OC fraction accounts for less than 1 µg/m^3 although weekly averages can be higher than 1.5 µg/m^3 .
Average proportions of inorganic and organic compounds are shown in Figure 12.

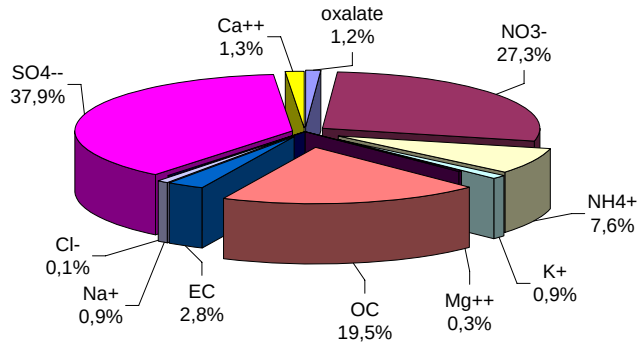


Figure 12: average fractions of inorganic and organic components of aerosol at puy de Dôme. Note that the NO3- fraction

Measurements with cascade impactors performed during 2006 and 2007 confirmed these proportions, showing both mono-modal, bimodal and trimodal size distributions depending on the events. Sulfate is the major component of the aerosol at puy de Dôme while the fraction of organic material varies from 15% up to 40% depending on the air mass.

4.2.2 Urban PM speciation

The field campaigns organised to assess the volatile part lost by automatic devices allowed to described the PM10 mass composition in winter and in summer for different types of sites. Figure 13 shows some of these results.

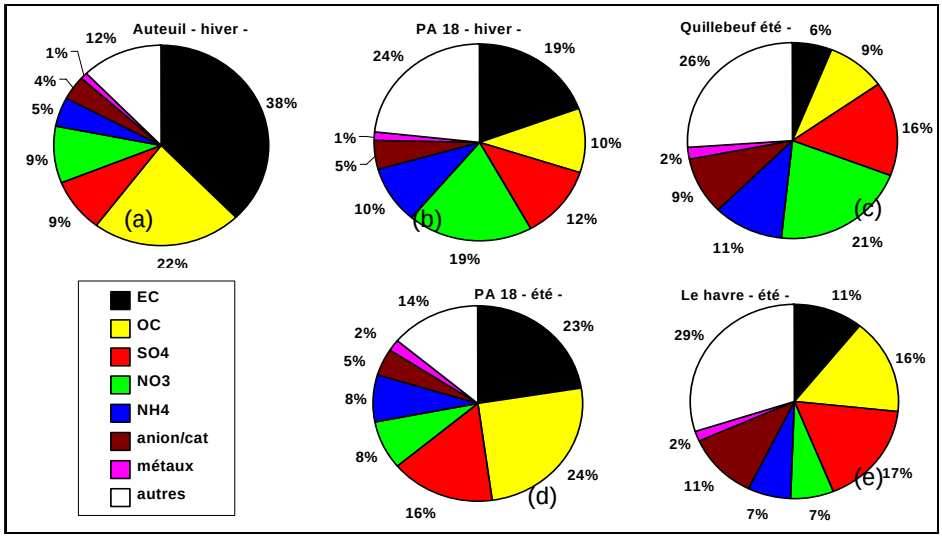
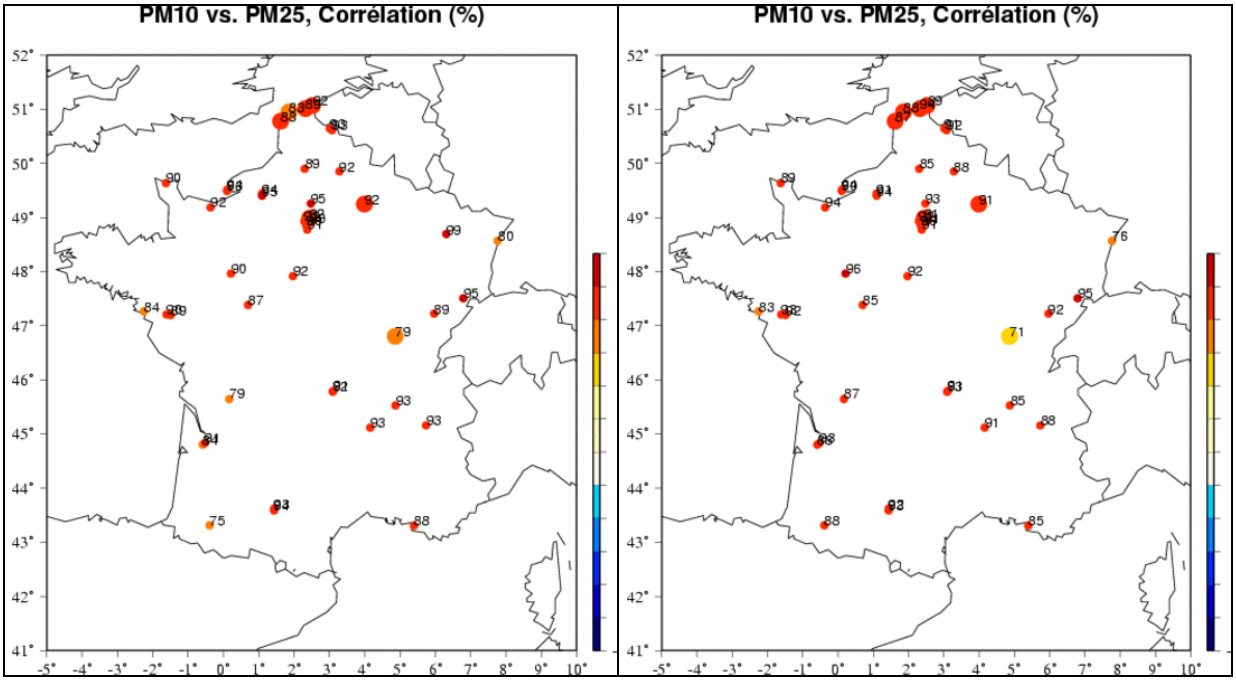


Figure 13 : PM10 mass composition for different situations in winter (a and b) and in summer (c to e)

4.3 Relationships between PM_{10} and $PM_{2.5}$ concentrations

4.3.1 Correlation between PM10 and PM2.5 mass concentrations (figure 14)

Correlation between PM10 and PM2.5 mass concentrations is excellent everywhere, higher than 75% (minimum in the South-West region). The score of 85% is often exceeded, with better results in winter or in summer depending on the geographic area. Correlation are higher in winter in Paris areas, in the North and in the Rhone-Alpes (Center-East) region.



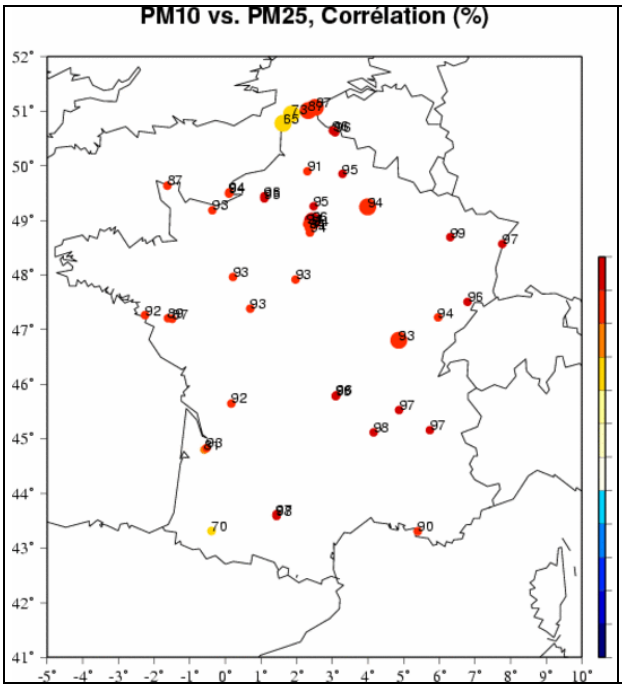
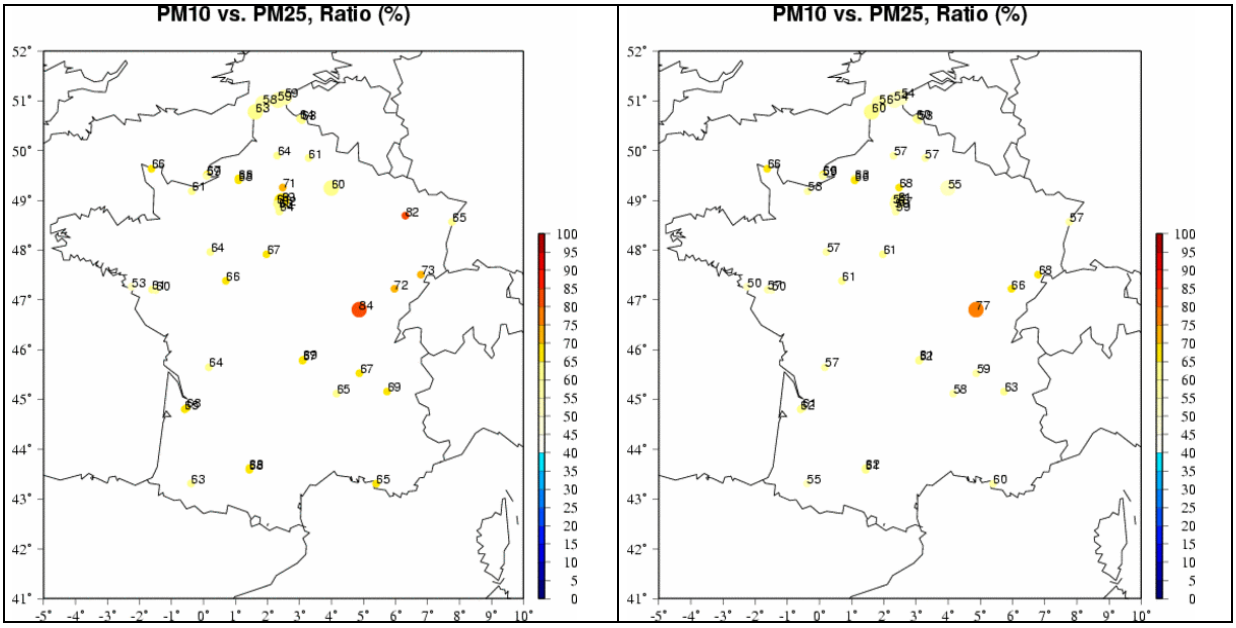


Figure 14 : Correlation between PM10 and PM2.5 concentrations. Suburban stations (large circles) and urban (small circles) are drawn for the 2003-2005 period (top left); summers 2003 to 2005 (top right) and winter 2003 to 2005 (bottom left).

4.3.2 Ratio PM2.5/PM10

Figure 15 shows ratios PM2.5/PM10 mass concentrations. This indicator varies depending on the geographic area considered. Highest ratios (72 to 84%) are seen in the eastern part of the country (Loraine, Vosges, Bourgogne), excepted in Alsace (65%). In Paris area PM2.5/PM10 ratio varies from 64 to 71%. It is quite homogeneous in the mid-south of France: from 63 to 69%. Ratios are lower at sites located in the western and northern coasts (53% at Nantes and 58_59% at Calais, Dunkerque).
PM2.5/PM10 ratio is almost higher in winter than in summer with a 5 to 10% increase compared to the average mean.



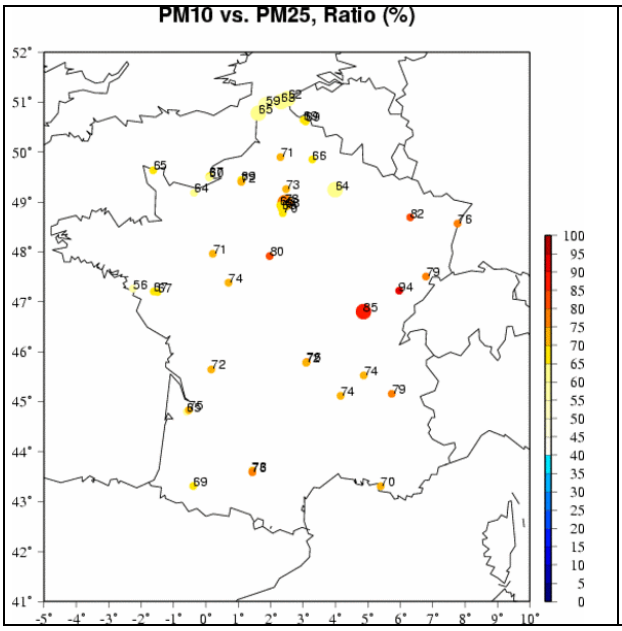


Figure 15 : PM2.5/PM10 ratios. Suburban stations (large circles) and urban (small circles) are drawn for the 2003-2005 period (top left); summers 2003 to 2005 (top right) and winter 2003 to 2005 (bottom left).

5 PM climatology in France

The Puy-de-Dome rural supersite helps to investigate the long range transport contribution to PM concentrations. It is clearly detected during advection of anthropogenic, marine and Saharan dust air masses to the sampling site. The contribution of the specific emission area is difficult to estimate without detailed modelling of transport mechanisms. However, results from cascade impactor measurements can provide a first estimate of the contribution of long-range transport, especially for marine and Saharan dust episodes. The contribution of marine aerosol (from the Atlantic Ocean) to the free troposphere in France remains extremely limited far from the Coastal areas. On average, we can consider that this contribution never gets above 0.1 µg m-3, that is, at the most, a few % of the total PM10 mass.
On the contrary, Saharan dust episodes have potentially a higher impact on the average PM10 mass measured at puy de Dôme. During Saharan dust episodes, the contribution of dust (mostly found onto large particles) reaches a few µg m-3 (2-6), and contributes to a substantial fraction of PM. It should be noted that the dust is mostly formed by Ca-containing material but also provides a surface for condensing HNO3 vapour that in turn will contribute to increasing PM. The contribution of NO3 condensed onto large particles can reach several tens of % of the total aerosol mass.
Finally, high concentration of particles is also measured during advection of air masses from Northern Europe (including the Paris area). These episodes are most intense following cold front when cyclonic conditions favour Northerly winds to the site. However, a precise contribution of long-range transport from anthropogenic activities is difficult to estimate due to the fact that the contribution of boundary layer air in embedded in the bulk filter (and impactor measurements). We can estimate that long range transport from Northern Europe increases the aerosol mass at the Puy-de-Dome supersite by a few µg/m^3 and is mostly composed of nitrate, sulphate and organic material.

Model experiments allow to represent the impact in France of particulate pollution episodes mainly due to inorganic particulate formation. The CHIMERE model [Bessagnet , 2005] has been developed to simulate primary and secondary PM concentrations. Its results have been assessed against observations and during European model intercomparison exercises [Cuvelier, 2006]. Provided that the emissions are reliable, confidence is built in model results especially for inorganic compounds. Thus, CHIMERE is able to correctly

detect ammonium nitrate episodes . This capacity is used in the PREV'AIR system, which is the french air quality forecasting platform (figure 16).
An example of ammonium nitrate episode simulated by CHIMERE is given in figure 17. Such episodes occurs several times a year, especially in spring because of favourable meteorological conditions and higher emissions of ammonium in the northern part of Europe.

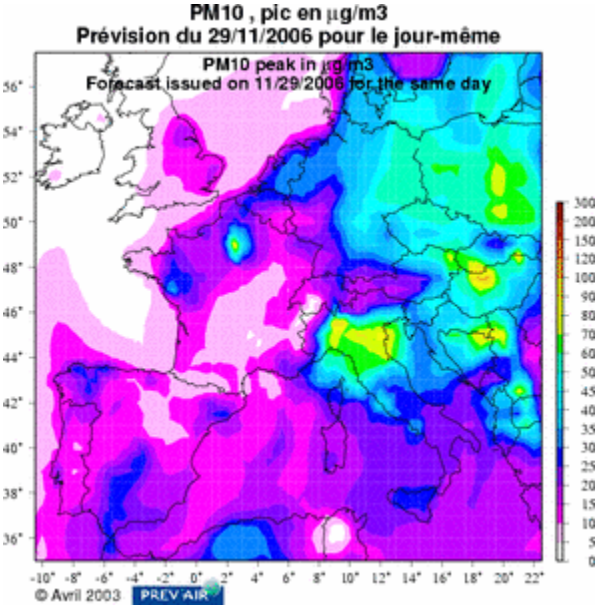
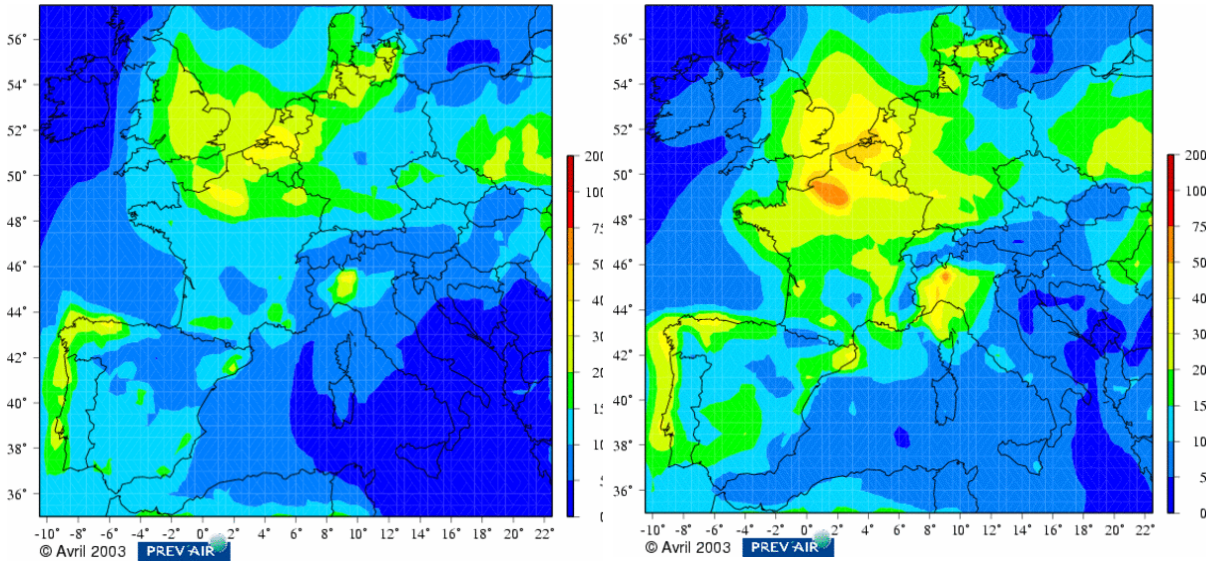


Figure 16 : PREV'AIR PM10 forecast

13 mars 2006 **14 mars 2006**



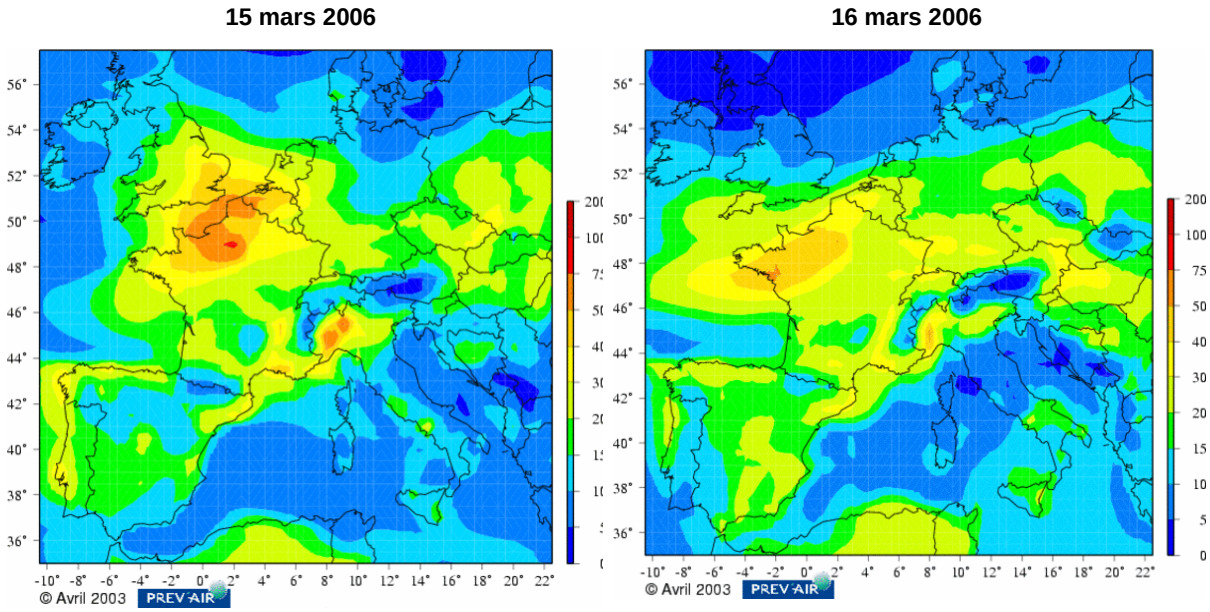


Figure 17: Ammonium nitrate episode simulated with the CHIMERE model (march 2006)

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