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Size distribution of fine Particles in Stack emissions of a 600-MWe coal-fired Power Plant

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Abstract

Monitoring emissions of particles with a diameter smaller than 10 μm (PM_{10}) has become of growing interest. However, measuring such particles is rather difficult. It appears as a result necessary to develop and assess measurement methods for the monitoring of PM_{10} from stack and fugitive emissions. The aim of the GAEC program [Granulométrie des Aérosols dans les Emissions Canalisées: Aerosol size distribution from stack emissions] is double: to develop monitoring methods and improve knowledge on fine particulate stack emissions. Three institutes were involved in the program: Séchaud Environnement (formerly LECES), INERIS and CERTAM.

This paper presents the main results of the evaluation of a cascade impactor (Johnas), aerosol size distribution measurement techniques (ELPI Dekati, FPS, SMPS TSI) applied on size characterization of fine particulate matter in stack emissions of a 600-MWe coal-fired power plant.

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Introduction

Emissions of particulate matter with an aerodynamic diameter smaller than 10 μm (PM_{10}) have become of growing interest. However, monitoring such particles is rather difficult. In order to address this issue, a standardisation frame has been defined: a standardisation working group dealing with PM_{10} and $\text{PM}_{2.5}$ emissions from stationary sources has been set up in 2003 (ISO TC 146 SC1 WG 20). The GAEC program presented here aimed at developing measurement techniques and generating experimental data on fine particulate emissions from stationary sources. Such developments may, for instance, help plant operators in providing more accurate data in the framework of the European Pollutant Emission Register (EPER). This study is supported by the French Environment Agency (ADEME) and the French ministry of environment (MEDAD) and deals more precisely with aerosol size distribution determination from stack emissions: PM_{10} , $\text{PM}_{2.5}$, PM_1 and $\text{PM}_{0.1}$. The working partners involved in this program are: Séchaud Environnement (previously LECES), INERIS and CERTAM.

A first step consisted of a state of the art of the health impact and the available measurement techniques. Some of the techniques have then been selected to be set up and assessed on site during measurement campaigns.

The first campaign took place in September 2005 in a cement plant; the results of this study have been presented at the Dustconf International Conference in Maastricht, the Netherlands in April 2007 (Fraboulet et al, Dustconf 2007). The present paper deals with the results of the second campaign that was carried out in March 2006 at a 600-MWe coal-fired power plant.

Experimental conditions

Measurement site

The sampling campaign took place at the EDF coal fired power plant in Cordemais, France. A schematic of the plant including the energy production and gas treatment unit is presented in Figure 1. The sampling point was located after the gas treatment unit. This unit is equipped with an electrostatic precipitator (ESP) dust filter and a desulphurisation unit (Figure 2).

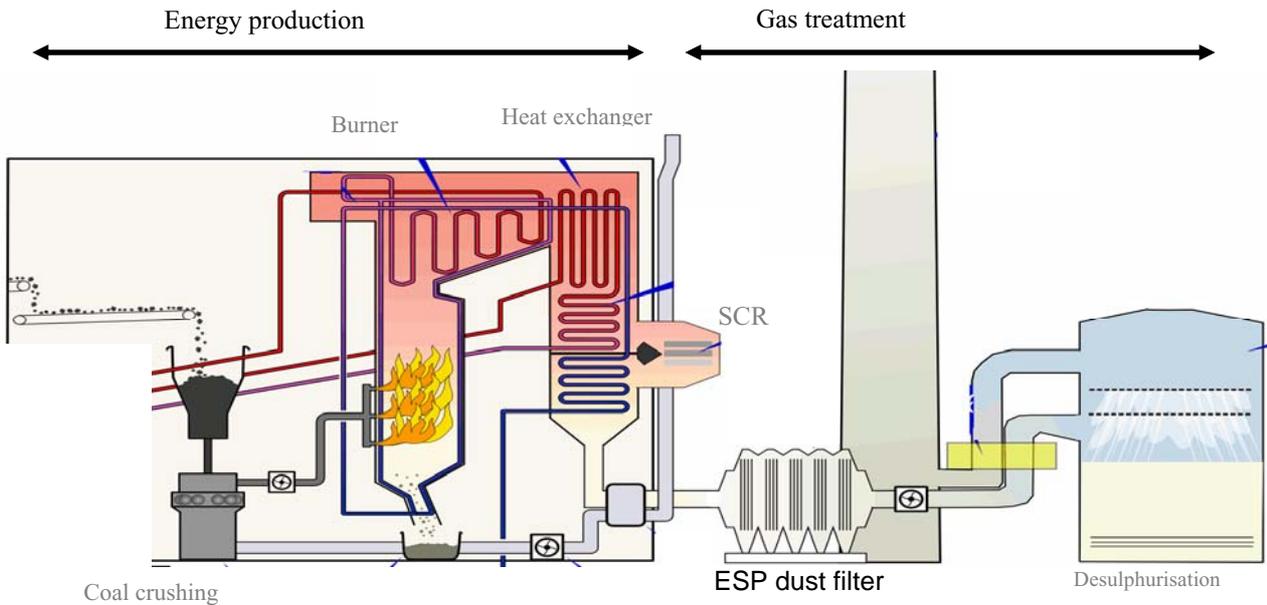


Figure 1 : Schematic of the Cordemais power plant process.

The desulphurisation consists in passing the gases through a wet scrubber where a lime (CaCO_3) saturated washing solution is sprayed. This leads to the formation of calcium sulphite (CaSO_3) which is afterwards oxidised into gypsum (CaSO_4). The clean gas is then emitted through the chimney and gypsum is collected to be sold to the cement or paper industry.

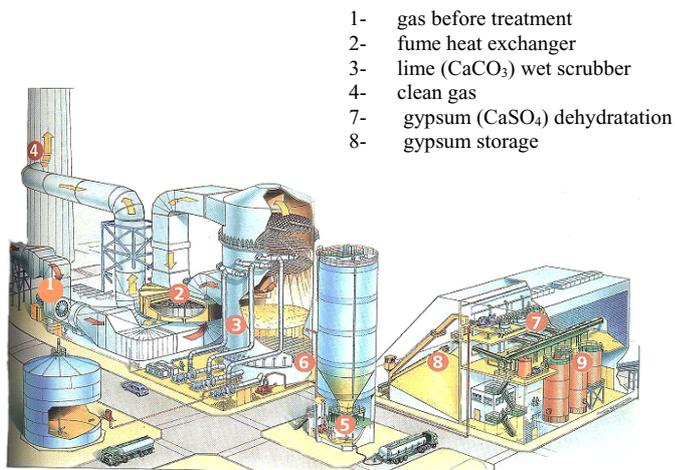


Figure 2 : Schematic of the desulphurisation unit

During the campaign, the power plant was operating at a power comprised between 480 and 600 MW. the following table presents the operating conditions associated. It is interesting to stress the fact that this stack showed very high values of gas velocity associated to relatively low TSP concentrations. Such conditions are likely to penalise the judgement on measurement techniques since they are close operating condition limits.

Table 1 : Sampling conditions

Gas velocity	34-43 m/s
Humidity	6-9 %
Temperature	91-94°C
O2 content	7-8 %
CO2 content	11 %
TSP	1-10 mg/m ³ ₀

Measurement techniques

The following techniques were selected to be set implemented during the campaign:

Manual methods:

- JOHNAS cascade impactor (John et al, 2003)
- TSP reference gravimetric method

Automatic methods: Aerosol size distribution

- ELPI : electronic low pressure impactor connected to a sampling and dilution probe called FPS (PM_{2.5} inlet), using smooth or sintered impaction plates
- SMPS : mobility analyser connected to a sampling and dilution probe called FPS (PM_{2.5} inlet)

Results and discussion

Manual techniques

Table 2 presents the comparison between the operating conditions and the specifications of the manual measurement techniques. This confirms the fact that the sampling conditions in term of concentration and gas velocity reach the limits of the technique specifications.

Table 2 : Comparison of the sampling operating conditions with the manual measurement technique specifications

Specifications	Gas velocity (m/s)	Temperature (°C)	Humidity (%)	Concentration (mg/m ³ ₀)
Johnas impactor	<35-40	250	<25	1-100
reference method		250	<25	>1
sampling conditions	36-39	91-94	6-9	1-10
agreement	limit	yes	yes	limit

Table 3 : Output power of the plant during the campaign

Trial	Power (MWe)
M4	600
M5	500 +/-60
M6	4-600

The filters collected with the two techniques showed a yellowish colour, which was very similar to the gypsum generated as a by-product of the desulphurisation. This suggests that the emitted aerosol could present a relatively high proportion of gypsum; this could mean that the nature of the emitted aerosol is more linked to the desulphurisation than to the coal combustion itself. However, this hypothesis would need to be confirmed by further analyses.

Concentrations measured were comprised between 1 and 6 mg/m³ i.e. in the low range of the specification of the measurement techniques. The small number of sampling trials performed is insufficient to conclude on correlation between the concentrations emitted and the output power of the plant.

The PM₁₀/PM_{2.5} ratio indicated that 90% of the mass of PM₁₀ was due to PM_{2.5}. These are very preliminary that would need to be taken into account with cautious and confirmed by microscopy analysis of the impaction filters in order to validate the distribution of the particles on the impaction stages and dismiss any risk of particle bounce.

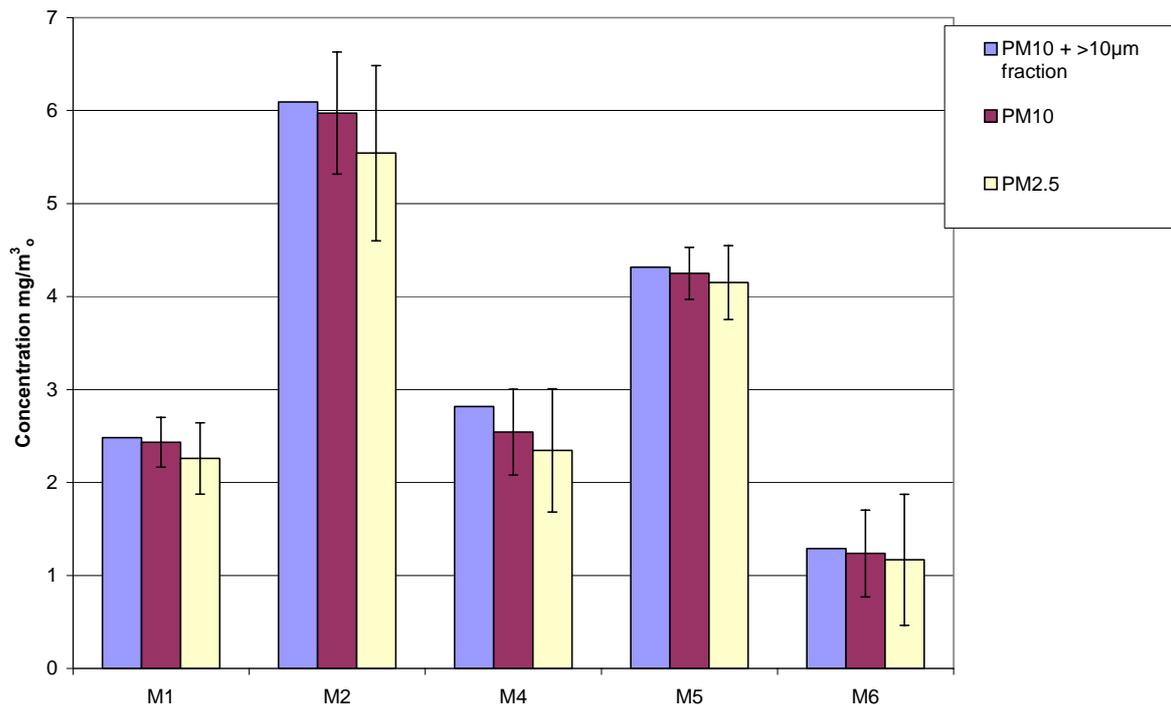


Figure 3 : PM₁₀, PM_{2.5} and sum PM₁₀ + > 10 µm fraction; mass concentrations measured with the Johnas impactor

Automatic techniques

The main interest of using automatic techniques for the determination of aerosol size distribution is to perform online monitor of the aerosol characteristics. This is especially interesting to assess process stability or to characterise process operating transitions.

The following figure presents the evolution of the operating output power as well as the number of particles measured by the ELPI. It shows that a decrease of the power of the boiler causes a decrease of the number of emitted particles. A similar trend could be observed with the SMPS.

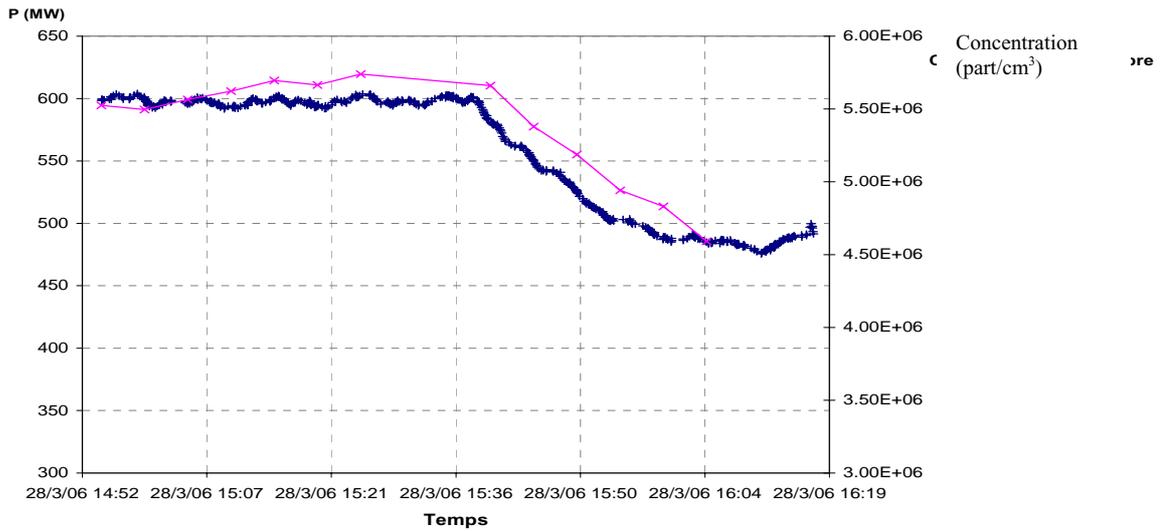


Figure 4 : Output power and particle number concentration (ELPI) monitoring

During the previous campaign that took place in a cement plant (Fraboulet et al, Dustconf 2007), the SMPS was used together with the ELPI equipped with smooth impaction plates, in order to characterise the aerosol (Keskinen et al, 1998; Marjamaki et al, 2000). For a density of 3, as shown in Figure 5, a good agreement of the SMPS and ELPI curves could be obtained in the range 0.1 to 1 µm. However, for particle with a diameter smaller that 0.1µm the ELPI was overestimating the amount of measured particles.

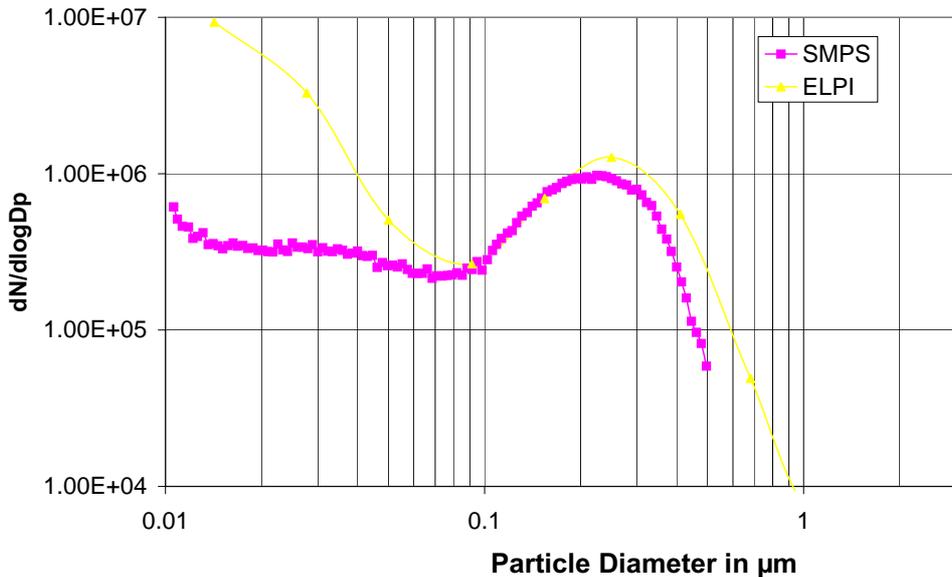


Figure 5 : Comparison of cement plant aerosol size distribution measured by the ELPI and the SMPS for a density value of 3

The most likely hypothesis was that particle bounce may cause this overestimation of the ELPI comparing to the SMPS. The use of smooth impaction plates, when characterising mineral particles, could indeed lead to particle bounce; a solution to this issue could be the use of sintered impaction plates.

In order to validate this hypothesis, the ELPI equipped with either smooth or sintered impaction plates was jointly used with the SMPS during the power plant campaign. The following figure shows the results obtained for a value of density of 1.5. It appears, as expected, that when using smooth impaction plates, the ELPI overestimates the particle number in the range of diameters smaller than 0.1µm. However when using sintered impaction plates, the size distribution obtained with the ELPI and the SMPS are in good agreement in the range of particles with a diameter bigger than 0.06 µm. In the

range of particles with a diameter smaller than $0.05\mu\text{m}$, the ELPI and the SMPS showed different size distributions. This may be due to the occurrence of a volatile unstable nucleation mode in the lower size range. In some cases, the use of low pressure to accelerate ultrafine particles in the area of the last impactation plates of the ELPI can lead to an evaporation of volatile compounds and as a result to an underestimation of the nucleation mode.

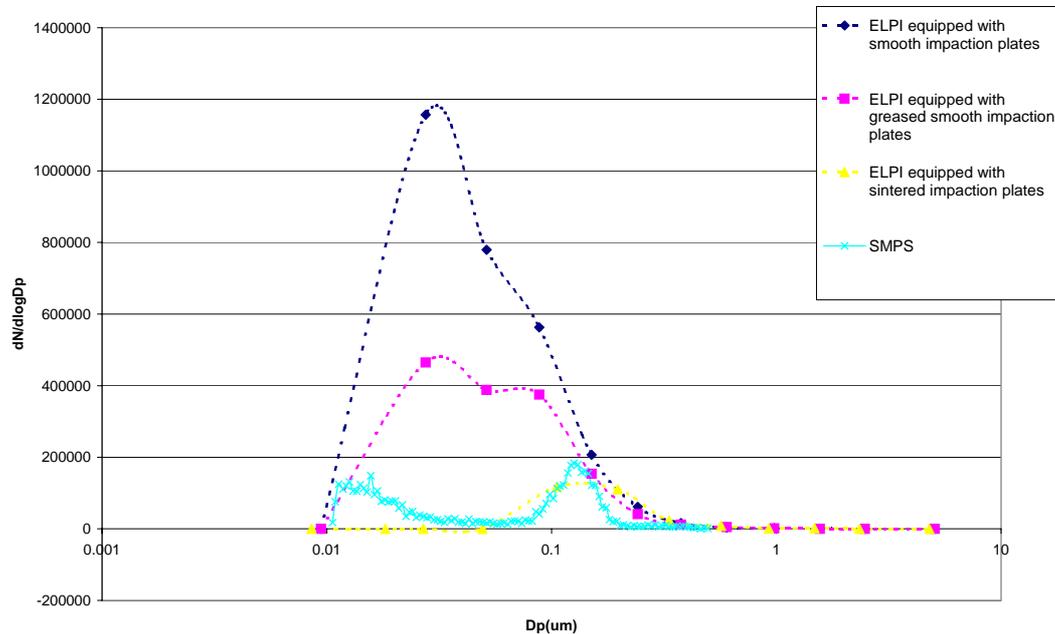


Figure 6 : Comparison of aerosol size distributions determined with the ELPI and the SMPS

CONCLUSION

This campaign took place at a power plant stack, the sampling points being located after an ESP dust filter and a desulphurisation unit.

The use of manual and automatic techniques led to the following results :

- TSP concentrations were comprised between 1 and 6 mg/m^3 , the $\text{PM}_{2.5}$ fraction represented 90% of PM_{10} .
- The number of particles emitted is correlated with the operating output power, monitoring of particle number appears then as an interesting tool for process monitoring.
- The use of the ELPI equipped with sintered impactation plates reduces the risk of particle bounce and leads to a good agreement with the SMPS.
- In the lower size range, a nucleation unstable mode could be observed.

Besides, the collected samples showed a yellowish colour characteristic of the gypsum generated as a by-product of the desulphurisation unit. This observation may indicate an important mineral proportion of the aerosol.

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