CONTROL OF FUGITIVE PARTICULATE EMISSIONS IN THE CERAMIC INDUSTRY

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ABSTRACT

Particle emissions into the atmosphere are one of the major impacts of the ceramic industry, by both channelled sources (stacks) and fugitive sources. Fugitive pollutant emissions (i.e. emissions which reach the atmosphere without channelling through a duct) have traditionally received little attention from either legal regulations or technical studies. However, this situation is changing given the relative importance of these fugitive emissions in certain industrial activities. In the particular case of the ceramic industry, interest is centred on the fugitive emissions of particulates, particularly in the raw materials preparation stages (e.g. manufacture of spray-dried granules). No standard measurement methodology has been established in the European Union for the control of these types of fugitive emissions, though different actions are currently being undertaken in this sense.

On the other hand, in the most recent EU legislation on air quality, particular attention is paid to the presence of particles, especially the PM_{10} fraction (particles with an aerodynamic diameter below 10 µm). The present study has developed a methodology for the control of fugitive particle emissions, based on the determination of the PM_{10} concentration in the industrial environment.

1. INTRODUCTION

1.1. LEGISLATION ON AIR QUALITY

The transposition of Directive 1999/30/EC, by approval in July 2002 of Royal Decree 1073/2002, modified Spanish legislation on the evaluation and management of environmental air quality in regard to particles and other pollutants. In the case of particulate materials this legislation no longer legislates settleable particles and total suspended particles; instead, based on human health protection criteria, following the recommendations of the World Health Organisation, it has focused on fixing air quality limits regarding PM_{10} particles, and more recently $PM_{2.5}$ particles.

In Spain the publication of Royal Decree 1073/2002 has not only meant an important change in the parameter to be measured in regard to particulates (going from TSP to PM_{10}), but also a greater restriction in the allowable limit values, which have gone from 150 μgTSP/m^3 to 40 μgPM_{10}/m^3[1].

The concentration limits of environmental PM_{10} particles, defined by the Royal Decree 1073/2002, are summarised in Table 1.

	PHASE 1	PHASE 2 (PENDING REVIEW)
Entry into force	2005	2010
Average of 1 civil Year	40	20
Average in 24 Hours	50	50
Max no. of limit oversteppings of the daily average (d/year)	35	7

Table 1. PM_{10} (µg/m^3) limits set by R.D. 1073/2002.

The most recent air quality studies conducted in the Castellón ceramic district[2-4] indicate that keeping the PM_{10} levels within the limits set in Table 1 is one of the most critical aspects in this regard. Therefore, to meet regulatory air quality requirements it is essential to control adequately the area's particle emission-generating activities in regard to both channelled and fugitive dust emissions, which include those of the ceramic industry.

One of the main problems of setting a single limit value is the definition of the location of the sampling points, so that they will be representative of the zones where the population might be exposed to these particulates. For this reason, these air quality limits would not seem to be applicable to industrial environments, either for their representativeness or for being a realistic objective[2].

1.2. FUGITIVE EMISSION CONTROL IN THE CERAMIC INDUSTRY

The main raw materials of the ceramic industry are of a powdery nature. Consequently, raw materials processing generate suspensions of the finest fractions in the surrounding air, particularly in transport, storage, mechanical treatments and subsequent handling operations of these raw materials. When these operations are confined (specific enclosures of a facility) the suspended particles are usually eliminated by extraction systems, collected in a treatment system, after which the cleaned stream is exhausted through a channelled source. However, not all suspended particles are captured by the extraction systems, nor are all the operations confined – some may also take place directly in the open air; non-channelled emissions of particles into the atmosphere are thus generated. These emissions are known as emissions from diffuse or fugitive sources, or simply fugitive emissions.

Traditionally legislation has proposed emission limit values for channelled sources, normally expressed as concentrations or emission factors. However, particle emissions through fugitive sources have not received the same legal treatment, and normally administrative regulations have been based on establishing a series of preventive measures in industrial processes, such as requiring asphalt paving of roads, keeping the materials stored outdoors wet or covered, etc.[5].

An extensive literature survey has shown that the methods for the control of fugitive suspended particle emissions in open-air activities, of an industrial as well as mining nature, can be classified in two groups:

a) Methods based on environmental **particle concentration measurement**:

- *Advantages:* Hardly any data treatment is required, the obtained values are direct. It measures a parameter (environmental concentration) directly related to the ones established in the legislation on air quality and industrial health (for example, the TLVs of the American Conference of Governmental Industrial Hygienists). From the point of view of industrial self-control it has the advantage of relative simplicity.

- *Disadvantages:* The overall emission of a facility is not quantified (environmental concentrations are measured), although it can be calculated by reverse modelling methods. It is necessary to have appropriate scientific equipment (capturing instruments and precision balances). The problem of the location of the apparatuses appears when the measurements are to be made (whether measuring occurs at internal points or on the perimeter of the facility). Hardly any reference values for this parameter are available in industrial environments. Safe, unencumbered locations with an electric power supply are required, which in practice involve a severe restriction.

b) Methods based on **estimating the mass flow** of fugitive particulate material emissions. These can, in turn, be divided into:

1. Methods based on the use of **emission factors** or **semi-empirical equations**[6]

- *Advantages:* No experimentation is required, and they are very useful for estimating emissions or efficiencies on an initial project. Little preliminary information is required, only some experimental parameters (equations).

- *Disadvantage:* They display high uncertainties, since the emission factors and conditions in which they have been obtained do not always match the actual conditions in the facility to be studied (material, machines, atmospheric conditions, etc.). The main difficulty is that at present little information is available on the specific emission factors of the ceramic industry and the quantitative efficiency of the applicable corrective measures.

2. Estimation of mass emission by **(reverse) dispersion models**[7]

- *Advantages*: The emission is calculated from experimental data on the facility. They enable readily simulating different situations by modifying some of the variables assumed by the model.
- *Disadvantages*: A relatively complex mathematical treatment is required. Instrumentation is required to obtain environmental data, of both an environmental and meteorological nature, in addition to an appropriate calculation program. These methods have hardly been standardised, although there are some initiatives in this direction. In the literature search conducted, we found a German standard from 2004 (VDI-4285)[8], and there are proposed standards, such as the one that this is being developed by the European Committee for Standardisation CEN/TC 264/N 863*: Fugitive and diffuse emissions of common concern to industry sectors-Fugitive dust emission rate estimates by Reverse Dispersion Modelling*, in whose workgroup ITC has participated.

2. OBJECT AND SCOPE OF THE WORK

With the premises set out in the introduction to this study, which is part of a larger project conducted by ITC and CIEMAT on particle emission control in the ceramic industry, this paper presents the results obtained in the development and evaluation of the usefulness of applying a fugitive emission control method to these facilities, based on the measurement of environmental particle concentrations.

The study has the following specific objectives:

1. To fine-tune a methodology for measuring particle concentrations in fugitive emission (PM_{10}), using gravimetric and continuous measurement equipment for this purpose.
2. To apply this methodology to facilities for the manufacture of spray-dried granules and ceramic tiles, to study whether the method offers a repetitive response in similar industrial measurement scenarios.
3. To determine PM_{10} levels and to evaluate these in order to draw conclusions concerning the environmental impact of the studied facilities. To verify the applicability of this type of measure for control of particulate emissions from fugitive sources in the ceramic industry.
4. To determine the possible use of this measurement methodology for controlling the degree of improvement achieved after the introduction of corrective measures.

3. EXPERIMENTAL DEVELOPMENT

3.1. EQUIPMENT FOR MEASURING ENVIRONMENTAL PARTICLE CONCENTRATIONS

The equipment proposed for the measurement of environmental particle concentrations for fugitive emission control is that used in air quality control. These systems can be divided into two types, according to the sampling period: discontinuous and continuous samplers.

Discontinuous samplers are based on the measurement of diffuse particle concentrations in a certain time range; the measurement time is usually 24 hours. The most widely used measurement method is the gravimetric method.

The measurement principle of a gravimetric sampler consists of taking in a known air flow that is held during the sampling period. This air flow crosses an appropriate particle cut-off head for the type of particle to be measured and then passes through a filter where the particles present in this stream are retained. The particulate concentration (µg/m^3) is determined by the difference in weight of the filter, dividing this by the total volume of sampled air.

European standard UNE-EN 12341 proposes, as a reference sampler of PM_{10}, a gravimetric apparatus with certain technical characteristics[9]. Samplers that do not have these technical characteristics need to demonstrate their equivalence with respect to the reference apparatus by performing field tests. The American Environmental Agency, U.S.-EPA, also has a reference apparatus according to section 40 CFR part 53.

Continuous samplers facilitate the concentration of the particles measured in real time. In addition, this type of equipment is able to perform simultaneous measurements of different particle size fractions depending on the measuring principle used. The most widely used measuring instruments are based on the following principles: β-radiation attenuation, tapered element oscillating microbalance (TEOM), light scattering, etc.

In the present project a continuous measuring instrument based on light scattering has been used. The operating mechanism in this type of instrument is based on making a known air stream flow pass through an entry head in order to measure, with a laser-based detector, the light scattering produced by different particles. The instrument enables obtaining particle concentration values per particle size fraction. These instruments are ideal for the continuous monitoring of the particle concentration at point sources.

ITC has four instruments for environmental particle measurement. The main characteristics of the instruments used in this study are in set out in Table 2.

REF	INSTRUMENT	TRADEMARK AND MODEL	TYPE OF SAMPLING	MEASURABLE PARAMETERS
CS	Sequential sampler	Leckel, SEQ47/50	Gravimetric	PM_{10}, $PM_{2.5}$
CAV	High-volume sampler	MCV, CAV-A/M	Gravimetric	PM_{10}
CBV	Low-volume sampler	R&P, Partisol 2000	Gravimetric	PM_{10}, $PM_{2.5}$
MC	Continuous sampler	GRIMM, 1108	Light scattering	PM_{10}, $PM_{2.5}$, PM_1, 15 sizes

Table 2. Characteristics of the measuring instruments used.

These instruments were initially designed for measuring air quality in urban contexts. Therefore, to enable them to be used for measurements in industrial and mining activities with much higher particle levels, a series of modifications and adaptations have been required to assure proper operation. These modifications have basically been: assuring good sealing and good cooling.

3.2. STUDY OF EQUIVALENCE

In the course of the work the instruments used were calibrated, based on standard UNE-EN 12341, which proposes a comparative methodology based on two points. The first of these is the comparability between the candidate samplers, in which two or more presumed equal instruments are compared with each other, while the second criterion is the comparability of the candidate sampler and the reference sampler.

The comparability of two samplers is made on the basis of the so-called equivalence function, which describes the relation between the mass concentrations measured by each compared instrument. A candidate sampler meets the equivalence requirements when this function is within the limits of the margin of relevant acceptance and the coefficient of variation R^2 of the calculated function is $\geq$0.95 in the working concentration range.

The considerations adopted when comparing the different instruments have been: **parallel sampling** in at least **two different locations** and covering different environmental scenarios, one during a cold season (**winter**) and another one during a warmer season (**summer**).

The minimum number of validated data (pairs of daily averages) has exceeded 25 for each instrument.

3.3. METHODOLOGY FOR THE MEASUREMENT OF FUGITIVE EMISSIONS IN INDUSTRIAL FACILITIES

3.3.1. Location of the sampling points

In general, whenever technically possible, the following criteria have been applied:

- Setting the sampling instruments in the predominant direction of the wind, leeward of the generating source, always within the perimeter of the property of the studied company.
- Setting the sampling instrument several metres away from any obstacles that might interfere with the measurement to be made (screening of the emission source) and avoiding sampling points close to emission sources not belonging to the studied company, for example: common traffic areas without asphalt paving, etc.
- Before the measurement campaign with gravimetric equipment, a map of concentrations in the company was drawn up with the continuous sampler (MC), within the perimeter of the company, with a view to establishing the most important fugitive emission generation sources. Based on these concentrations, the most significant sampling points have been selected.

3.3.2. Monitoring meteorological variables

In order to know and determine the influence of the meteorological variables on the concentration and scatter of fugitive emissions, different parameters have been measured with the help of a weather station. The measured parameters were: wind speed and direction, temperature, relative humidity and rainfall. The positioning of this instrument should be representative of the sampling location; in principle, it should be located at a site away from any obstacles that might affect the measurements by screening.

3.3.3. Determination of PM_{10} concentration

3.3.3.1. Discontinuous measurements: gravimetric method

The gravimetric measurement of suspended particles was conducted with a sampler. The sampling time to collect sufficient mass of sample depends on the intake flow rate of each type of sampler. Table 3 summarises the operating conditions of these instruments during the work.

REFERENCE	CHARACTERISTICS	SAMPLING TIME
CS	15 filters, flow rate 2.3 m^3/h	24 hours
CAV	1 filter, flow rate 30 m^3/h	24 hours
CBV	1 filter, flow rate 1 m^3/h	48 hours

Table 3. Operating conditions of the sampling instruments.

In particle sampling, filters of quartz and glass fibre have been used, specifically: Schleicher & Schuell, quartz fibre QF-20, 47 and 150 mm diameter, and glass fibre, GF-9 of 47 mm diameter; and Whatman, glass fibre GF-A, 150 mm diameter.

The filters were appropriately prepared for weighing before and after sampling. Weighing was performed on a balance with a resolution of 0.1 mg. In the weighing of the filters after sampling we ensured that ambient conditions in the weighing room (temperature and relative humidity) were within the allowable tolerance in regard to the environmental conditions of the initial weighing.

When the instrument used was not a reference apparatus according to the standard UNE-EN 12341, its own equivalence function was applied to it.

3.3.3.2. Continuous measurement: light scattering

To conduct this type of sampling, the continuous suspended particle sampler indicated in Table 2 has been used. This instrument is used for drawing up the initial map of concentrations, carrying out measurements for short times at each location ($t<1h$), or for the measurement in a particular location for long times ($t>24h$). The integration time is variable; in this study we have used a time of 1 minute for the short-time measurements and 10 minutes for long times ($t>24h$).

4. RESULTS AND DISCUSSION

4.1. STUDY OF EQUIVALENCE

To calibrate the measuring instruments, reference instruments must be used; this reference equipment differs in Europe and the United States (Table 4).

Reference	Standard UNE-EN 12341	Standard USEPA
CS	Reference	--
CBV	Equivalent	Reference

Table 4. Gravimetric samplers used as reference instruments in Europe and the United States.

The equation of equivalence has been calculated in relation to CS (reference instrument according to the European standard) for each sampler. The calibration period includes different periods of the year, in winter and in summer, while the location has been limited to different companies which manufacture spray-dried granules and ceramic tiles in Castellón province, entailing minimum variability in the composition of the collected particles and in the climatic conditions. Figures 1 to 3 show the comparison of the measuring instruments used.

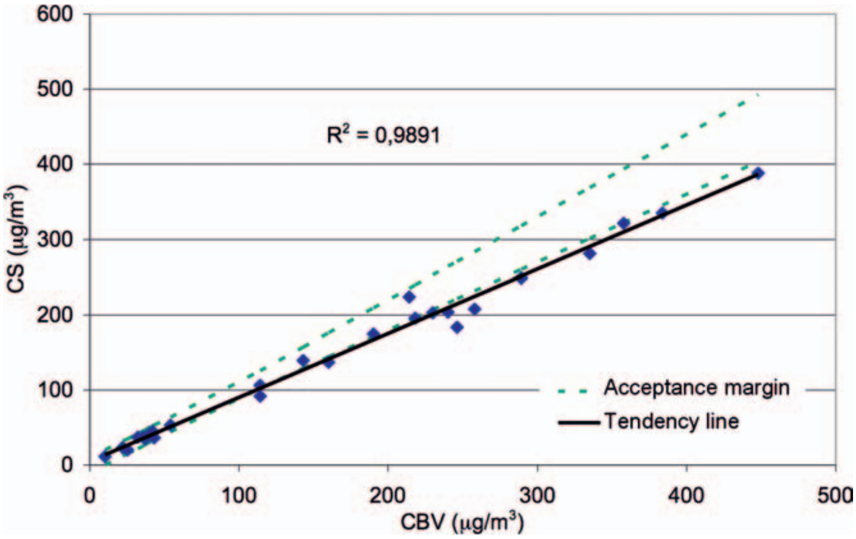

Figure 1. Calibration of the sequential sampler

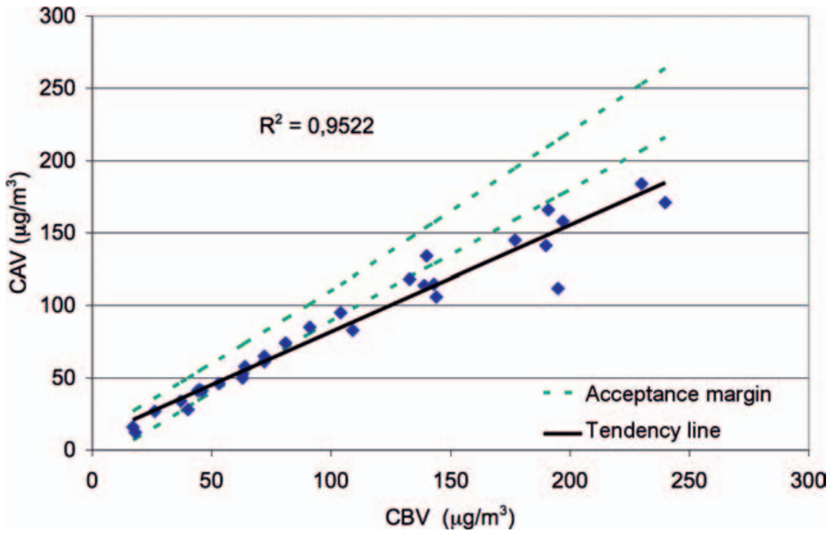

Figure 2. Calibration of the high-volume sampler

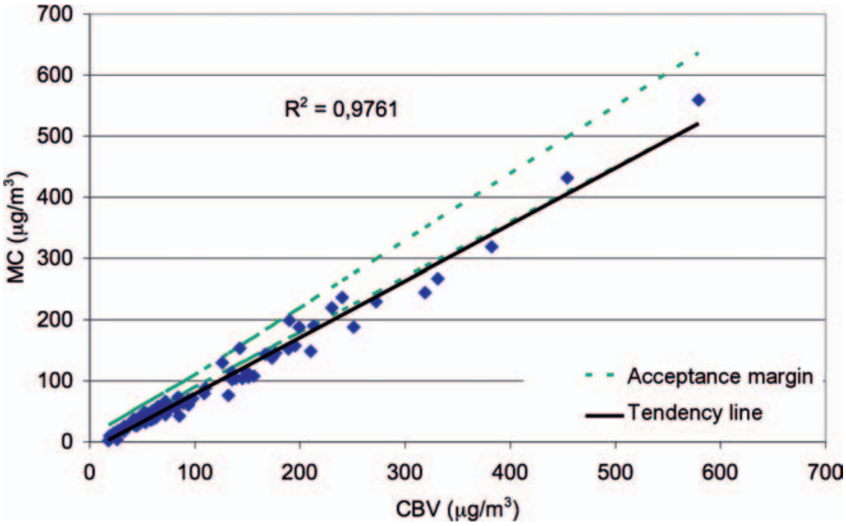

Figure 3. Calibration of the continuous sampler

Table 5 details the number of values used for the calculation of the equivalence function, the study range for each instrument, as well as the equivalence function obtained between the different instruments used in the study.

REF.	NUMBER OF PAIRS OF VALUES	RANGE OF VALUES (µg/m³)	EQUIVALENCE FUNCTION
CS	25	[10, 448]	1
CAV	30	[0, 240]	CAV = 0.8543 · CS + 5.1613
CBV	n.a.	n.a.	CBV = 1.1619 · CS – 4.2447
MC	77	[0, 600]	MC = 0.9330 CS +16.1295

Table 5. Equivalence functions between the different sampling instruments used

4.2. INDUSTRIAL MEASUREMENTS

4.2.1. Repeatability

To validate the methodology, we studied the repeatability of the obtained environmental concentration data. For this, two series of experiments were conducted, which consisted of repeating samplings at a same location in different periods of time, holding, as far as possible, the technological scenario (production, corrective measures, types of raw materials, etc.). This repetition was carried out at several locations to attempt to cover the usual range of concentrations at the most representative sampling points with regard to type of company and generating sources at the company, and periods of workdays and weekends. In all the cases the data of each series were always superior to two days' sampling. Each measurement point was positioned by a global positioning system (GPS) in order to repeat exactly the location of the different measurement points in the different series.

Table 6 summarises these average concentration levels together with the range of concentrations recorded, the sampling period and the number of samplings performed in each series.

PERIOD	LOCATION	PM_{10} (μg/m^3)					
		SERIES 1			SERIES 2		
		AVERAGE	RANGE, n	PERIOD	AVERAGE	RANGE, n	PERIOD
Workday	Location 2	**60**	[55, 77] n=2	Oct Nov 2003	**61**	[66, 73] n=2	Nov 2004
	Location 3	**107**	[80, 134] n=4	Oct Nov 2003	**108**	[89, 130] n=7	Nov 2004
Weekend	Location 1	**98**	n=1	Sept 2003	**93**	n=1	Nov 2004 Jan 2005
	Location 2	**48**	n=1	Oct Nov 2003	**43**	n=1	Nov 2004
	Location 3	**71**	n=1	Oct Nov 2003	**68**	n=1	Nov 2004

Table 6. Average PM_{10} concentrations in 24 hours obtained in the repeatability study.

The data in Table 6 confirm the repeatability of this methodology in the usual range of concentrations at the measurement locations in which no significant changes in the manufacturing process have taken place.

4.2.2. Industrial application: fugitive emission control by performance of measurements at internal points in the companies

This methodology has been used to characterise the PM_{10} levels in numerous industrial facilities for the manufacture of spray-dried granules and ceramic tiles, and has proven its applicability for companies of quite different size and configuration.

The measurement points have been located in every case with a global positioning system (GPS), in order to repeat the samplings at the same location in successive campaigns. During each measurement campaign, the PM_{10} concentration levels and most important meteorological variables were collected. All these values were subsequently treated; thus, the ones obtained in extreme meteorological conditions (for instance, strong winds, persistent rain, etc.) were discarded.

Though it makes no sense to compare the PM_{10} values recorded at points located on company grounds with the legal limits on air quality, even when they are taken in the open air, they still need to be compared with the limits on labour environments. However, the experience gathered in the course of this work indicates that analysis of these data by comparing them with values recorded in different measurement campaigns in the same facility is a very useful tool for diagnosing and controlling fugitive particle emissions.

On the other hand, to define limit values in an industrial environment is problematic, since they must be sufficiently realistic to allow conducting the proper activities of the production process, applying the best available techniques (BAT), yet sufficiently restrictive to assure the protection of human health and the environment as a whole. In this sense, in the few literature references found in which control of PM_{10} within the perimeter of industrial or mining facilities is proposed, a daily limit value of 150 μg/m^3 is established[10,11]. Therefore, in the measurements performed at different companies in developing this methodology, a target value has been established of 150 μgPM_{10}/m^3 (as an average in 24 hours), which it is attempted to achieve at all the internal points of the company. However, the information compiled to date is insufficient to judge whether this objective is realistic for all industrial facilities, since it needs to be validated with a significant number of data from industrial facilities in which the Best Available Techniques have been largely implemented. On the other hand, one of the problems arising in this type of measurement is the high scatter between points located in different positions, as a result of the strong variability of the fugitive particle emission-generating activities in the studied sampling points: traffic of trucks, crushing machines, loading and unloading areas of powdery materials, etc.

By way of example, Table 7 sets out the results obtained in a company that manufactures spray-dried granules, and stores raw materials outdoors, at which three measurement campaigns were conducted in a period of three years, and at which corrective improvements have been introduced to reduce fugitive particle emissions during the period of study.

CONCENTRATION RANGE ($\mu g/m^3$)	FREQUENCY 2001 (%)	FREQUENCY 2003 (%)	FREQUENCY 2004 (%)
0-50	0	0	0
50-100	0	25	14
100-150	14	13	50
150-200	14	25	14
200-250	0	25	14
250-300	14	0	0
300-350	14	13	7
>350	43	0	0

Table 7. Improvement of PM_{10} concentration levels at an industrial facility

This table shows a favourable evolution, year after year, of the concentration values below 150 $\mu g/m^3$, which go from 14% in 2001 to 64% in 2004. This means that the number of values below 150 $\mu g/m^3$ has increased by 50% due to the improvements implemented in the company. The opposite effect is observed in the values above 250 $\mu g/m^3$, which have diminished notably. In accordance with the set target value (150 $\mu gPM_{10}/m^3$ at 24 hours), in this case the company has been urged to continue introducing improvements in order to reach this criterion.

The obvious disadvantage of this control system, as indicated above, is that it does not enables quantifying the fugitive emissions in mass flow, for which data treatment is required, in addition to a more sophisticated sampling plan.

Monitoring a given activity or particular company zone in which important fugitive emissions are generated can be completed with an analysis performed with a continuous sampler (MC), which enables identifying high concentration episodes and assigning their causes in a temporal series. Figure 4 is presented as an example of this methodology.

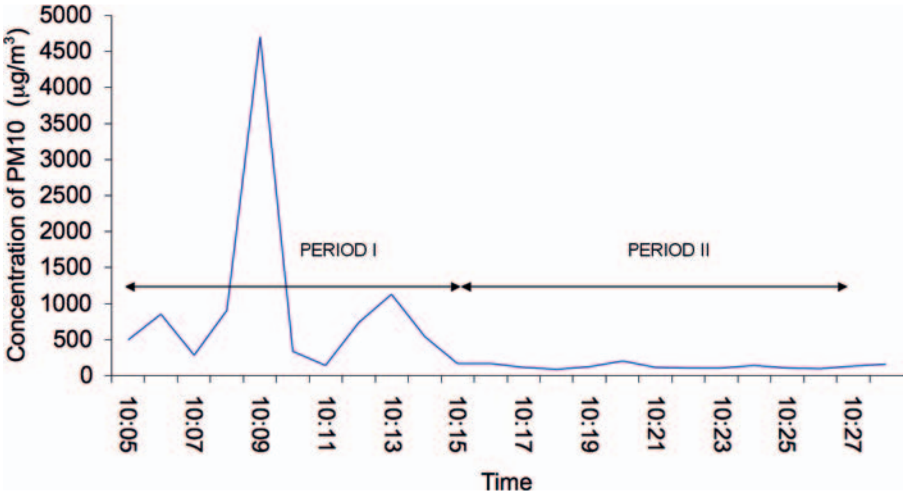

Figure 4. Monitoring PM_{10} levels at a location.

Figure 4 shows that two different periods can be identified at the measurement point. When these data were compared with information from the company (activities during the sampling) and with the meteorological information (absence of rain and gusts of wind), the main cause of fugitive emissions in period I was found to be heavy vehicle traffic. In the absence of traffic (period II), the activities adjacent to the sampling point were not significant.

5. CONCLUSIONS

- A methodology for control of fugitive PM_{10} emissions in the ceramic industry has been proposed and validated, based on the measurement of PM_{10} concentrations at different outdoor points of an industrial facility. This control system, despite the limitations remarked, is considered relatively simple and economic, in both execution and the interpretation of results. However, it requires adaptation of the commercial air quality measurement instruments to industrial conditions.
- The measurement of the PM_{10} concentrations in an industrial facility in different measurement campaigns has proven to be a valuable tool for establishing a method of internal control on the PM_{10} generating activities and for detecting points of conflict.
- The measurement of PM_{10} concentrations in ceramic industry facilities is considered to be a good parameter for control of fugitive emissions, although it entails the problem of not allowing comparison of the obtained values with the legal limits on air quality. In this sense, it might be interesting to propose setting a PM_{10} concentration limit value or daily target in industrial environments. Though the information compiled is insufficient to allow proposing a specific numerical value, which requires more exhaustive analysis and needs to be compatible with the existing regulations on air quality, it does appear interesting to consider differentiated limit values, as a function of the prevailing use of the ground and/or the distances to urban centres. An example in line with this proposal could be the legislation on acoustic pollution, which establishes different sound level limits depending on the prevailing use of the ground. In these cases, the values would obviously need to refer to points on the perimeter of the facilities.

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