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DEVELOPMENT OF REAL TIME DETECTOR FOR FLUORESCENT PARTICLES

C. PREVOST*, A. SEIGNEUR**, J. VENDEL*

*Institut de Protection et de Sûreté Nucléaire
Département de Prévention et d'Etude des Accidents - DPEA/SERAC
CEA/Saclay, Bâtiment 383 - 91191 GIF-SUR-YVETTE Cedex, France
**LETI (Direction des Techniques Avancées)
Département d'Electronique et d'Instrumentation
CEA/Saclay, Bâtiment 528 - 91191 GIF-SUR-YVETTE Cedex, France

ABSTRACT

Aerosols tagged by a fluorescent dye are a worthwhile tool within the framework of ventilation and filtration studies. The detection in real time of a specific particulate tracer allows characterization of ventilation behaviour such as air change rate, the determination of a good or bad mixing zone and transfer coefficient, or the determination of the decontamination factor for High Efficiency Particulate Air (HEPA) filters.

Generally, these tests require specific aerosols in order to get rid of the atmospheric aerosol background.

Until now the principle of fluorescent aerosol concentration measuring has only allowed an integral response with a time lag by means of sampling on filters and a fluorimetric analysis after specific conditioning of these filters.

To make it possible to detect a specific tracer in real time, a new monitor has been developed to count these particles on the following basis :

- fluorescent particles pass through a sampling nozzle to a specially designed measurement chamber ; ducting flow rate is used to contain the test aerosol in the sample flow rate at the nozzle outlet ;

- interception of this stream with a laser beam allows aerosol detection and characterization, particle by particle ;

- optical signals are produced by fluorescence emission of glycerol particles tagged with fluorescein sodium when they are excited by a laser beam emitting on a suitable wavelength (ionized argon laser) ; these signals are transmitted to a photodetector by a patented optical arrangement.

The paper presents the detection concept and the experimental device. The main results achieved in this study show the feasibility of such a technique providing aerosol tracer detection in real time.

I. INTRODUCTION

Aerosols artificially tagged with a fluorescent substance are a powerful tool for the specific follow-up of a polluting agent in an atmosphere naturally free from such particles. Most tests carried out within the framework of research into air contamination, confinement or else purification act on these aerosols both in some industrial fields and in specialist laboratories.

The detection of such particles can thus help in understanding the various complex phenomena produced inside premises and thus enable them to be controlled. In fact, the combination of ventilation and pollutant is a major concern when designing ventilation plant and making it comply with the relevant standards.

Therefore, the follow-up in real time of the transfer of contamination from a harmless polluting agent injected instead of a source of contamination enables the location of detection markers to be optimized and the rate of homogenization of the pollutant in the premises determined. Optimization of the rate of renewal is then conceivable when making a "map" of the concentration of the pollutant inside the premises. This reveals the presence of a "dead zone", the site of pollutant accumulation, and thus enables it to be eliminated.

This particle tracing technique can also be used in designwork concerning in particular dynamic confinement, the validation of aeraulic design calculation codes, the modelling of flows of air loaded with particles or even the qualification of the filtering equipment.
The principle of measuring such specific aerosols previously only allowed for integral measurement of their concentration with a time lag through taking samples on filters, and then preparing the samples for analysis by fluorimetry.

As has previously been explained, the need for following up the concentration of a tracer in the air in real time has led us to show the feasibility of a new technique for detecting fluorescent aerosols.

In this paper, we explain the principle of detection which we apply, the experimental device made, and the main results achieved during our research.

II. PRINCIPLE OF MEASUREMENT

The main objective of our research lies in designing a technique for characterizing and counting aerosols, based on detection of their fluorescence, in real time [4].

When a fluorescent fluid aerosol intercepts a beam of monochromatic light of a wavelength included within its spectrum of absorption, it sends back a light of a higher wavelength due to the fluorescence.
With constant lighting, its intensity is in proportion to the amount of fluorescent substance contained in the particle.

It's a question of collecting this fluorescent light using a suitable optical system on a photosensitive detector in order to quantify the fluorophore content [5].

$$Q_f = k.P_L.\lambda_a.\Phi.\varepsilon.N_f.t$$

where Qf = amount of fluorescence (number of photons)
k = proportionality factor
P_L = power of the laser beam (Watt)
λ_a = absorption wavelength (nm)
Φ = quantum yield of fluorescence
ε = molar absorption coefficient (L/mol.cm)
t = time during particle remains in the light beam (s)

An aerodynamic focusing device aligns the particles behind each other. Fine focusing of the excitation light enables lighting as a particle moves.

Using a laser source of light facilitates concentration of the excitation power of the beam on a narrow spot of light.

The volume of measurement can be estimated at about 3.10^{-6} cm^3 ; the maximum concentration which can be detected with a rate of coincidence below 5% must not exceed 10^4 particles/cm^3.

III. MEASUREMENT CELL AND ASSOCIATED OPTICS

The measurement cell essentially comprises :

→ a sampling system (Figure 1) in brass with a low surface finish index for the inner surface, ending in a small diameter nozzle (d_{bp} = 0.6 mm) which allows the aerosols to be routed into the measurement chamber within a centered jet.

A ducting air system improves the aerodynamic centering of the output jet in the same time as it prevents any droplets being deposited on the walls of the collector optics.

→ a measurement chamber (Figure 2), the special geometry of which enables the maximum fluorescent light emitted to be collected. The fluorescence collection performance is in the region of 15 %. This collector optics is covered by a patent [1]. Its rear face, centered on the jet, is metallized which allows the light emitted to the rear to be returned to the place of emission where it's added to the flow emitted forward.

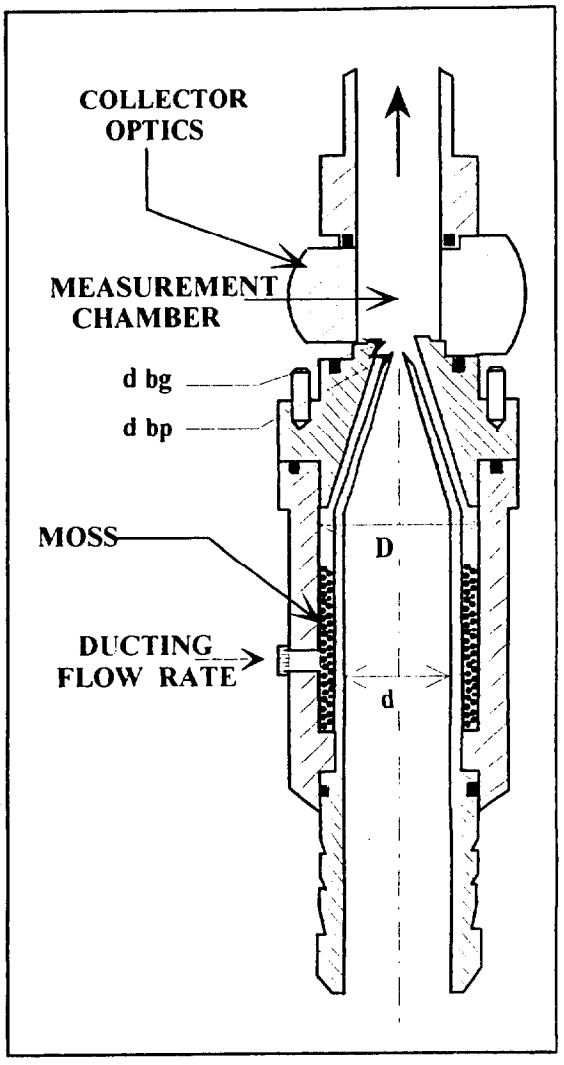


Figure 1 : Sampling system

Specification of the nozzle : D = 10.6 mm
d = 6.0 mm
d_{bp} = 0.6 mm
d_{bg} = 2.6 mm

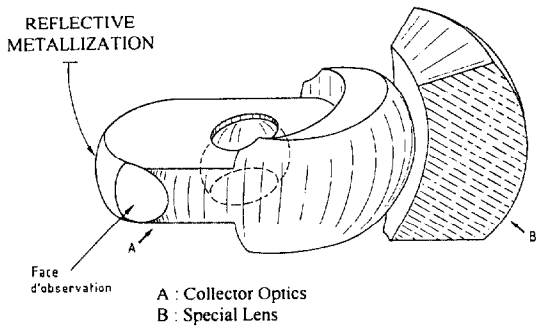


Figure 2 : Measurement chamber

Lens B catches the flow of light and sends it all back to the rest of the optic arrangement.

IV. CHOICE OF FLUOROPHORE

The fluorophore selected for carrying out our tests is soda fluorescein (C_{20} H_{10} O_5 Na_2) ; it is an excellent product to use in tracing work, essentially because it's non toxic in the usual concentrations and also because it is soluble in water and in glycerol, often used when producing calibrated droplets.
Further, it's used in a standardized measurement method (NF Standard X 44.011) for the test of HEPA filters efficiency.
This tracer has very good fluorescence properties in solution in glycerol (quantum performance evaluated at 92% under the best conditions) and can easily be detected by any fluorimetric technique.

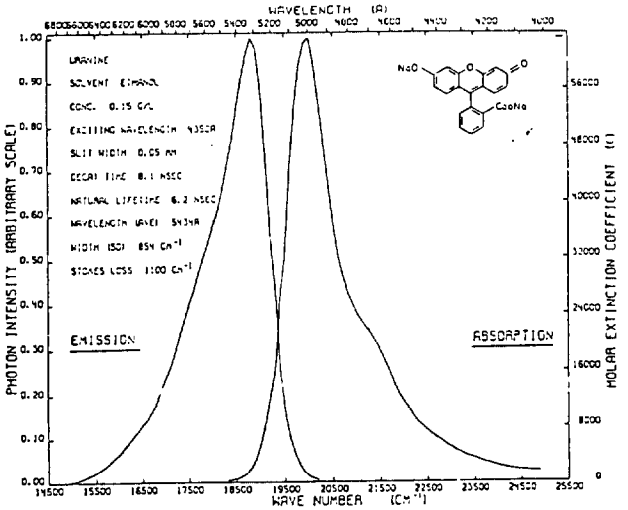


Figure 3 : Spectrum of soda fluorescein absorption and emission [3].

V. SETUP OF THE EXPERIMENT

Two main sections can illustrate the principle of operation of our arrangement (Figure 4) :

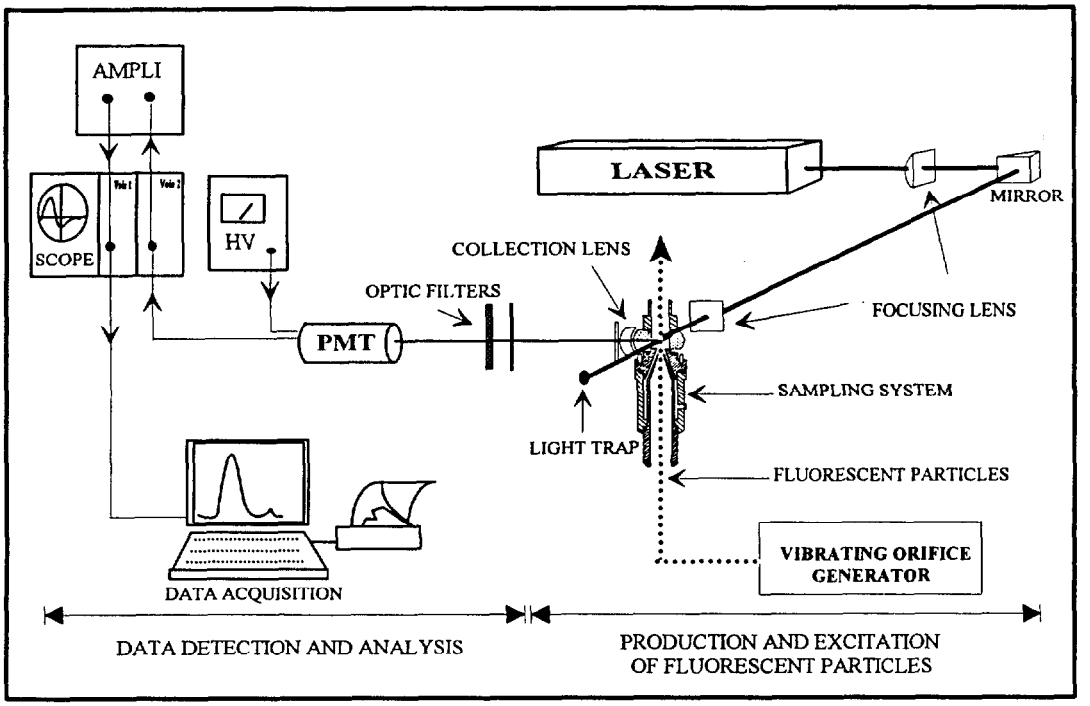


Figure 4 : Overall view of the experimental device

V.1. Production of droplets

A vibrating orifice generator (TSI Inc., Model 3050) provides for producing monodispersed droplets of glycerol tagged with soda fluorescein at concentrations of 1 and 10 g/L.
The range of diameters of aerosols produced extends from 1 to 10 µm (aerodynamic diameter), i.e. quantities of fluorophore contained per particle between 3.10^{-16} and 3.10^{-12} g/particle inclusive i.e. 6.10^{5} to 6.10^{9} molecules/particle.
It has been possible to check the particle size features of the test aerosols by means of an APS (Aerodynamic Particle Sizer, Model 3310, TSI Inc.).
The spectrum illustrated in Figure 5 is found when producing droplets with a diameter of 5 µm.

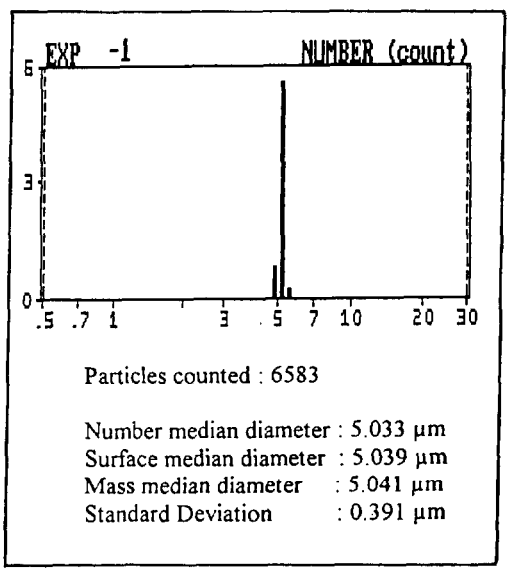


Figure 5 : Results from APS System

V.2. Dilution and sampling

The reference sample is delivered by an ejector with the flow rate controlled by a calibrated sonic orifice (SAPELEM) at the required rate (5 L/min.).
The total sample (sum of ducting and sampling flow rates) is delivered by two vane pumps mounted in series (RIETSCHLE).

The ducting deliveries and total sampling are checked by calibrated mass flowmeters (BROOKS, Models 5851) in the range of outputs used ; the difference corresponds to the delivery of particles taken.

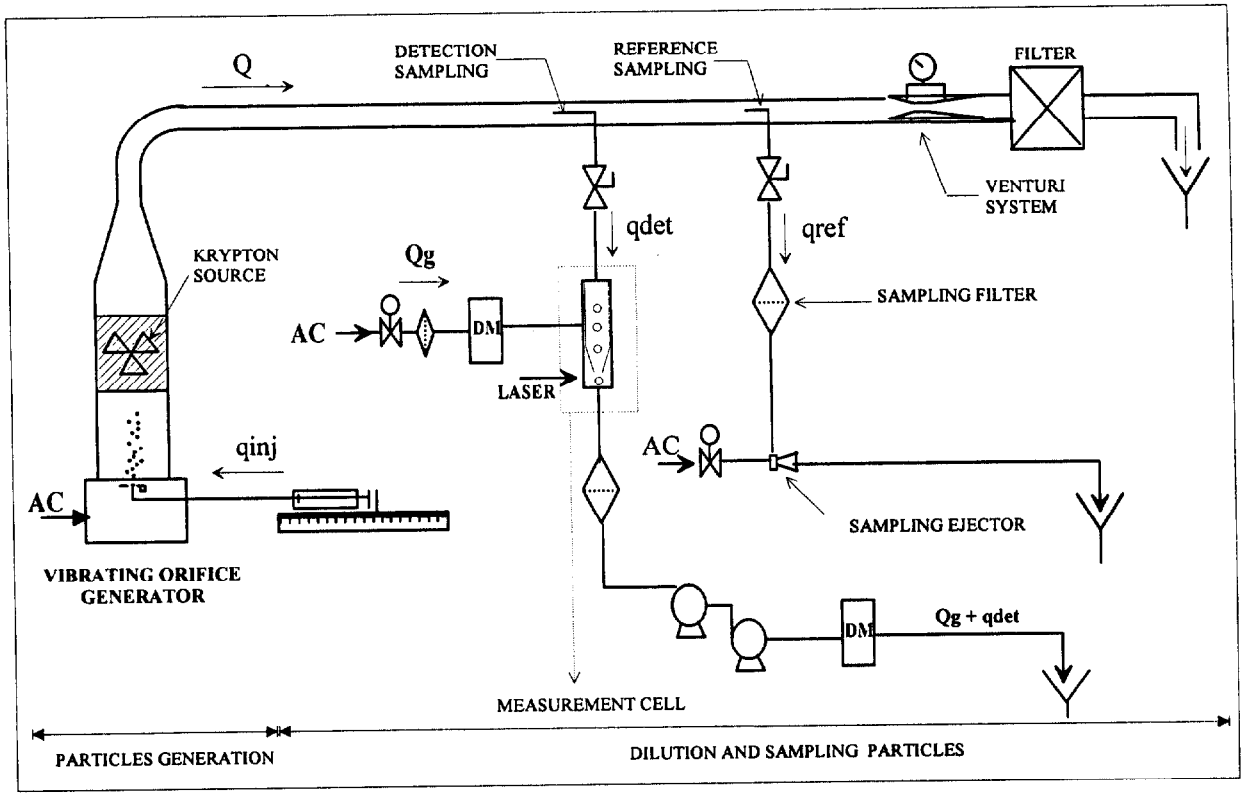


Figure 6 : Aeraulic diagram for the counter qualification

KEYS:
Q = dilution output of aerosols in the main line (m^3/h)
Qg = delivery of ducting air in the measuring cell (L/min)
qdet = sample flow rate in the detection cell (L/min)
qref = sample flow rate on the reference filter (L/min)
qinj = injection delivery of fluorescent solution into the generator (mL/min)
DM = mass flowmeter
AC = compressed air

VI. DETECTION OF AEROSOLS

VI.1. Laser source

The intention to make a device for measuring high sensitivity has led us to adopt a laser as the source of very bright light, monochromatic, easy to focus and, moreover, stable.

So we use an ionized argon laser (SPECTRA PHYSICS, Series 2000) which emits on wavelength 488 nm in accordance with the absorption wavelength of fluorescein sodium.
The power is intentionally limited from 10 to 300 mW. It supplies a beam on TEMoo mode, i.e. its profile of intensity is Gaussian.

VI.2. Focusing the beam

Figure 7 shows the distribution of the intensity of the laser beam at the level of the particles loaded air jet.

The focusing device is large in front of the diameter of the sample stream ; the intensity of the particle excitation is thus uniform, even for particle positions which fluctuate slightly in the lighted area.
The combination of special lenses enables the beam to take on the appearance of a sheet of light at the level of the flow of particles, the focusing thickness of which is fine so as only to illuminate a single particle going through (see Principle of Measurement).

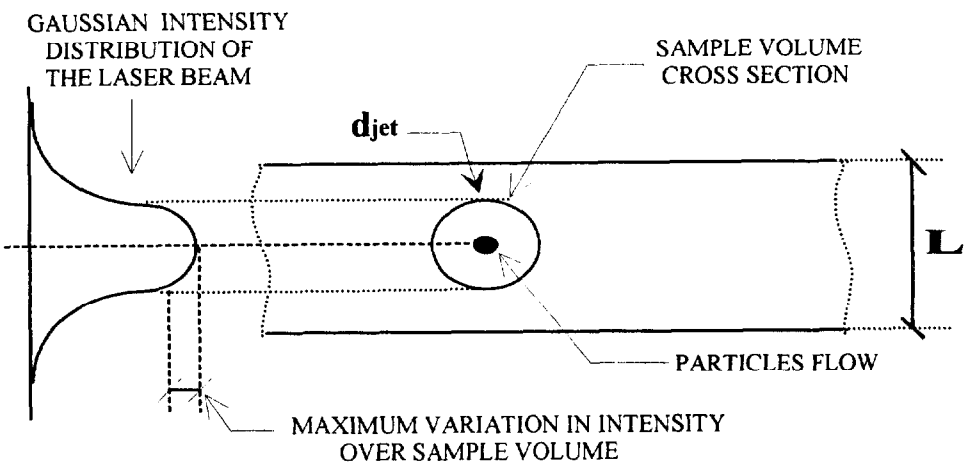


Figure 7 : Distribution of the laser intensity at the level of the jet as it leaves the nozzle.

VI.3. Chromatic filtering on the fluorescence light analysis channel

The association in series of an interference filter (CORION, LL500) and a colored filter (OG515) discriminates well between the wavelengths transmitted in comparison with the excitation light ; only fluorescence signals coming from the particles reach the detector.

Therefore, we clear the parasite light reflected mainly by the walls of the cell and the diffusion light also produced by the particles.

VI.4. Detection and collection of data

A photomultiplier (HAMAMATSU, Type R928) collects the fluorescence photons and converts them, depending on the performance of the photocathode and its gain, into electric current ; the signal leaving this detector can be observed directly on one of the two channels of an oscilloscope before it is transmitted to the amplifier (SCHLUMBERGER, Type 7129) where it is formed and amplified.
The outlet from the amplifier is connected to the second channel of the oscilloscope mounted in parallel with the data collection line.

The data collection line consists of a computer (PC 386) completed with an interface card, type Accuspec NaI (CANBERRA) ; the coding is done on 2048 channels.
We thus have a high performance pulse analyzer which reconstitutes the spectrum of quantities of fluorescent photons detected ; it delivers histograms worked out by adding up the number of events filed according to their amplitude.

This arrangement allows us to follow in real time the detection of fluorescence signals emitted by the particles, simultaneously on the dial of the oscilloscope and on the computer. It is then easy correctly to adjust the focusing of the laser beam on the jet of particles by adjusting the micrometric settings of the cell (XY movement). The interaction between the laser beam and the particle flow is thus optimized by finding the maximum amplitude signal which offers the best resolution.

VII. MAIN RESULTS

VII.1. Estimating the rate of retention in the measurement cell

One of the main stages in the qualification of our device is to validate that laser detection is representative in relation to a reference sampling made in parallel. To do this, we have estimated the rate of retention "T" of the droplets right inside the device.

Three series of ten tests with simultaneous sampling reference and downstream of the laser detection device (Figure 6) carried out for each particle diameter between 1 and 10 μm inclusive, enable us to draw a retention curve characteristic of the device (Figure 8) after conventional fluorimetric analysis of the sampling filters.

The bars associated with the results correspond to uncertainty over the results, given with 95% probability.

$$T = \frac{Cref - Cdet}{Cref}$$

where Cref = reference concentration (g/L)
Cdet = concentration (sampling) measured downstream of the device (g/L)

The characteristic spectrum found during the detection of fluorescent particles is illustrated by few results recorded when detecting particles of different diameters (Figure 9).

The scale of the abscissa is in direct proportion to the amount of fluorescein measured per particle counted off, i.e. to the volume of the particle for a constant fluorescein concentration in the solvent (glycerol).

As far as the scale of the ordinates is concerned, it totals the number of particles detected per unit of time.

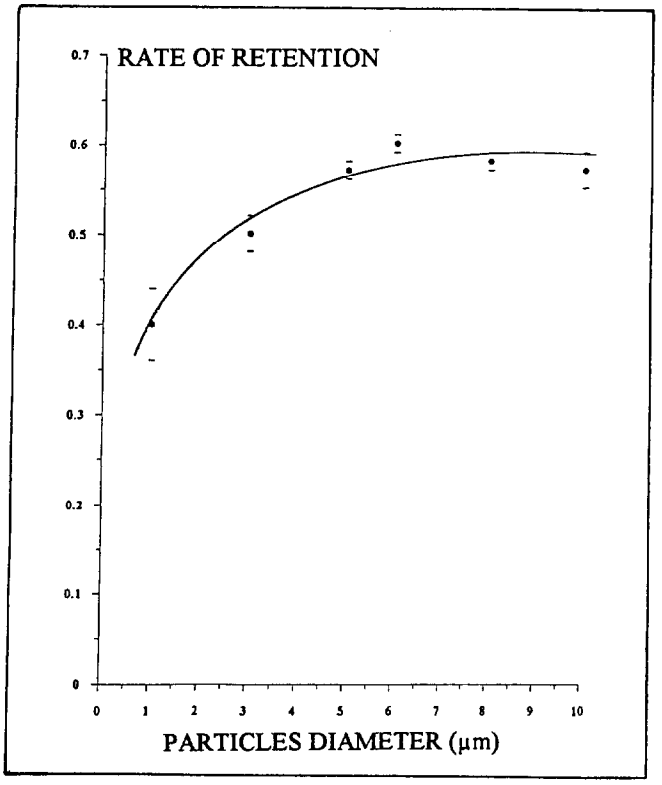
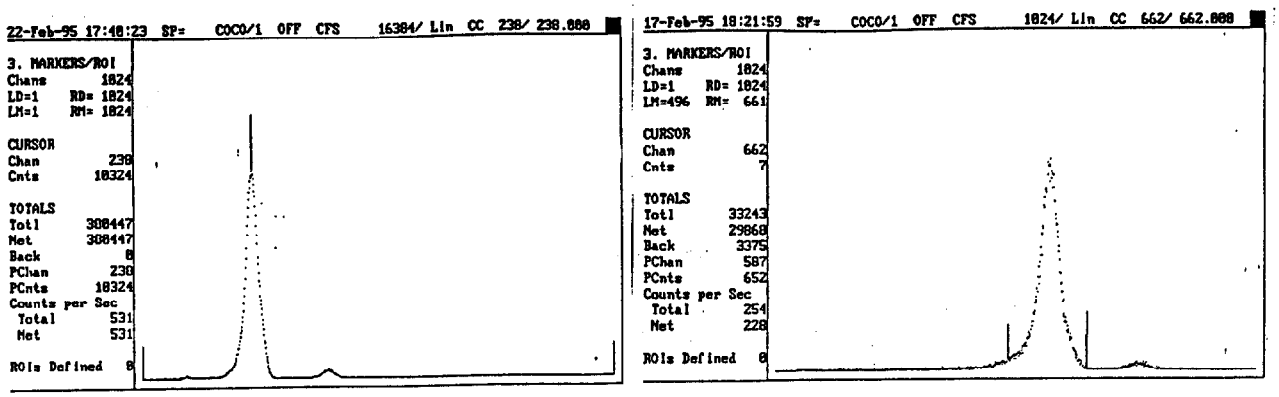


Figure 8 : Rate of retention versus particle diameter.

VII.2. Analysis of the signal given

Under good test conditions, i.e. optimum focusing of the laser beam on the jet of particles and a population of monodispersed droplets, the histogram found takes on a gaussian pace centered on the average value of the intensity of fluorescence measured.

Integration under the peak allows for determining the total number of particles detected during the analysis time.



dae = 5 µm dae = 10 µm

Figure 9 : Examples of spectra recorded

VII.3. Effect of different parameters

Several parameters can change the rate of the spectra recorded for a given diameter of particle with a fixed fluorophore concentration ; they are mainly :

- power of the laser beam (therefore of the excitation lighting) ;
- high voltage applied at the photomultiplier (PMT) ;
- time during particle remains in the laser beam ;

- alignment of the excitation beam on the jet of particles ;
- centering of the flow of particles (action of the ducting air flow rate).

The first three parameters quoted act essentially on the limit of detection of the device ; as for the other two, they can entail the spectrum being wrongly interpreted when they are incorrect.

Figures 10 and 11 show that the figures for the power provided by the laser and for the high voltage applied at the PMT must be optimized to maintain the linearity of the detection signal with the fluorescence of standard particles.

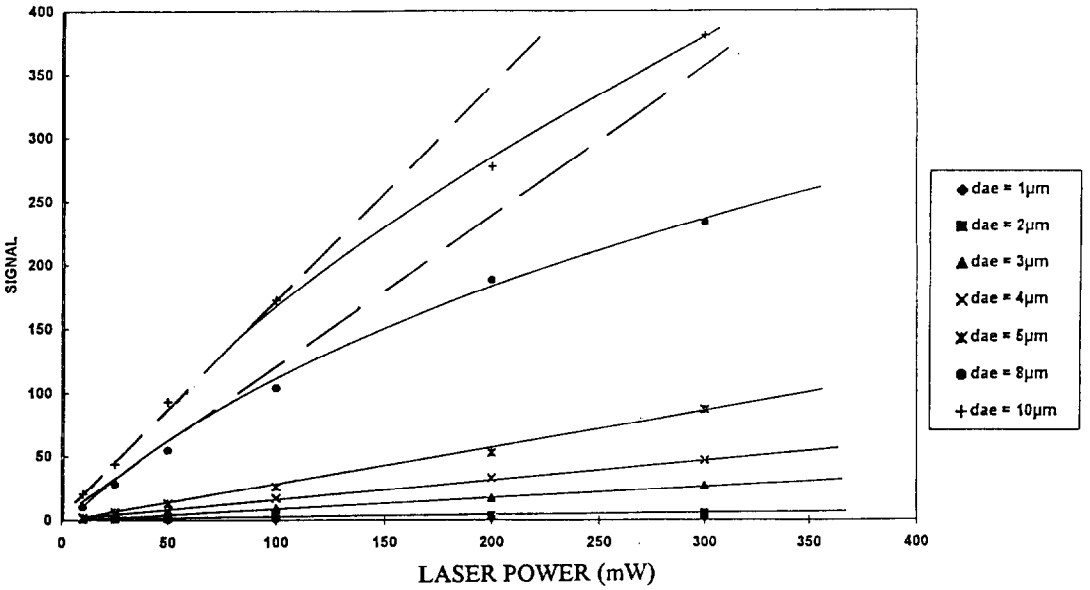


Figure 10 : Effect of the laser power on the fluorescence signal.

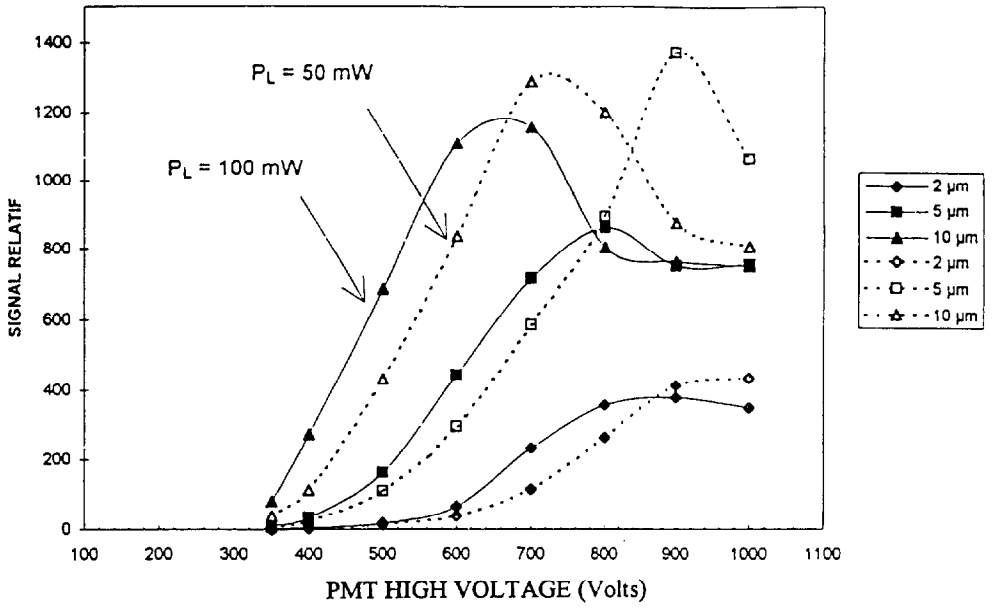


Figure 11 : Effect of the High Voltage of the PMT on the fluorescence signal

VII.4. Calibration test

A calibration curve can be drawn from the time one is working with all different optimized and fixed parameters.
The fluorescence signals emitted depend on the weight of fluorophore contained per particle, this is shown in Figure 12.

However, beyond certain conditions concerning laser power, high voltage of the PMT, size of droplets or else the fluorescein concentration, we encounter problems both with the limit of detection and with electronic saturation of the system.

In fact, when any of these various parameters increase, the linearity of the signal is only maintained in a certain field.

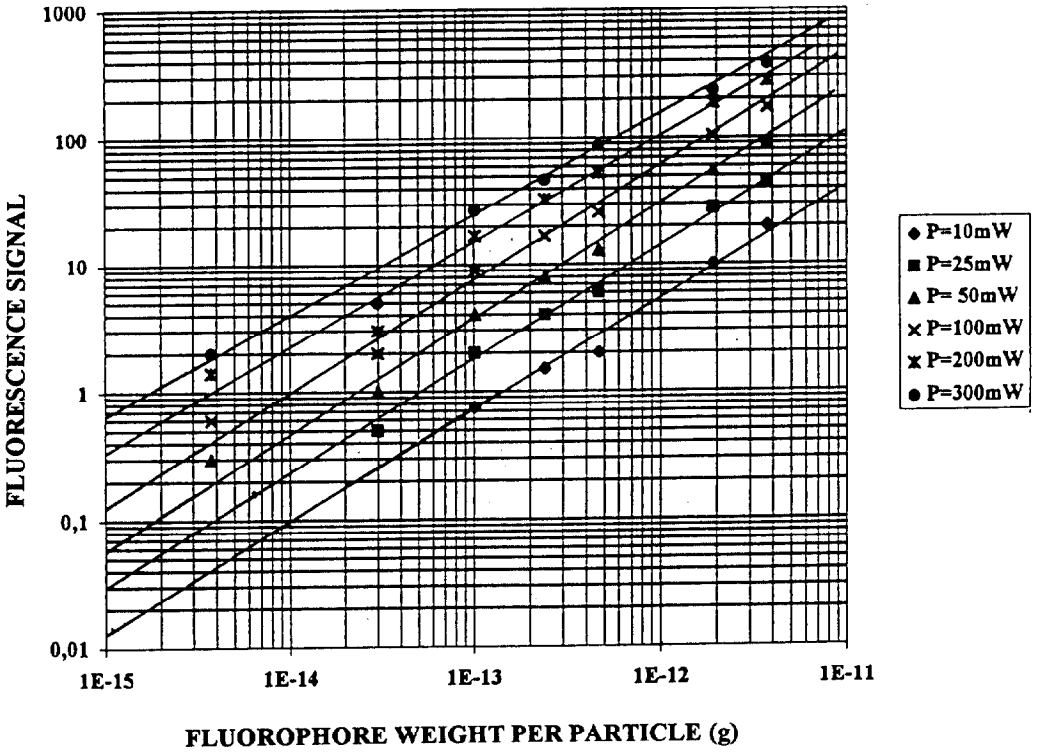


Figure 12 : Fluorescence signal versus weight of fluorophore contained per particle.

VII.5. Threshold of detection

The detection threshold of the device depends on the noise disturbing the signal for a fixed high voltage and laser power.

Thus, the detection threshold characteristic of our device is expressed as the lowest amount of fluorophore which can be detected. It's about 6.10^6 molecules per particle (10^{-15} g of soda fluorescein) detected at a laser power of 50 mW .

This limit value of detection can also be expressed in terms of the minimum particle diameter which can be measured, variable in accordance with the fluorophore concentration used.

VII.6. Comparison of results with conventional fluorimetry

Various tests have enabled us to compare the two differents systems by estimating the mean deviation between the results achieved by laser detection with those achieved with conventional fluorimetry when measuring an identical particulate concentration, for different droplet diameters.

We find results which are quite closed between the two techniques since the difference is below 10% in all instances in which the spectra obtained can be used (separation of the fluorescence signal with that corresponding to the electronic or optic noise).
The parity diagram (Figure 13) illustrates that the laser detection is a reliable method of particles counting versus the conventional fluorimetry method.

The analysis times using the laser counting method are reduced and depend only on the particulate concentration present, for a constant delivery : it is desirable to wait for detection for a minimum of 10^4 events in total in order to reach satisfactory counting statistics (2 % accuracy) which, depending on the concentration measured, can involve counting times of less than one minute.

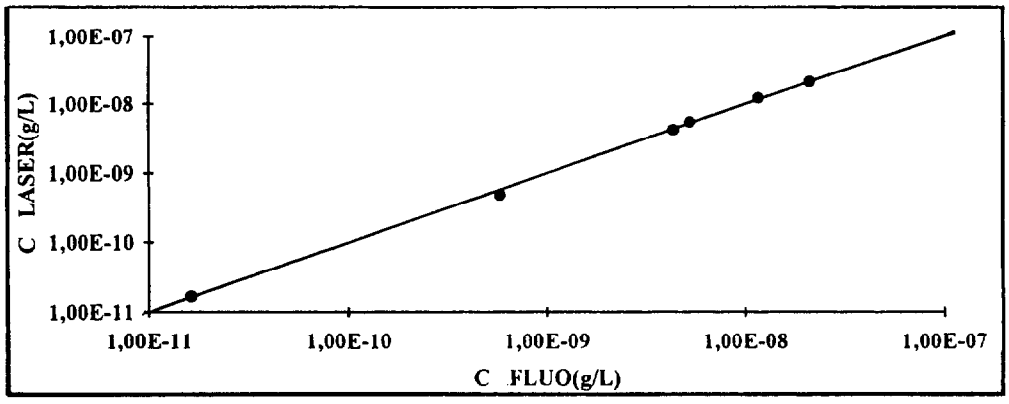


Figure 13 : Parity diagram

where C_{LASER} = results from laser detection (g/L)
C_{FLUO} = results from conventional fluorimetry (g/L)

Our device allows great selectivity of detection since it only counts fluorescent particles. Spectra which have nil counting rates are found when samples are taken direct in the ambient air.

VIII. CONCLUSIONS AND PROSPECTS

Using a suitable laser source associated with high yield collection optics contributes to making a high performance fluorescent aerosol detection device for follow-up in real time and continuously of a test particulate concentration injected into a room or test rig.

Through the data collected the technique allows direct access to particle counting rates while controlling the particle size of the aerosols produced.
Estimating the total weight of fluorophore detected is facilitated for monodispersed particle analysis as soon as the content of each of them is known.

The limitations of this technique are directly connected with the noise level (optics and electronics) produced by the device as well as by the physical and chemical properties of the fluorophore used.

Manufacturing in the course of time a portable unit for taking measurements on site, calls for a technology which is compact and light and that essentially limits the size of the laser source used. In spite of the fact that there are ionized argon lasers on the market producing powers in the region of 20 to 100 mW, it would be wise to consider new excitable dyes in the red to permit the use of laser diodes which are very small and inexpensive. However, the dyes used must be quite harmless to human beings, which considerably limits the choice. In spite of our various researchs, only one dye is appropriate, the allophycocyanin that is a very costly product.

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