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Physical characteristics of generated micron tracer particles

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Summary

In the framework of the TITANS research programme, micron-sized particles containing a tracer are to be generated in order to quantify their dry and wet deposition on terrestrial vegetation during experimental campaigns. These inert particles must have the same characteristics as the tritiated particles emitted during the dismantling works of nuclear facilities. The aim of this report is to present the physical characteristics that the particles to be generated must have in order to be representative of the particles emitted by these dismantling works, the generation method chosen and the characteristics of the particles that will be generated using this method in the framework of the project.

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TITANS

Tritium Impact and Transfer in Advanced Nuclear ReactorS

PHYSICAL CHARACTERISTICS OF GENERATED MICRON TRACER PARTICLES

WP 3

21th November 2023

P. ROUPSARD *(IRSN/PSE-ENV/STAAR/LERTA)*

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Abbreviation and Acronyms

Acronym	Description
ELPI+	Electrical Low Pressure Impactor +
EU	European Union
LPI	Low Pressure Impactor
WP	Work package

Executive Summary

In the framework of the TITANS research programme, micron-sized particles containing a tracer are to be generated in order to quantify their dry and wet deposition on terrestrial vegetation during experimental campaigns. These inert particles must have the same characteristics as the tritiated particles emitted during the dismantling works of nuclear facilities. The aim of this report is to present the physical characteristics that the particles to be generated must have in order to be representative of the particles emitted by these dismantling works, the generation method chosen and the characteristics of the particles that will be generated using this method in the framework of the project.

Keywords

Micron-sized particles, generation, fluorescein, particle size distribution

Introduction

During the dismantling phases of nuclear fusion or fission equipment and facilities, tritiated aerosols may be produced as a result of the surface treatment of concrete or the cutting of stainless steel pipes. If confinement is lost during this works, tritiated dusts may be dispersed in the atmosphere and lead to exposure of the environment or the surrounding population, particularly through their dry deposition (in the absence of precipitation) or wet deposition (in the presence of precipitation).

In the framework of the TITANS (Tritium Impact and Transfer in Advanced Nuclear ReactorS; 2022-2025, Horizon Europe Project) research programme, the dry and wet deposition of these particles on terrestrial vegetation will be measured in the environment during experimental campaigns in order to improve understanding of the physical deposition processes. However, the experiments will be based on the use of non-radioactive particles generated and dispersed in the environment (tritiated particles cannot be intentionally dispersed in the environment for research purposes). A tracer attached to these particles, fluorescein already used in the environment by Maro *et al.* (2014), will make it possible to quantify their dry and wet deposition. The generated fluorescein particles must have the same characteristics as the tritiated particles produced by the dismantling work, as far as existing technical solutions allow.

The aim of this report is to present:
- the characteristics of the particles to be generated, based on knowledge of the tritiated particles that may be put into suspension during dismantling phases;
- an ad hoc method for generating particles containing a tracer that can be used in the environment;
- the physical characteristics of the particles that will be generated and dispersed in the environment during the experimental campaigns.

1 Characteristics of tritiated concrete and stainless steel particles

Relatively few data are available in the literature on the characteristics of concrete and stainless steel particles suspended during dismantling work. The known data comes from the work of the TRANSAT project (2017-2021, EU Horizon 2020 programme), Sow *et al.* (2020) and Payet (2023), in the framework of the present TITANS project.

1.1 TRANSAT project (2017-2021)

The general aim of the TRANSAT project was to improve knowledge about tritium management in nuclear fusion and fission facilities. Part of the project was devoted to the impact on populations of tritiated particles releases, more specifically through radiotoxicity, radiobiology and dosimetry studies.

In the framework of the TRANSAT project, Gensdarmes *et al.* (2019) has used an optical particle counter to characterise the diameter of stainless steel particles produced by cutting operations in a glove box. The produced particles presented a mean geometric diameter of 4.1 µm (corresponding to an equivalent optical diameter).

In their study, they has recommended the use of spherical stainless steel substitution particles with a median mass diameter (equal to the volume diameter) equal to 4.7 µm, in equivalent optical diameter, and a geometric standard deviation of 1.35 (Figure 1) to simulate particles suspended by cutting operations.

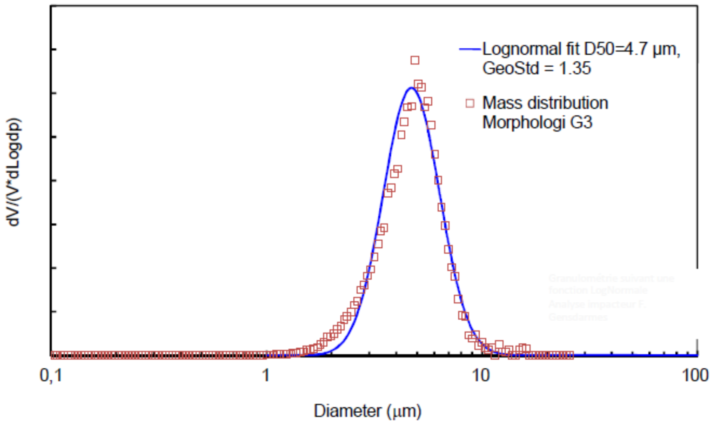

Figure 1: Volume (and mass) particle size distribution of the stainless steel substitution particles used by Gensdarmes *et al.* (2019) (in equivalent optical diameter; V: particles volume, dp: particles diameter, D50: median diameter, GeoStd: geometric standard deviation, Morphology G3: optical microscope type)

The mass median aerodynamic diameter associated with these stainless steel particles is 13.3 µm.

1.2 Sow *et al.* (2020)

Sow *et al*. (2020) has characterised the particle size distribution of concrete particles suspended by scarifying operations in a confined experimental tent. Measurements were realized using an Andersen cascade impactor, which gives aerodynamic diameters, and an optical particle counter, which gives optical diameters. Their measurement results are shown in Figure 2 and Figure 3.

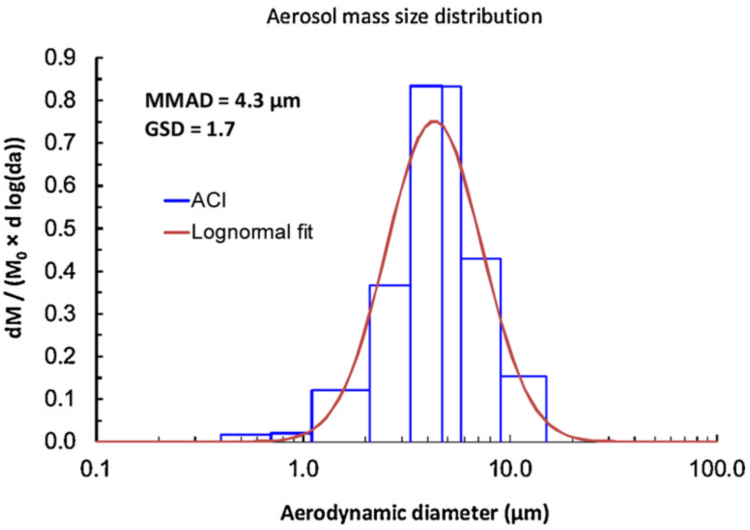

Figure 2: Mass size distribution of concrete particles suspended during scarifying operations, sampled by Andersen cascade impactor (Sow *et al.*, 2020; M: particles mass, M0: total particles mass, da: aerodynamic diameter, MMAD: mass median aerodynamic diameter: GSD; geometric standard deviation, ACI: Andersen cascade impactor)

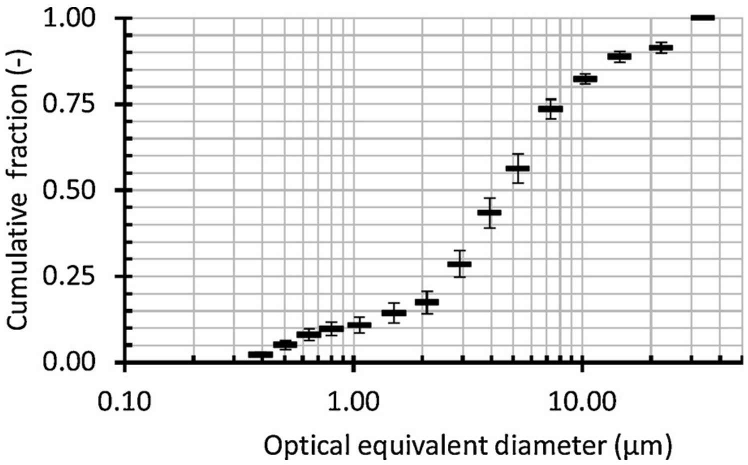

Figure 3: Cumulative mass size distribution of concrete particles suspended during scarifying operations, measured by optical counter (Sow et al., 2020)

The mass median aerodynamic diameter of the particles was 4.3 µm, with a geometric standard deviation of 1.7 (Figure 2). The mass median optical diameter is between 4 µm and 5 µm (Figure 3). The size distribution of the concrete particles is polydispersed.

1.3 WP3 of the TITANS Project (2023)

In the framework of the WP3 of the TITANS project, the diameter of concrete particles produced by abrasion in a glove box and the diameter of stainless steel substitution particles recommended by Gendarmes *et al.* (2019) has been characterised using an Andersen cascade impactor, following the method developed in TRANSAT (Rose *et al.*, 2019). The measurement results are shown in Figure 4.

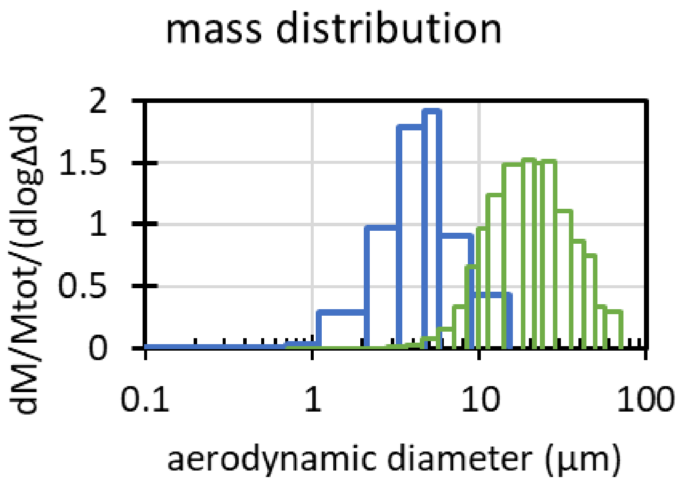

Figure 4: Mass size distribution of concrete particles (blue) suspended by abrasion and distribution of stainless steel substitution particles (green), sampled by Andersen cascade impactor (WP3 of the TITANS Project, 2023)

The mass median aerodynamic diameter of concrete particles is 4.2 µm. The mass median aerodynamic diameter of stainless steel particles is 13.3 µm.

1.4 Required characteristics of the particles to be generated

The data presented above show different diameter classes depending on the nature of the particles, concrete or stainless steel. Concrete particles have median aerodynamic diameters of between 4 and 5 µm, while stainless steel particles have a median aerodynamic diameter of 13.3 µm. Both types of particle have polydispersed distributions with geometric standard deviations ranging from 1.4 to 1.7. Table 1 summarises these literature data.

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Nature of the particles	Authors	Mass median aerodynamic diameter and associated geometric standard deviation	Mass median optical diameter and associated geometric standard deviation
Stainless steel	Gensdarmes *et al.* (2019)	13,3 µm / 1,4	4,7 µm / -
	WP3 TITANS Project (2023)	13,3 µm / -	
Concrete	Sow *et al.* (2020)	4,3 µm / 1,7	4 µm to 5 µm / -
	WP3 TITANS Project	4,2 µm / -	

Table 1: Summary of literature data on stainless steel and concrete particle size distributions

In the framework of the TITANS project, it has been decided that dry and wet deposition would only be studied for one of these particle types, concrete or stainless steel. Concrete particles has been retained with a polydisperse distribution centred on a mass median aerodynamic diameter of between 4 µm and 5 µm. This choice was made for scientific reasons. The range of concrete particle diameters is:

- representative of a dispersible fraction of particles in the atmosphere with a residence time ranging from one day to several days (Jaenicke, 1988), which enables their transport in the environment over long distances;
- deposited under the action of several physical phenomena: impaction, sedimentation and interception; with different intensities to be quantified according to micro-meteorological conditions;
- associated with a lack of data on dry and wet deposition, which limits the improvement of modelling of these depositional processes.

2 Material and method for generating and characterising particles

2.1 Material

2.1.1 Generating particles

The generator used is the MICRONICE® 12 µm (associated with the P&S 360 kit) produced by TEKCELEO (Figure 5). This generator technology is based on the use of a vibrating micro-perforated membrane coupled to a piezoelectric transducer and supplied with liquid. When the membrane vibrates at a certain frequency, it ejects fine calibrated drops of liquid through the micro-perforations to form a cloud of micro-droplets

Figure 5: View of the TEKCELEO MICRONICE® 12 µm generator (P&S-360) in operation (source: TEKCELEO.com)

The use of this generator model to generate micron-sized particles has already been studied for IRSN's internal needs and has been the subject of an internal report (Kort *et al.*, 2022). During this study, particles with diameters of the order of 5 µm were generated. In addition, the stability of the generation rate, given at 4.5 mL of particles produced per minute, was demonstrated over a period of around 50 minutes.

The principle of micron particle generation with this generator is based on the use of a solution containing a quantifiable tracer, a low-volatility liquid (the main constituent of the generated particles) and a volatile solvent that evaporates quickly following the generation of the micro-droplets. The particle size distribution can be modified by adjusting the concentrations of the main component (low volatility) and the tracer in the solution. With this method, the particles have a tracer mass proportional to their total mass (Kort *et al*., 2022).

The generator is used with its P&S 360 kit in the same configuration as shown in Figure 6. The P&S 360 kit includes a generator nozzle (integrating the generator) and its tripod, an electronic controller for the generator, an IHM (Human Machine Interface, not visible in the figure), a peristaltic pump equipped with its solution flow controller, a tank for the solution, pipes for supplying the generator with solution and cables (not visible in the figure). This assembly supplies solution to the generator and controls particle generation.

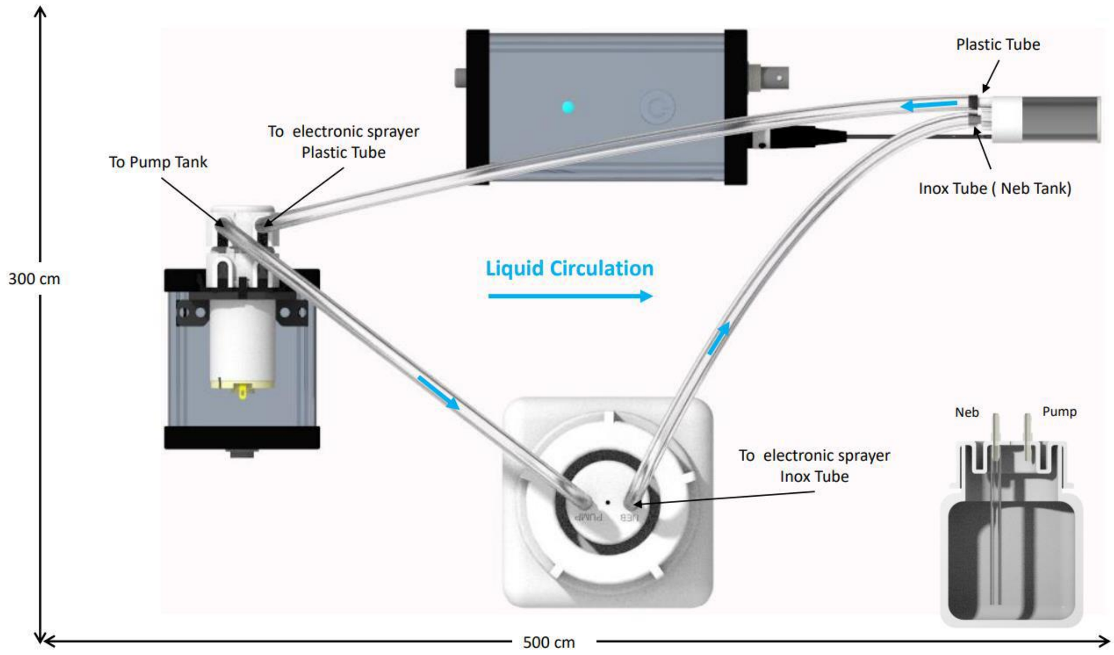

Figure 6: Overall view of the assembly of the MICRONICE® 12 µm generator and the P&S 360 particle generation kit (IHM not visible) (source: TEKCELEO.com)

In the present study, the following compounds are used to generate particles:

- tracer: sodium fluorescein;
- main constituent (low volatility liquid): glycerol;
- volatile solvent: pure ethanol.

2.1.2 Particle characterisation

The particle size distribution of the generated particles is characterised by direct measurement of their diameter and by indirect measurement, based on a sample taken with a cascade impactor followed by deferred measurement of fluorescein by spectrometry.

Samples for deferred fluorescein measurements are realized using a Low Pressure Impactor (LPI) from DEKATI Inc (Figure 7).

Figure 7: View of an LPI DEKATI Inc. (source: DEKATI.com)

The LPI is a column made up of 13 stages of stacked conventional inertial impactors, operating at low pressure (100 mbar at the foot of the column). Assembled in cascade, the first stages collect the largest particles, while the smallest impact on the lower stages. The cut-off diameters of these stages correspond to aerodynamic diameters. Particles are separated into 13 particle size classes ranging from 30 nm to 10 µm (or even more, with the first stage collecting all particles larger than 10 µm).

Fluorescein is measured using a JOBIN YVON HORIBA Fluoromax-3 fluorescence spectrometer (Figure 8).

Figure 8: View of a HORIBA Fluoromax-3 (source: HORIBA.com)

The spectrometer measure by fluorescence the concentration of fluorescein in solutions. For the measurement, particles taken from the LPI's impactors are dissolved in known volumes of aqueous solution. These solutions are measured and the masses of fluorescein (and therefore of particles) sampled are determined by calculation. The fluorescence spectrometer returns a "counts per second" signal for each sample. To calculate the concentrations associated with the "counts per second" signals, fluorescein standard solutions are used to determine a calibration curve.

Direct measurement of particles concentrations, in particular as a function of their diameters, is carried out using an Electrical Low Pressure Impactor+ (ELPI+) from DEKATI Inc (Figure 9).

Figure 9: View of an ELPI+ DEKATI Inc. and of its impaction column (source: DEKATI.com)

The ELPI+ measures particles in real time by electrical discharge in a low-pressure cascade impactor (same operating principle as the LPI), after they have been charged by the corona effect. It measures the concentration of particles in the air for 14 size classes between 6 nm and 10 µm. The result is a concentration measurement for each class and therefore a particle size distribution as a function of their aerodynamic diameter.

2.2 Method

The development of the in-environment generation method of particles between 4 µm and 5 µm and the characterisation of their size distribution are carried out in 2 steps:

- a first step to determine the appropriate composition of the solution to be used to generate particles with the wanted distribution. This stage is carried out in the laboratory;
- a second stage in which the generator and the solution are tested outdoors, under test conditions.

During the first step, consecutive tests are carried out with different compositions of solutions until the right composition is found. The chosen composition must be able to generate particles with diameters of between 4 µm and 5 µm and have a high enough fluorescein content to enable them to be quantified after dispersion and deposition in the environment.

The solutions are tested in the laboratory with the generator placed in a fume hood (Figure 10). Measurements are realized in real time using the ELPI+.

Figure 10: View of the generator and a solution containing fluorescein and glycerol during a test in the laboratory in a fume hood

Following this first stage, the generator is tested, in the second step, with the solution of the chosen composition (Figure 11). The generator is battery-powered and positioned outdoors in real atmospheric conditions. Characterisation of the particle size distribution is then determined on the basis of a sample taken with the LPI (measurement with the LPI enables only fluorescein to be quantified, whereas the ELPI+ counts all the particles present in the air, regardless of their chemical nature). This step ensures that the particle size distribution does not change between the two configurations, in the laboratory and outdoors (test conditions).

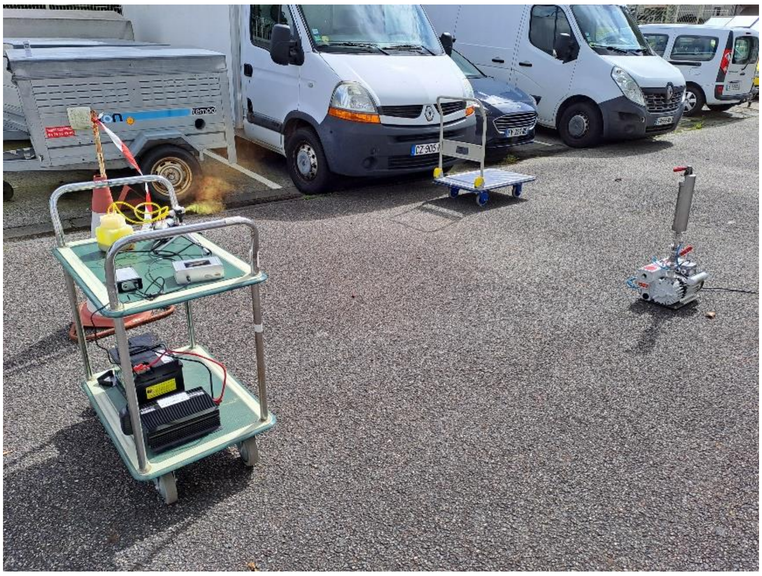

Figure 11: View of the generator in "test" configuration and the LPI during an outdoor test

3 Results - characteristics of the generated particles

3.1 Laboratory tests

A total of 9 tests have been carried out in laboratory. The concentrations of the components of the tested solutions have varied in the following ranges:

- sodium fluorescein: from 4 g.L^{-1} to 8 g.L^{-1} (dissolved in the solution);
- glycerol: de 1 % à 10 % by volume;
- pure ethanol: from 90% to 99% by volume.

Table 2 provides a summary of the tests. It shows the compositions of the solutions, the mean and median mass aerodynamic diameters, and the associated geometric standard deviations, calculated from measurements realized with the ELPI+.

The mass mean aerodynamic diameters obtained from a lognormal fit on the experimental points measured with the ELPI+ have varied between 3.2 µm and 5.5 µm, with geometric standard deviations varying from 1.8 to 2.0.

The mass median aerodynamic diameters have been determined using a Henry curve. They have varied between 2.9 µm and 4.9 µm, with geometric standard deviations varying between 1.8 and 2.1.

Reproducibility tests have been carried out for 3 solution compositions in tests 1, 2, 4, 5, 8 and 9. The results show a good reproducibility of the particle generation, with particle size distributions well preserved between two tests with the same solution.

Based on these results, the composition of the solution selected for outdoor testing was that tested in trials 8 and 9: 8 g.L^{-1} fluorescein, 7% glycerol by volume and 93% pure ethanol by volume. The particle size distributions obtained with this solution show a mass mean aerodynamic diameter of 4.8 µm with a geometric standard deviation of 1.9, and a mass median aerodynamic diameter of 4.5 µm with a geometric standard deviation of 2.1. These values are very close to those reported in the literature for concrete particles. The particles generated are 5% to 7% larger in diameter than concrete particles.

Test number	Solution composition (% by volume)	Mass mean aerodynamic diameter and associated geometric standard deviation	Mass median optical diameter and associated geometric standard deviation
1	Fluorescein 4 g.L^{-1} / Glycerol 1 % / Ethanol 99 %	3,2 µm / 1,8	2,9 µm / 1,8
2	Fluorescein 4 g.L^{-1} / Glycerol 1 % / Ethanol 99 %	3,4 µm / 2,0	3,1 µm / 1,9
3	Fluorescein 4 g.L^{-1} / Glycerol 2 % / Ethanol 98 %	3,7 µm / 1,9	3,3 µm / 2,0
4	Fluorescein 8 g.L^{-1} / Glycerol 4 % / Ethanol 96 %	4,1 µm / 1,9	3,8 µm / 2,0
5	Fluorescein 8 g.L^{-1} / Glycerol 4 % / Ethanol 96 %	4,2 µm / 1,9	3,8 µm / 2,1
6	Fluorescein 8 g.L^{-1} / Glycerol 10 % / Ethanol 90 %	5,5 µm / 2,0	4,9 µm / 2,2
7	Fluorescein 8 g.L^{-1} / Glycerol 8 % / Ethanol 92 %	4,7 µm / 1,8	4,5 µm / 2,1
8	Fluorescein 8 g.L^{-1} / Glycerol 7 % / Ethanol 93 %	4,8 µm / 1,9	4,4 µm / 2,1
9	Fluorescein 8 g.L^{-1} / Glycerol 7 % / Ethanol 93 %	4,8 µm / 1,9	4,5 µm / 2,1

Table 2: Summary of laboratory tests

The particle size distribution obtained with the ELPI+ for the particles generated with this solution in test 9 is shown in Figure 12.

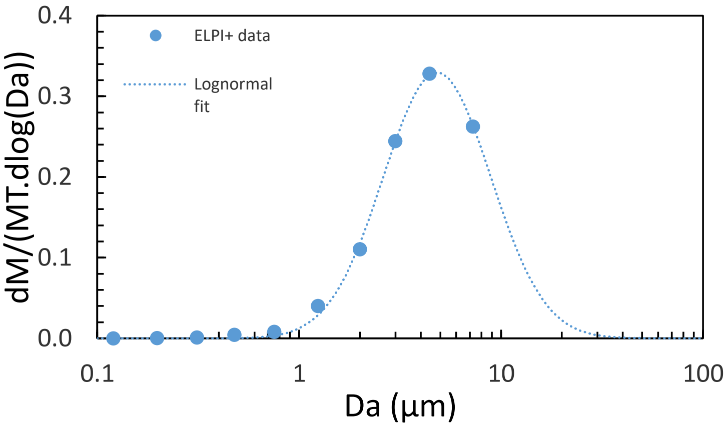

Figure 12: Particle size distribution obtained with the solution selected for being tested in an outdoor test configuration (experimental points) and fitted lognormal distribution for determining the mass mean aerodynamic diameter and geometric standard deviation

3.2 Outdoor test

During this test, the generator has been tested in "field" configuration, i.e., powered by a battery and a voltage converter, with the solution chosen in the configuration shown in Figure 11. The generator has been placed on a trolley at a height of around 90 cm. The LPI was positioned downwind of the generator, 2.5 m away.

Meteorological conditions were dry (22°C and 46% relative humidity).

The generator has emitted particles for 15 minutes. The particles generated have benne sampled with the LPI throughout the generation period, and the samples taken from each impactor have been then measured in the laboratory using the fluorescence spectrometer.

The particle size distribution obtained is shown in Figure 13.

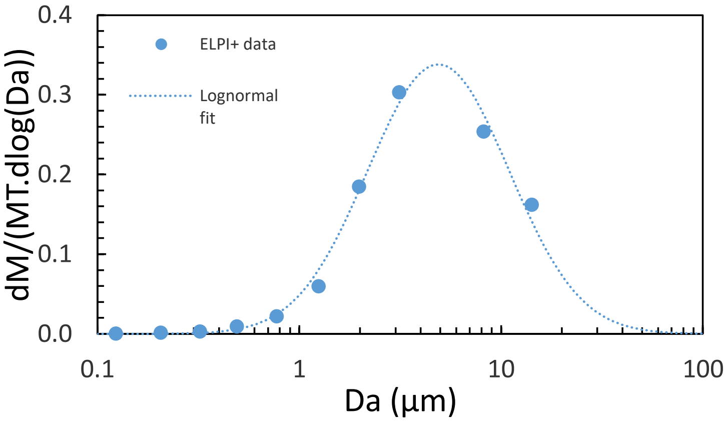

Figure 13: Particle size distribution of particles generated outdoors (experimental points) and fitted lognormal distribution for determining mass mean aerodynamic diameter and geometric standard deviation

The particle size distribution obtained outdoors shows a mass mean aerodynamic diameter of 4.9 µm with a geometric standard deviation of 2.2, and a mass median aerodynamic diameter of 5.2 µm with a geometric standard deviation of 2.6. The generated particles are 21% to 24% larger in diameter than concrete particles.

These values are very close to those obtained in the laboratory and validate the method of generating, outdoors, polydisperse micron particles of 4 µm to 5 µm in diameter, representative of concrete particles suspended during dismantling work.

Conclusion

The data in the literature was used to determine the particle size distributions of concrete and stainless steel particles that may be suspended during dismantling work. A polydispersed particle size distribution with a mass median aerodynamic diameter of the order of 4 µm to 5 µm, representative of concrete particles, was selected for the study of dry and wet deposition of tritiated particles in the environment.

A method for generating particles containing a tracer has been tested in the laboratory. It permits to generate polydispersed particles of glycerol containing fluorescein with a mass median aerodynamic diameter of 4.5 µm and a geometric standard deviation of 2.1.

This method has been successfully tested under environmental conditions. The generator is battery-powered and the particle size distribution is maintained.

The particle generation method presented in this report can be used to measure dry and wet particle deposits in the environment.

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