

SF1- FILTRATION TESTS FOR OPTIMIZATION OF SCCD TREATMENT

1.1- Experimental protocol

The experimental test bench used to carry out the optimization phase, in terms of pressure and temperature, of the SCCD protocol is presented in Figure SF1- 1.

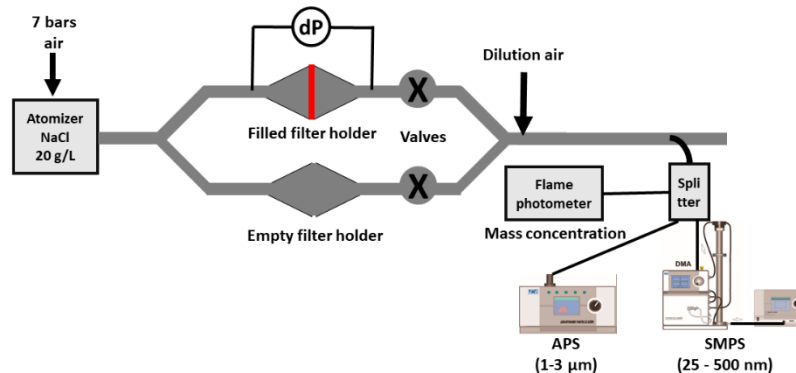


Figure SF1- 1: experimental test bench used to carry out the optimization phase, in terms of pressure and temperature, of the SCCD protocol

Sodium chloride aerosol is produced by a Collison type atomizer (Collison 4100 250F). Count median diameters of aerosol of 74 nm (IRQ of 79 for Q25-Q75 of 44-123 i.e. for a size distribution with a Geometric Standard Deviation of 2.04, characteristic of requested polydisperse) were reported for both lines 1 and 2 (see Figure SF1- 2). Prior measurement, carried out with an Electrical low pressure impactor, has confirmed an aerodynamic mass median diameter of 610-640 nm for this atomizer (with a Geometric Standard Deviation of 3). Filtration efficiency measurement is based on two separate aerosol transport ducts, both instrumented with the same filter holder. 47 mm diameter samples (real filtration surface associated to 36 mm diameter) are prepared directly from FFP2 mask and placed in the upper filter holder (line 1). The second filter holder is left empty and is used to characterize the aerosol size distribution upstream tested filter (line 2). To support this measurement protocol, aerosol size distributions for both filter holder lines have been measured with a Scanning Mobility Particle Sizer (X-ray neutralizer 3088, classifier 3082 with long DMA column 3081 and particle counter CPC 3775 from TSI). Comparison of measured size distributions are presented in Figure SF1- 1 part right and a close agreement could be noticed, confirming similar transport of aerosol within both lines. In addition to number-based SMPS measurement, aerosol mass concentration is measured by emission spectroscopy using MOORE'S Low flow photometer type 1250. A second size spectrometer is also reported in Figure SF1- 1 (aerodynamic particle size TSI 3021) for measuring particle size in the range 1-3 μm (in terms of aerodynamic diameter) for testing surgical or barrier masks⁴. Since present paper focus on FFP2 mask, we won't discuss results obtained with APS and will only consider SMPS and emission spectroscopy measurements.

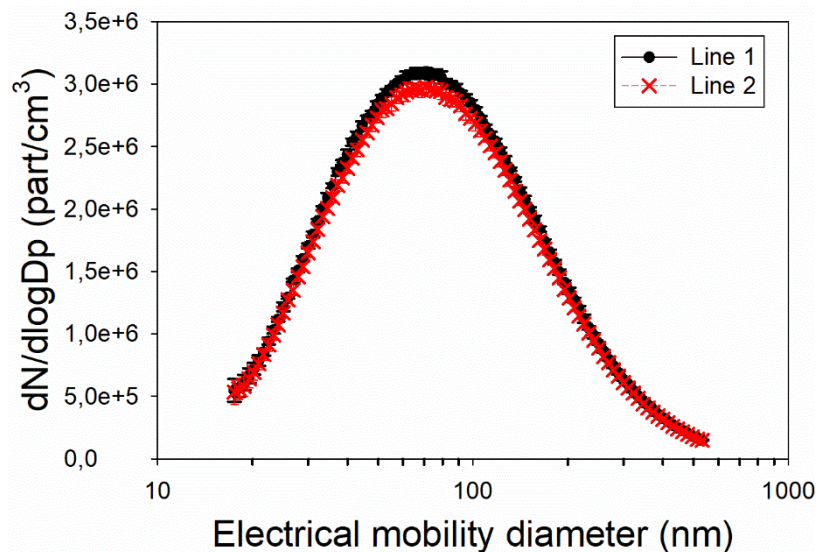


Figure SF1- 2: comparison between aerosol size distributions measured in lines 1 and 2

Experimental protocol considered for measuring the filtration efficiency (number or mass-based at 9.6 cm/s) during the optimization phase is described hereafter:

- Measurement of two size distributions by APS and SMPS on the « empty line » corresponding to the first upstream analysis (upstream 1), in parallel, mass concentration of sodium chloride particles is measured by emission spectroscopy,
- Measurement of six size distributions by APS and SMPS on the « filled line » corresponding to downstream analysis (downstream), in parallel, mass concentration of sodium chloride particles is measured by emission spectroscopy,
- Measurement of two additional size distributions by APS and SMPS on the « empty line » corresponding to the second upstream analysis (upstream 2), in parallel, mass concentration of sodium chloride particles is measured by emission spectroscopy,
- Computation of mean aerosol size distribution measured by APS and SMPS on one hand from all upstream measurements (« upstream 1 » and « upstream 2 ») and on the other hand from the size distribution measurements conducted downstream the filter,
- Computation of mass and number-based filtration efficiencies.

1.2- Optimization phase

The optimization phase was achieved by comparison of six different SCCD protocols in terms of filtration efficiency on FFP2 from Kolmi OpAir Pro Oxygen and 3M VFLEX 9152 with close initial efficiencies (see Figure SF1- 4). Figure SF1- 3 presents the evolution of non-dimensional mass-based filtration efficiency as a function of pressure and temperature imposed during the SCCD protocol. Dealing with pressure, the highest non-dimensional filtration efficiency is noticed for the lowest pressure (7.5 MPa). Regarding temperature, since optimization has been conducted for a pressure of 14-15 MPa, we were not able to reach a filtration efficiency close to initial one, nevertheless as temperature increases, filtration efficiency increases. Mutual effects of both pressure and temperature were considered by computing CO₂ density (Figure SF1- 3, right). Most relevant SCCD conditions were finally identified as 7.5 MPa, 343 K corresponding to an optimum CO₂ density of 0.15 g/ml.

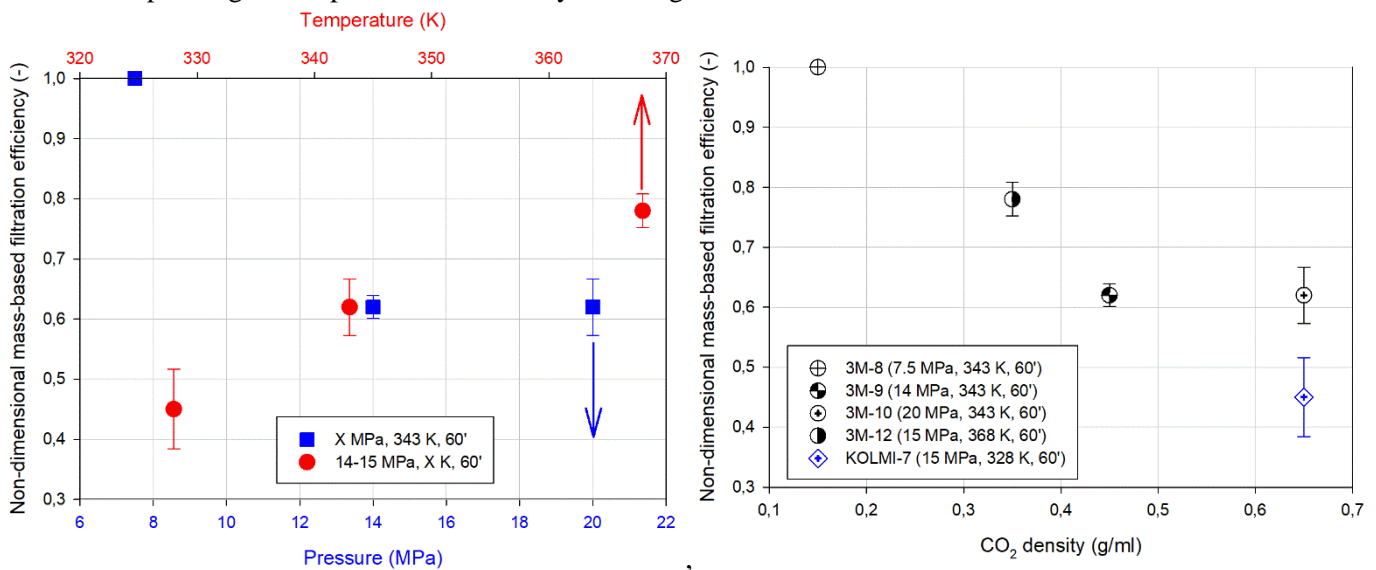


Figure SF1- 3: evolution of non-dimensional mass-based filtration efficiency as a function (left) of pressure (blue squares), temperature (red dots) and CO₂ density (right)

Beyond this statement, analysis of filtration efficiency versus particle diameter brings additional information on the evolution of filtration efficiency after SCCD treatment. Figure SF1- 4 presents the evolution of number-based filtration efficiency as a function of electrical mobility diameter (measured by the SMPS) for three different FFP2 masks untreated (3M, KOLMI and Paul Boyé) and after SCCD (3M and KOLMI) or dry/wet cleaning protocols (Paul Boyé). Figure SF1- 4 shows, for the non-optimised SCCD protocols (3M-9, 3M-10, 3M-12 and KOLMI-7), a significant decrease, associated to a shift of the diameter with minimum efficiency (corresponding to the most penetrating particle size MPPS) from nearly 20-40 nm for untreated mask to 200-300 nm for SCCD treated ones. This shift, also noticed in Figure SF1- 4, for dry and wet cleaning reported (Paul Boyé masks), was identified in the classical filtration theory for electrically charged filtration medium and is due to a modification of the electrostatic properties of the medium.

For optimised conditions (3M-8), no significant shift of MPPS could be identified and the overall and spectral filtration efficiencies remain poorly affected confirming relevant temperature, pressure and CO₂ density.

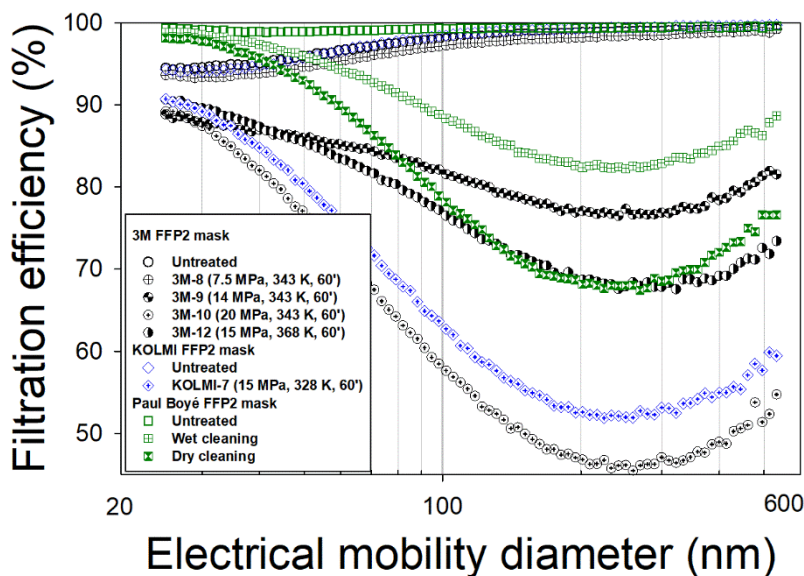


Figure SF1- 4: evolution of number-based filtration efficiency (measured for a filtration velocity of 9.6 cm/s) as a function of electrical mobility diameter for untreated and treated masks. Samples 3M-8, 3M-9 and 3M-10 were considered for investigating SCCD pressure effect while samples 3M-9, 3M-12 and KOLMI-7 were considered for the temperature effect.

Table SF1- 1 presents experimental conditions associated to tested samples (at 9.6 cm/s) and their mass-based filtration efficiency.

Table SF1- 1: samples properties and experimental conditions

Sample	Treatment protocol	Mass-based filtration efficiency
3M VFLEX 9152	untreated	99.3 +/- 0.1 %
3M-8	SCCD 7.5 MPa, 343 K, 0.15 g/ml CO ₂ , 60'	99.2 +/- 0.1 %
3M-9	SCCD 14 MPa, 343 K, 0.45 g/ml CO ₂ , 60'	84.8 +/- 1.9 %
3M-10	SCCD 20 MPa, 343 K, 0.65 g/ml CO ₂ , 60'	61.4 +/- 4.7 %
3M-12	SCCD 15 MPa, 368 K, 0.35 g/ml CO ₂ , 60'	77.7 +/- 2.8 %
KOLMI OpAir Pro Oxygen	Untreated	99.5 +/- 0.1 %
KOLMI-7	SCCD 15 MPa, 328 K, 0.65 g/ml CO ₂ , 60'	45.1 +/- 6.6 %
Paul Boyé untreated (MPB2.1)	Untreated	99.7 +/- 0.1 %
Wet cleaning	60°C in water with detergent	89.8 +/- 1.2 %
Dry cleaning	Dry cleaning	82.2 +/- 2.1 %

SF2- FILTRATION NORMS, PROTOCOL, RESULTS

2.1- Filtration test set-up and protocols

2.1.a- Norms defining protocols and limits for FFP2 certification

This work was designed to support the particulate filter test requirements according to EN149: 2001+A1: 2009(F) and to EN13274-7: 2019(F), respectively for FFPs and for filters cut in FFPs (cf. Table SF2- 1). Resistance to air inhalation (pressure drop) and mass filtration efficiencies were measured, for filters cut in FFP challenged with liquid Paraffin aerosol at 15.8 cm/s face velocity, related to peak Inhalation flowrates (PIF of 95 Lpm on 100cm² filtering media).

Table SF2- 1: Test conditions and classification criterion of FFPx (1/2 Filtering Face Piece not covering the eyes, FFPx), according to EN149: 2001+A1:2009 (F) ; with the same data over lined for the filtering media, called filters cut in FFP2 according to EN 13274-7 :2019 (F) June 2019.

Filtering medium or filter cut in 1/2 masks	FFP1	FFP2	FFP3
Penetration at 15.83 cm/s (%) Solid Aerosol: NaCl, D _{mode} = 0.06 µm and D _{50 mass} = 0.6 µm	20	6	1 %
Penetration at 15.83 cm/s (%) Liquid Aerosol : Paraffin D _{mode} = 0.3µm et D _{50 Num} =0.4µm	20	6	1 %
Inhalation pressure drop at 30 Lpm (5 cm/s)	60	70	100 Pa
Inhalation pressure drop at 95 Lpm (15.83 cm/s)	210	240	300 Pa
Exhalation pressure drop at 160 Lpm (- 26.9 cm/s)	300	300	300 Pa
Maximal total incoming leak for wear FFP _x	22	8	2 %

2.1.b- Set-up for filtration performances measurements

Figure SF2- 1 depicts the set-up to measure Filtration performances of filters cut in FFP2. This includes the resistance to respiration flow (pressure drop of the filter) and both the total mass efficiency (mass fraction of particles collected in the filter) defined as the complement to unity of penetration (mass fraction of particles passing through) and the fractional efficiency versus the test aerosol diameter. The total mass filtration efficiency is defined as:

$$\text{Efficiency} = 1 - P = 1 - \frac{C_{\text{downstream}}}{C_{\text{upstream}}} = 1 - \frac{C_m^{\text{OUT}}}{C_m^{\text{IN}}} \quad ; \quad \text{Penetration} = \frac{C_{\text{downstream}}}{C_{\text{upstream}}} = \frac{C_m^{\text{OUT}}}{C_m^{\text{IN}}}$$

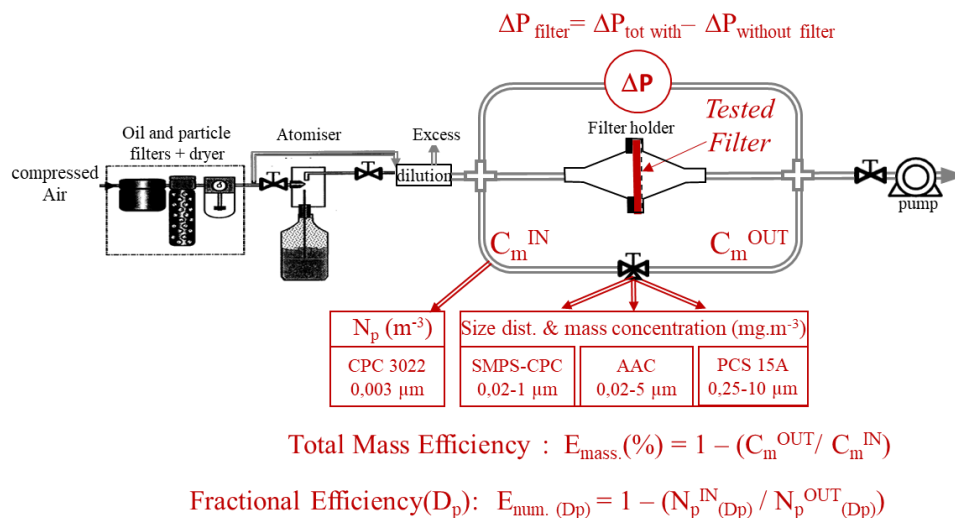


Figure SF2- 1: Filtration test set-up

- Aerosol generator is a constant output nebuliser (TSI 3076): Liquid particles of Paraffin are produced with the corresponding unimodal number size distribution depicted on Figure SF2- 2b (modal diameter of 0.3 µm, with median diameter, $d_{50}^{\text{NUM.}} = 0.4$ µm, and Geometric Standard Deviation of 1.65 as defined on fig. 6 p14 in the NF76-

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007, from the EN13274-7:2019 (F)). A dilution device then adjust the droplet concentration from 10^8 to 10^5 cm^{-3} , down to normative test conditions for concentrations from 15 to 25 mg/m^3 .

- Filter holder according to norms with fitting screws and support grid, ($S_{\text{Filtration}} = 8.04$ cm^2 for 32 mm diameters filters cut in FFP2). The actual filtration surface is lower than the recommended 100 cm^2 to limit eventual heterogeneities of filters. However, this does not affect the reproducibility of pressure drop and mass efficiencies on different filters from the same mask or even from different masks from the same lot (defined for each treatment and each FFP2 trademark).
- Manometer and flow meters calibrated in January 2020, to measure the pressure drops at the 3 normative references velocities of 5 and 15.83 cm/s for inhalation and -26.9 cm/s for expiration. Before any measurement, the tightness of the test unit is checked by following the pressure after depressurising the line.
- Aerosol measurements: to operate in stable conditions of aerosol production, a Condensation Particle Counter CPC (CPC 3022 from TSI Inc® Minneapolis, calibrated in June 2019) follows the total upstream droplets concentration, in parallel with size and mass distributions upstream and downstream the filters, from 0.25 to 10 μm (PCS Palas GmbH® Karlsruhe, calibrated in July 2019). This optical detection used for particulate respirator certification was implemented by measurements of size distributions and total mass concentration of suspended droplets upstream and downstream the challenged filters for mass efficiencies calculations over the complete size range from 0.02 to 3 μm of the test aerosol. Selection versus the diameter of suspended droplets was performed based either on electrical mobilities using a Scanning Mobility Particle Sizer from 0.02 to 1 μm (Kr neutralizer 3077, with long DMA classifier column 3071 and particle counter CPC 3022 i.e. SMPS+CPC from TSI Inc.® Minneapolis USA) or on aerodynamic properties using the Aerodynamic Aerosol Classifier from 0.02 to 3 μm (AAC from Cambustion Ltd.®, Cambridge UK).

2.1.c- Filter samples preparation and protocol of filtration tests

Widely used masks labelled as FFP2 according to EN149 were tested. These FFP2 from Paul Boyé ® with a “duckbill” shape are made of two flat parts without expiration valve with a nasal clip for adjustment and two welded elastic binders.

FFP2 are stored in ambient conditions (HR from 40 to 90 % and T from 15 to 30 °C) in their original conditioning before being cut with a 47 mm diameter-cutting tool, just before the tests on 32 mm diameter of filter exposed to aerosol.

Each lot of FFP2 is labelled according to its initial state (new or soiled filters), the number of treatments with conditions (P, T, duration).

Filtration performances for FFP2 certification ($\Delta P < 240$ Pa and/or mass efficiency $> 94\%$) have been measured according to EN149 and EN13274-7. Measurements were thus repeated on at least three different filters (labelled 1, 2, 3 for the superior face with the nasal clip and 1', 2' et 3' for the lower face) and for different FFP2 (A, B, C...) from the same lot.

At last, measurements are not impacted for the few minutes tests in dry air, below the minimum relative humidity of 40 %, imposed by norms for exposition test of few hours performed on worn FFPs.

The initial efficiency, the resistance to inhaled air (pressure drop in Pa) and their evolution upon loading were measured three times 30 seconds, successively upstream, downstream and upstream the filter. Short term and time-averaged mass concentrations, with related mass-based filtration efficiencies were then calculated.

Finally, the exposition or loading test was performed on one sample. The mass efficiency and the pressure drop are continuously measured during the loading of the filter with 4 mg of liquid test aerosol (as extrapolated from the 120 mg loading recommended by EN149 for a FFP2 of 204 cm^2 for the Paul Boyé masks under test), then stored for 24 hours before the final measurements.

2.2 Properties of test Aerosol

2.2.a- Size distributions and concentrations of liquid test aerosol (Paraffin droplets)

Size distributions and concentrations of liquid test aerosol (paraffin droplets) first have to be validated with respect to normative test conditions to warrant the reliability of mass efficiencies calculated from these distributions. To achieve a complete size distribution including unmeasured smaller droplets than the optical cutting diameter of 0.25 μm , complementary size measurements are performed by SMPS from 0.02 to 1 μm or directly measured on the overall size range of oil droplets by aerodynamic method (AAC from 0.02 to 3 μm).

The Figure SF2- 2a depicts the three size distributions measured with different principles, either optical requested by EN149 and EN13274-7 for the Particle Sizer from Palas ® (PCS), sometimes implemented by electrical mobility selection to include droplets smaller than the optical cutting diameter of 0.25 μm or by aerodynamic principle (Aerodynamic Aerosol Classifier AAC from Cambustion® Cambridge UK).

Figure SF2- 2b depicts the number size distributions so reconstructed as well as the cumulated mass concentration.

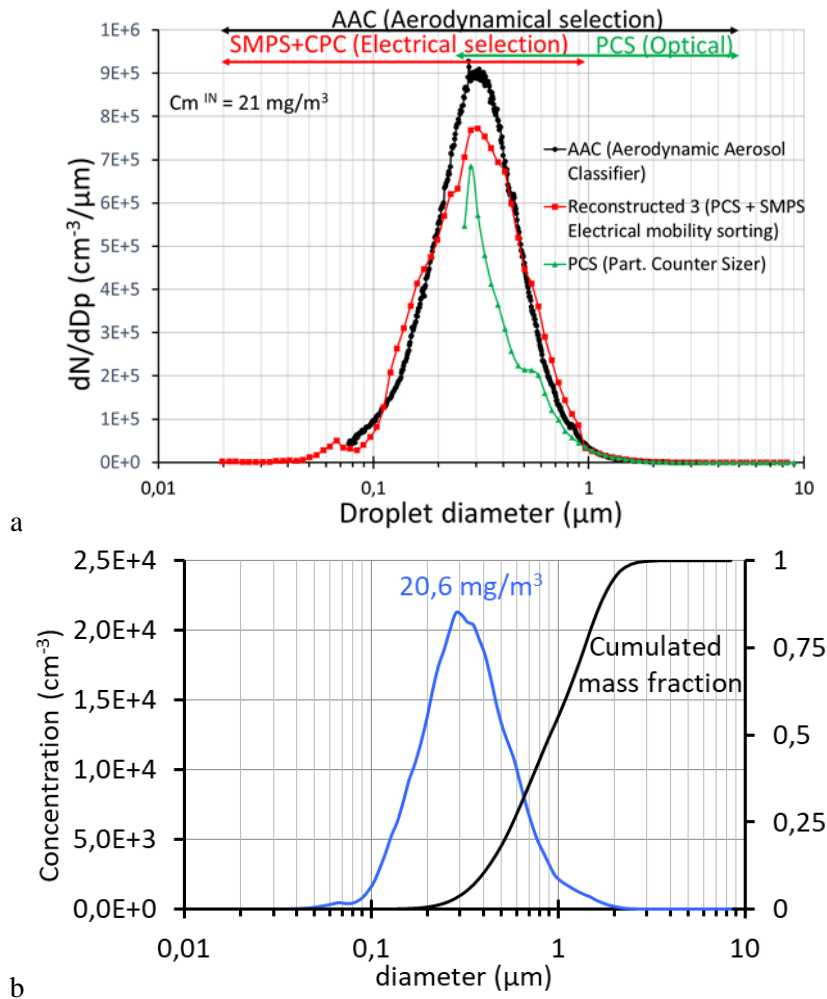


Figure SF2- 2: (a) Number size distributions of Paraffin droplets as challenge liquid aerosol defined from EN149: 2001+A1:2009 (F) and EN 13274-7 :2019 (F), normalized by the width of size intervals for comparisons of distributions measured by light diffusion with the PCS 2010-15A from Palas® GmbH, Karlsruhe (from 0.25 to 10 μm), by electrical mobility selection (SMPS+CPC 3022 from TSI.® Inc, Minneapolis, from 0.02 to 1 μm) and by aerodynamic Aerosol Classifier (AAC from Cambustion Ltd.®, Cambridge UK from 0.02 to 3 μm) (b) Complete size distribution and cumulated mass fraction.

The modes of the three number size distributions is closed to 0.3 μm with a median diameter of 0.397 μm (0.290-0.549 i.e. an IRQ of 0.259 i.e. for a size distribution with a Geometric Standard Deviation of 1.65, characteristic of requested polydisperse Aerosol), measured using optical PCS measurements by light diffusion, as well as electrical principle by SMPS or aerodynamic ones with AAC (cf. Figure SF2- 2a).

Figure SF2- 2b highlights that more than 95 % of the total aerosol mass is carried by droplets larger than 0.25 μm , within the measurement range of optical detection tools. This supports the interest of such optical measurements recommended by norms to calculate mass efficiencies.

2.2.b- Electrical charge of the test aerosol

The net electrical mean charge of paraffin droplets was measured by a Faraday cup Electrometer 3068 from TSI. Inc® Minneapolis.

The fraction of positively charged particles lies below 0.1% of the total number concentration of paraffin droplets with a net charge measured as positive but below the fA detection limit, corresponding to a mean charge per particle lower than 10^{-5} e, meaning that only one particle is singly charged positively over 10^5 .

2.3- Experimental results

Table SF2- 2 describes samples and test conditions of all filters (including reference, serial number of FFP2, numbers and locations of filter samples cut in FFP2, flowrates) for the different lots of FFP2 i.e. versus the initial new or soiled filter, the trademark and the treatment with conditions. It synthesises the measured values of pressure drops versus flowrates and mass efficiencies at 15.8 cm/s, with standard deviations for each filter as well as the mean values among different filters per lot with standard deviation.

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Table SF2- 2: Labelled samples of filters cut in FFP2 with references from the producer, test conditions, mass efficiencies and pressure drop for each filter tested with geometric standard deviation of measurements performed at least three times in a row for three times 30 seconds, as well as their mean values with standard deviation for different filters from the same lot, at 15.83 cm/s with a liquid Paraffin aerosol according to EN149:2001+A1:2009(F) and EN13274-7:2019(F).

Minimal mass Efficiency > 94% with maximal $\Delta P < 240$ Pa at 15.8 cm/s for FFP2 certification according to EN149

Treatment / trade mark of filters cut in FFP2	Type of FFP2, Ref, N° lot - expiration date	Number and location of filters cut in FFP2	initial Pressure drop (Pa) = ΔP_{tot} with - $\Delta P_{without\ filter}$			Filtration Efficiency (% at 15.8 cm/s) collected fraction of C_{mass}^{IN} [15-25] mg/m ³ of Liq. Paraffin Aerosol -- $-d_{mod}=0.3$ et $d_{50\ Num.}=0.4$ μm -
			2.44 Lpm 5cm/s	7.6 Lpm 15.8 cm/s	13 Lpm 26.7cm/s	
			- 8 Pa	- 55 Pa	- 140 Pa	
NEW reference filters						
NEW Paul Boyé CE 0333 EN 149: 2009 in tight conditioning for 10 masks	FFP2 NR MPB2.1 136/20 M4 15/24	ICMCB-A2		175		99.9 ± 0.02
		ICMCB-A2'		165		99.9 ± 0
		CHU2-B2'	54	150	260	99.9 ± 0
		CHU2-A2		150		99.7 ± 0.01
		CHU2-A2'		155		99.8 ± 0.04
		MEAN (5 filters)		159 ± 11		99.8 ± 0.05
Super Critical CO₂ (SCCD 60' @ 343 K and 7.5 MPa, 0.5 mL solution at 50/50 EtOH/H₂O₂ 30%)						
L15 (1*CO ₂) / new Paul Boyé CE 0333 EN149:2009	FFP2 NR, ref : MPB2.1 136/20 M4 Lot -15/24	A2	49	150	250	99.4 ± 0.1
		A2'	44	145	240	99.4 ± 0.02
		A1	44	145	240	99.4 ± 0.04
		MEAN (3)		147 ± 3		99.4 ± 0
L18 (1*CO ₂) / new Paul Boyé CE 0333 EN149:2009	FFP2 NR, ref : MPB2.1 136/20 M4 Lot -15/24	A2	44	145	240	98.4 ± 0.1
		A2'	44	145	240	98.5 ± 0.03
		A1	40	130	230	
		MEAN (2)		140		98.5 ± 0.1
		± 9				
1*CO₂/ new Paul Boyé L15+L18		MEAN (5 = L15+L18)		143 ± 7		99 ± 0.5
L2sccd (2*CO ₂) / new Paul Boyé CE 0333 EN149:2009	FFP2 NR, ref : MPB2.1 136/20 M4 Lot -15/24	A2		155		98.8 ± 0.04
		A2'		165		99.2 ± 0.03
		A1		135		98.2 ± 0.01
		MEAN (3)		152 ± 15		98.8 ± 0.5
L29'-30' (1*CO ₂) /P.Boyé soiled with spores CE 0333 EN149:2009	FFP2 NR, ref : MPB2.1 136/20 M4 Lot -15/24	A2		155		98.8 ± 0.1
		A2'		155		99 ± 0.04
		A1		150		98.8 ± 0.1
		MEAN (3)		153 ± 3		98.9 ± 0.1
EXPOSITION and STORAGE TESTS / new PBoyé post- SCCD CE 0333 EN149:2009	FFP2 NR, ref : MPB2.1 136/20 M4 Lot -15/24	A3' at t°		120 Pa		99.4 ± 0.03
		at t _{f Expo} (+ 4 mg)		130 Pa		99.1 ± 0.04
		at t _{f storage} = t _{f Expo} + 24 h		127 Pa		98.2 ± 0.2
Dry cleaning "optimised 1" (15' with soap(Intense ® & Activate ®) + 5' spinning + 40' drying)						
L266: dry cleaning / used Paul Boyé collected the 2nd/04/2020	FFP2 NR, unreadable reference	A2		155		87 ± 0.1
		A2'		165		87 ± 0.2
		A1		135		87 ± 0.2
		MEAN (3)		152 ± 15		87 ± 0.3
Autoclave 20' @ 394 K (with liquid water condensation)						
L262 / used Paul Boyé collected the 2nd/04/2020 CE0333 EN 149 2009	FFP2 NR, ref : MPB2.1 067/20 M3 11/24 -	A2				90.5 ± 0.2
		A2'				90.3 ± 0.1
		A1				90.7 ± 0.1
		MEAN (3)				90.5 ± 0.2

NB: Efficiencies are given with one significant digit above 90% and approximated to unity below, here and in Figure 4 of the article, due to larger uncertainties from tenth of % from 99 to 99.9 % efficiencies, to few % for lower ones below 90%, as detailed below in uncertainties section.

2.3.a- Reproducibility of measurements

These calculated pressure drops and mass efficiencies are reproducible for all trademarks, on different filter samples from the same FFP2 as well as from different masks from the same lot (cf. SD in Table SF2- 2). This supports the tightness of the test set-up and the stability of test conditions, which are critical for the reproducibility of pressure drops versus face velocities and mass concentrations measurements, i.e. for the reliability and reproducibility of calculated efficiencies at 15.8 cm/s.

New and treated FFP2 are homogeneous, since different filters from the same lot lead to close measured values of mass concentrations and efficiencies with standard deviation of mean efficiencies per lot, addressed in Table SF2- 2 always lower than 0.5 %, in the worst case. This homogeneity of filtering media cut in FFP2 confirms discrepancies recorded from one lot to another (i.e. for different initial new or soiled filter, trademark or treatment of each lot) are significant, as detailed below.

2.3.b- Uncertainties, significant discrepancies between measured efficiencies and normative limits

Uncertainties on pressure drop

Despite measurements uncertainties of 5%, mainly due to precision reading on differential pressure with reference atmospheric pressure, uncertainties on calculated values is lower than 9 % for pressure drop ($\Delta P = 195 \text{ Pa} \pm 17.6 \text{ Pa}$) and stands for pressure drops from 120 to 240 Pa.

Uncertainties on mass concentrations

C_m^{IN} and C_m^{OUT} are the mass concentrations, respectively upstream and downstream the filter.

Uncertainties on concentration measurement C_m (written as $U(C_m)$) are first calculated with 95 % confidence interval from the theory of uncertainties propagation, as the square root of the sum of the three kinds of uncertainties:

- $U_{Rep}(C_m)$ is related to the reproducibility defined as the standard deviation of measurements (σ_m) in a given condition $U_{rep}(C_m) = 2.7 \sigma_m$ where 2.7 is the coefficient for a number of measurements below 100, at 95 % confidence interval.
- $U_{App}(C_m)$ is related to the resolution and the calibration of the measurement apparatus. This uncertainty is evaluated by comparison with measurements based on different physical principles by mobilimetry and aerodynamic measurements.
- $U_{Cond}(C_m)$ is related to the discrepancies between real conditions and the conditions taken for the calculation (surface of the filter, gas flow rate...)

The mass concentration is calculated as:

$$C_m = \sum_i \frac{4 \pi \rho_{paraffine} R_{p,i}^3 N_i}{3 v_p S_{detector} \Delta t_{measurement}}$$

where $\rho_{paraffine}$ is the density of the paraffine, $R_{p,-i}$ is the mean radius of a droplet in the measurement range i , N_i is the number of particle detected during the measuring time $\Delta t_{measurement}$ through the detector surface $S_{detector}$ and v_p the velocity of the droplet in the measuring volume. $\frac{u(R_{p,i})}{R_{p,i}}$ is calculated using the size range for a diameter of 1 μm i.e. 0.075 μm .

$$\frac{U_{Cond}(C_m)}{C_m} = \frac{U(\rho_{paraffine})}{\rho_{paraffine}} + 3 \frac{U(R_{p,-i})}{R_{p,i}} + \frac{U(N_i)}{N_i} + \frac{U(v_p)}{v_p} + \frac{U(S_{detector})}{S_{detector}} + \frac{U(\Delta t_{measurement})}{\Delta t_{measurement}}$$

Finally, the total uncertainties on the mass concentration detailed in Table SF2- 3 is calculated as:

$$U(C_m) = \sqrt{U_{rep}(C_m)^2 + U_{app}(C_m)^2 + U_{cond}(C_m)^2}$$

Uncertainties on mass filtration efficiencies $E_m = 1 - \frac{C_m^{OUT}}{C_m^{IN}}$

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This uncertainty is calculated with respect to the theory of uncertainties propagation as the square root of the sum of the squares of $U_{Rep}(E_m)$ (with at least 3 measurements) and of the partial derivative of E_m with respect to C_m^{IN} and C_m^{OUT} times the uncertainties on C_m^{IN} and C_m^{OUT} respectively, neglecting correlation between C_m^{IN} and C_m^{OUT} .

$$U(E_m) = \sqrt{U_{Rep}(E_m)^2 + \left(\frac{\partial E_m}{\partial C_m^{OUT}}\right)^2 U(C_m^{OUT})^2 + \left(\frac{\partial E_m}{\partial C_m^{IN}}\right)^2 U(C_m^{IN})^2}$$

with $\frac{\partial E_m}{\partial C_m^{OUT}} = \frac{1}{C_m^{IN}}$ and $\frac{\partial E_m}{\partial C_m^{IN}} = -\frac{C_m^{OUT}}{C_m^{IN^2}}$

$$U(E_m) = \sqrt{(2,7\sigma(E_m))^2 + \left(\frac{1}{C_m^{IN}}\right)^2 (U_{Rep}(C_m^{OUT})^2 + U_{App}(C_m^{OUT})^2 + U_{Cond}(C_m^{OUT})^2) + \left(-\frac{C_m^{OUT}}{C_m^{IN^2}}\right)^2 (U_{Rep}(C_m^{IN})^2 + U_{App}(C_m^{IN})^2 + U_{Cond}(C_m^{IN})^2)}$$

The different uncertainties required to evaluate mass and efficiency total uncertainties are given in Table SF2- 3.

The mass measured by the mandatory optical measurements with respect to normative protocols is systematically underestimated for both C_m^{IN} and C_m^{OUT} . Thus, to confirm the validity of measurements presented here, these mandatory optical measurements were also compared with others based on different physical principles by mobilimetry and aerodynamic measurements.

The underestimation of mass concentrations by optical measurements is larger for C_m^{OUT} than for C_m^{IN} due to the smaller size of the aerosol downstream the filter, closer or even below the minimal optical cutting diameter (0.25 μm). This induces a systematic over-estimation of the filtration efficiency calculated from optical measurements, as confirmed by the measurements based on other principles. Thus, uncertainties on mass and efficiency are not symmetrical, as depicted in Table SF2- 3 from calculation described above with the following assumptions. For systematically underestimated mass concentrations, the positive uncertainties are larger than negative ones (i.e. using $U_{App}^-(C_m) = 0$). Reversely for systematically overestimated filtration efficiency, the smaller positive uncertainty on efficiency is calculated using the smaller uncertainty on mass concentration (i.e. $U_{App}^-(C_m) = 0$), and the larger negative uncertainties on efficiencies using the larger uncertainty on mass concentration (i.e. $U_{App}^+(C_m) > 0$).

Table SF2- 3: Uncertainties on mass and efficiency

		Measurements		Uncertainties					Corresponding values			unit	
		mean	standard deviation	U_{Rep}	$U_{App+/-}$	U_{Cond}	U^-	U^+	min	mean	max		
New Paul	C_m^{IN} (mg/m3)	24.053		1.055	12.0267/7	2.405	2.626	12.310					mg/m3
	C_m^{OUT} (mg/m3)	3	0.3910	0.031	0	0.004	0.032		21.43	24.05	36.36	3	mg/m3
	Efficiency	0.0481	0.0118	0.001	0.0423/0	8	1	0.0531	0.02	0.05	0.10	3	%
Boyé's FFP2	Efficiency	0.9980	0.0005	0.001			0.002	8	0.0019	99.52	99.80	99.99	%
1*SCCD	C_m^{IN} (mg/m3)	21.550		1.533		2.155	2.645	11.094					mg/m3
	C_m^{OUT} (mg/m3)	0	0.5680	0.005	10.775/0	0	0	9	18.91	21.55	32.64	3	mg/m3
	Efficiency	0.2155	0.0020	0.013	0.3233/0	6	2	0.3240	0.19	0.22	0.54	3	%
/ new PB	Efficiency	0.9900	0.0050	0.013			0.020	9	0.0136	96.91	99.00	100.00	%
1* SCCD / PB	C_m^{IN} (mg/m3)	22.050		1.676		2.205	2.770	11.367					mg/m3
	C_m^{OUT} (mg/m3)	0	0.6210	0.015	11.025/0	0	1	7	19.28	22.05	33.42	3	mg/m3
	Efficiency	0.2426	0.0058	0.002	0.3638/0	3	8	0.3650	0.21	0.24	0.61	3	%
soiled with spores	Efficiency	0.9890	0.0010	0.002			0.017	7	0.0033	97.13	98.90	99.23	%
2*SCCD	C_m^{IN} (mg/m3)	21.550		1.533		2.155	2.645	11.094					mg/m3
	C_m^{OUT} (mg/m3)	0	0.5680	0.018	10.775/0	0	0	9	18.91	21.55	32.64	3	mg/m3
	Efficiency	0.2586	0.0069	0.013	0.3362/0	9	8	0.3377	0.23	0.26	0.60	3	%
/ new PB	Efficiency	0.9880	0.0050	0.013			0.021	6	0.0137	96.64	98.80	100.00	%

Minimal and maximal efficiency measurements for FFP2 certification and rejection

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Based on similar uncertainties calculations (not detailed here), only filters reaching 96.6 % efficiency from optical measurements are considered as FFP2, fitting the normative limits of 94 % as minimal mass efficiency (with pressure drop below 240 Pa). Besides, filters with efficiencies lower than 93.1 % from optical measurements can be rejected. This maximal efficiency of 93.1 % for rejection is closer to 94% than the minimal measured efficiency for certification above 96.6 %, as expected from the smaller positive than negative uncertainty on efficiency, detailed just above. Despite a large range of efficiencies between 93.1 and 96.6 % which for the filter can not be certified, all the measured efficiencies are out of this ambiguous range and thus reliable for FFP2 certification above 96.6 % or exclusion below 93.1 %.

Student test to confirm significant discrepancies between mean measured efficiencies per lot and normative limit

To test the hypothesis that a sample of size n with mean filtration efficiency E_m and standard deviation σ has a significant discrepancy with the limit value of 94 %. The formula for the t -statistic parameter $t = \frac{E_m - 94}{\sigma / \sqrt{n}}$ has been used.

The p-value are reported in Table SF2- 4 for each filtration efficiency.

Table SF2- 4: p-values to prove the significant discrepancy between measured filtration efficiency and the critical value of 94 %

	efficiency E_m (%)	standard deviatio n (%)	number of measurement s	t- coefficient /94 %	p-value
New Paul Boyé's FFP2	99.8	0.05	5	259.38	< 0.0001
1*SCCD / new PB (L15+L18)	99	0.5	5	22.36	< 0.0001
1* SCCD / PB soiled with spores (L29'-30')	98.9	0.1	3	84.87	< 0.001
2*SCCD / new PB (L2scdd)	98.8	0.5	3	16.63	< 0.005
Autoclave 394 K (L262)	90.5	0.19	3	-7.58	< 0.03
Optimized dry cleaning 1	87	0.3	3	-8.08	< 0.03

2.4- Complementary results for Paul Boyé's FFP2s

2.4.a- Resistance to respiration flow (pressure drop of filter) versus inhalation face velocity of filtration

Filters are tested at different face velocities corresponding to Peak Inhalation Flow at 15.83 cm/s and to Low Inhalation Flow for moderate work activities at 5 cm/s, with respect to EN 149 for certification of FFPx. As FFP2, these filters should not overcome 70 Pa at 5 cm/s (30 Lpm on 100 cm² corresponding to 2.41 Lpm on 8.04 cm²) and 240 Pa at 15.83 cm/s, for 95 Lpm sur 100 cm² i.e. 7.64 Lpm on 8.04 cm²). Figure SF2- 3 first confirms the increasing pressure drop at higher filtration velocities^{1, 2}.

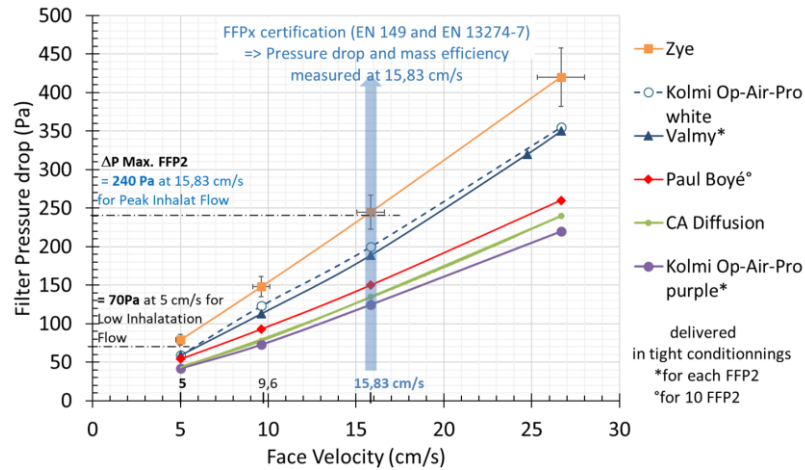


Figure SF2- 3: Pressure drops versus face velocity related to inhalation flow rates for filters cut in FFP2 of different trademarks. NB: For readability, error bars are only plotted for “Zye” filters but stand for all measurements.

For all tested trademarks, Valmy, Kolmi Op-Air-Pro (purple and white), Paul Boyé and CA Diffusion, pressure drops of 120 to 200 Pa are recorded at 15.8 cm/s. These different trademarks thus respect the maximal pressure drop authorized for certification as FFP2, as depicted on Figure SF2- 3 and in Table SF2- 2.

For the particular case of Paul Boyé's FFP2 presented here to check the SuperCritical Carbon Dioxide (SCCD) treatment for reuse of FFP2, three different lots of new FFP2 have been tested with pressure drop between 150 to 175 Pa, as well.

It has to be noted, since not detailed here that most treatments such as autoclave, dry cleaning and SCCD, lower the pressure drop of 10 to 30 Pa with respect to the original one of new filters.

2.4.b-Exposition and storage tests

Figure SF2- 4 highlights the evolution of pressure drop with the efficiency drop related to the loading with liquid Paraffin on surface as well as within the filter volume by fibre coating and pore clogging³. Pressure drops of Paul Boyé's filters cut in new FFP2 as well as after SCCD treatment increase of 5 to 15 Pa for few minutes exposition test when loaded with 4 mg of Paraffin droplets.

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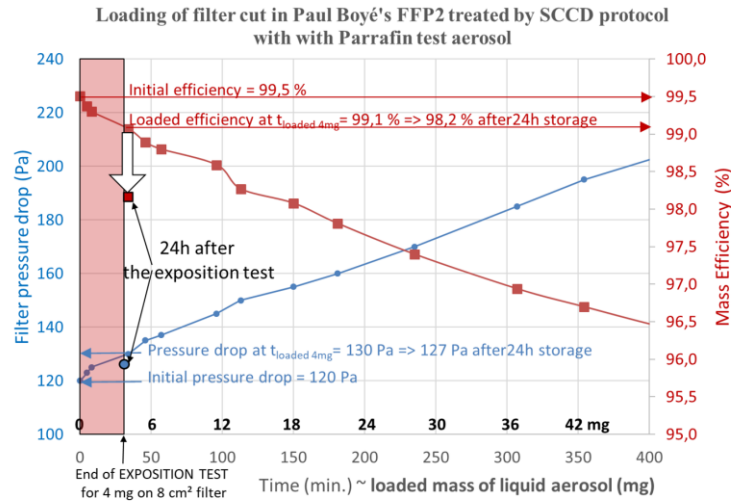


Figure SF2- 4: Filtration performances during the exposition test of filter cut in Paul Boyé’s FFP2 treated by SCCD protocol, with lined symbols for measurements performed after 24h storage at the end of the exposition test, for the storage test.

Evolution of pressure drop and mass efficiency upon loading with 4 mg of liquid Paraffin droplets (as required for a 8 cm² filter corresponding to 120 mg on the 204 cm² of the FFP2, according to EN 149 and EN 13274-7).

The loading of filters during the exposition test does not change the certification as FFP2, neither during the exposition test (even with few tens of milligrams, cf. Figure SF2- 4), nor 24 hours after the loading, for the storage test. In other words, these filters cut in Paul Boyé’s FFP2 filters successfully passed the exposition and the storage tests for both new and treated by SCCD protocol proposed here for the reuse of FFP2. As a result, these FFP2 would probably deserve to be considered for certification as resistant to oil and reusable.

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