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### VALIDATION OF KRYPTON AS A NEW TRACER GAS FOR THE STANDARDIZATION TESTS OF COLLECTIVE AND INDIVIDUAL PROTECTION SYSTEMS

### Elise Cabaset<sup>1</sup>, Mathieu Marchal<sup>1</sup>, Audrey Santandrea<sup>1</sup>, Olivier Herbinet<sup>2</sup>, Emmanuel Belut<sup>1</sup>, Stéphanie Marsteau<sup>1</sup>, Jean-Pierre Leclerc<sup>1</sup>

<sup>1</sup> INRS, Département d'Ingénierie des Procédés, 1 rue du Morvan, CS 60027 54519 Vandoeuvre les Nancy Cedex
<sup>2</sup> Université de Lorraine, CNRS, UMR 7274 - LRGP - Laboratoire Réactions et Génie des Procédés, F-54000 Nancy, France

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### ABSTRACT

Sulfur hexafluoride (SF<sub>6</sub>) is the reference tracer gas in many international standards for characterizing respiratory protective devices, fume cupboards and other devices. However, due to its significant impact on global warming, its use is becoming increasingly restrictive. Krypton was selected based on theoretical criteria for the properties that a substitute gas should possess. While compliance with these criteria is generally sufficient to guarantee the reliability of the choice, it is essential in the case of widespread use such as a standard to validate experimentally that this tracer has the same behavior as SF<sub>6</sub>. In this regard, numerous tests have been carried out to characterize the face leakage of respiratory protective devices and the rupture of containment of fume cupboards test under different operating conditions. The results obtained are identical with both tracers, and lead us to propose the use of krypton as the new reference gas in standards for which SF<sub>6</sub> was used.

Keywords: tracer, sulfur hexafluoride, krypton, fume cupboard, respiratory protective device, standardization

## **INTRODUCTION**

The use of a tracer to obtain information on the flow pattern or flow defects (short circuits, stagnant volume, leaks...) is widespread in many fields. It was first proposed by Danckwerts (1953) to estimate the residence time distribution in reactors. Since then, numerous methods have been developed to study the flow of liquid, gaseous or solid phases and for applications as varied as chemical processes, water treatment, hydrogeology, oil fields, ventilation, etc. Depending on the applied field, very precise restrictive hypotheses on the choice of the tracer as well as on the operating conditions of the tracing must be respected to obtain reliable information.

If the use of tracers is widely developed in the world of research and industry to characterize flow behavior of processes in order to model the studied systems, many uses can also be found in the context of verification or validation of the efficiency of reactors, processes or systems involving the flow of a fluid. The rules for using a tracer are then rigorously detailed in standards.

By definition, a standard is an official document released by standardization committees in order to establish rules, guidelines or definitions. Standards are developed by combining knowledge of manufacturers, consumers and regulators of a product, material, service or process. Although constraining, standards are a powerful tool to combine efficiency, quality and safety. Among all the existing standards, sulfur hexafluoride (SF<sub>6</sub>) is often used as a gas tracer to detect leaks in various fields. Indeed, in addition to being chemically inert and non-toxic (Lester and Greenberg 1950), its low and constant ambient concentration make it an interesting tracer gas. Sulfur hexafluoride is also easily measurable thanks to the high absorption of infrared light, even in low concentrations (Clemons et al. 1968). Examples of applications and the associated standards are presented in Table 1.

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Applications	Standards		
Ventilation and indoor air quality	EN 12599 (2012); ISO 16000-8 (2012)		
Biotechnology	EN 13311-3 (2001)		
Transport of radioactive material	EN ISO 12807 (2021)		
Gas infrastructures	EN 12327 (2012)		
Protective clothing	ISO 17491-2 (2012); EN 941-1+A1 (2019)		
Respiratory protective devices	EN 136 (1998); EN 140 (1998); EN 13274-1 (2001)		
	ISO 16900-1 (2019)		
Fume cupboards	EN 14175-3 (2019)		

Table 1 - Examples of applications of sulfur hexafluoride in European and internationalstandards

However, sulfur hexafluoride also has a non-negligible drawback: it is a powerful greenhouse gas. To compare and evaluate the impact of different gases on the global warming, the Global Warming Potential was developed, using the global warming impact of carbon dioxide as a reference. Sulfur hexafluoride has a particularly high GWP, reaching 23 500 over 100 years (Myhre et al. 2013), inducing that one ton of this gas will absorb 23 500 times more energy than one ton of carbon dioxide. Moreover, its long atmospheric residence time of 3200 years (Ravishankara et al. 1993)

implies that almost all of the gas released will accumulate in the atmosphere, thus continuously increasing its impact on global warming. Due to these characteristics, sulfur hexafluoride is regulated by the Kyoto Protocol aiming at reducing greenhouse gases emissions. Finding an alternative to sulfur hexafluoride as a tracer gas is therefore necessary. Several tracer have been proposed in the few last years but all of them have drawback that limit their uses to some specific applications. A gas tracer for standard should be adapted in all the configurations included in the given standard.

Alkanes such as decane-D22 have been proposed as a tracer gas for indoor air quality (Paralovo et al. 2022). However, this compound is in a liquid state in ambient conditions, and its vapors are flammable, making it unsuitable. Due to the wide range of applications to cover, including tests performed on human subjects, safety and toxicity considerations discredit some tracer gases studied for laboratory fume hoods, such as nitrous oxide (Burke et al. 2014) or isopropanol (IPA) (Bittner and Moirandat 2022). Other potential substitutes have a significant presence in the atmosphere or a variable ambient background such as carbon dioxide, widely used for assessing natural airflows (Remion et al. 2019), or helium, used for characterizing ventilation and capture efficiency (Niemala et al. 1991). This paper focuses on the replacement of sulfur hexafluoride by krypton for the characterization of collective and individual protection systems through two applications: respiratory protective devices (RPD) and fume cupboards.

The overall performances of a RPD generally rely on the filter(s) efficiency, but also on the tightness between the face and the face seal of the respirator (Grinshpun et al. 2009). If the latter point strongly depends on the adequacy between the face of the wearer and the respirator model and size, manufacturers are required to ensure that the respirators can fit. Thus, the inward leakage, i.e. the leakage at the face seal, must not exceed 0.05% for full-face masks (EN 136 1998) and 2% for quarter and half-masks (EN 140 1998). International standard ISO 16900-1 (2019) and European standard EN 13274-1 (2001) specify the method and equipment to use for the inward leakage measurement on human subjects. A test subject equipped with the RPD to test performs a serie of exercises in an atmosphere containing a known concentration of a test substance: the test agents mentioned in standards are sodium chloride, corn oil, and sulfur hexafluoride. The first part of this work then aims at comparing the inward leakage of a full-face respirator determined based on ISO 16900-1 (2019) and EN 140 (1998) using sulfur hexafluoride and krypton as test agents.

The performance of laboratory fume hoods is assessed by evaluating its fume containment. The ability of a laboratory hood to provide protection for the user at the face of the hood is strongly influenced by its environment but there is a need for a test method that can be used to evaluate the performance of a laboratory hood in the ideal environment and in the field to establish an "as used" performance rating. ANSI/ASHRAE Standard 110-2016 (2016) proposed a three parts method to do such an evaluation: flow visualization, face velocity measurement and tracer gas containment. The tracer gas containment test consists in injecting a tracer in various positions within the fume hood enclosure while measuring the concentration of the tracer in the breathing zone of a mannequin in front of the fume cupboard. The effect of the passage of a mobile obstacle on a potential rupture of containment is also assessed. The European standardization approach is somehow similar with EN 14175-3 (2019) containment and robustness of containment tests. The principle of the containment measurement consists of generating a tracer gas at a known flow rate at specified locations in the working volume of the fume cupboard and sampling the atmosphere at specified locations in the plane of the opening. The measurement in the plane of the opening makes

it possible to quantify, with precision, the leakage of chemical substances from the interior of the working volume of a fume cupboard towards its installation room. A rupture of containment test is also proposed through the robustness of containment test described. To simulate the disturbance caused by a person walking in front of the fume cupboard or by air disturbances, a rectangular plate is moved in front of the fume cupboard front at a speed of  $1 \text{ m.s}^{-1}$ . At the same time, a tracer gas is generated in the working volume of the fume cupboard. The robustness of the containment is evaluated by measuring the leakage of tracer gas following the back and forth movement of the plate by taking samples of the atmosphere 5 cm upstream of the front of the fume cupboard.

The first section of this study consists in verifying the pertinence of using krypton by checking the different criteria defining a relevant tracer gas. Then, the present article divides in two tests for the validation of krypton as a substitute to sulfur hexafluoride for standards. The first test was conducted on a bench to measure the inward leakage of a respirator equipped on a headform. The second test consists in assessing the rupture of containment of fume cupboards during a robustness test.

#### Theoretical validation of the tracer criteria

In order to replace the sulfur hexafluoride during characterization of various individual and collective protection systems, one needs to ensure that the chosen gas is a relevant tracer gas. Depending on the objectives of the tracer experiments, selected gas must respect some criteria. For the present applications, the selected gas must then meet the following criteria (Sherman 1990):

1) The behavior of the tracer gas must be similar to that of the fluid it characterizes.

A gas tracer gas should ideally have the same density and diffusion properties than the studied gas, air in our case (Hitchin and Wilson 1967), which is difficult to achieve in practice. The density of sulfur hexafluoride compared to air is 5.14 while that of krypton is 3.12, making it a more interesting tracer. Moreover, its diffusion coefficient in air is of  $14.5 \ 10^{-6} \ m^2.s^{-1}$ , which is closer to that of air than sulfur hexafluoride and other well-known tracers such as helium or nitrous oxide. The density and diffusion coefficient in air of the common tracers are presented in Table 2 and are extracted from Niemala et al. (1991).

Tuble 2 - Characteristics of alferent common tracer gases (from Niemata et al. (1991))				
Gas	Density compared to air (at STP	Diffusion coefficient in air $(m^2.s^{-1})$ at		
	conditions)	20°C		
$SF_6$	5.14	8.2 10 <sup>-6</sup>		
He	0.14	59.7 10 <sup>-6</sup>		
$N_2O$	1.53	13.2 10-6		
Air	/	17.5 10-6		
Kr	3.12	14.5 10 <sup>-6</sup>		

Table 2 - Characteristics	of different common tr	racer pases (from	Niemala et al. (1991))
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2) The tracer gas must be easily measurable and quantifiable, and be easily recognized from all other constituents of air.

Krypton can be measured using a mass spectrometer. Interferences between indoor pollutants measurements and krypton could be induced by the presence in sufficient quantities of molecules having a molecular mass close to that of krypton, i.e. 84 g.mol<sup>-1</sup>. As volatile organic compounds (VOCs) are very common gaseous contaminants in interior air quality studies, a particular attention should be paid to interferences due to this group of substances. A review of the spectrum from  $C_1$  to  $C_{12}$  and their fragments analysis conducts to four potential interferences: cyclopentanol ( $C_5H_{10}O$ ), nonanal and 2-nonanone ( $C_9H_{18}O$ ) and pentanoic acid ( $C_5H_{10}O_2$ ). However, these four pollutants are liquid in ambient conditions. Moreover, interferences can be avoided by good laboratory practice such as background noise subtraction and apparatus calibration.

3) The tracer gas should have negligible presence in typical indoor environments, particularly having no significant indoor sources.

Contrary to other potential tracer gases such as argon, krypton has the advantage of being of low abundance in air, very stable and in a gaseous state under ambient conditions. The atmospheric concentration of krypton is  $1.14 \text{ ppm}_{v}$ .

4) The tracer gas must be available and affordable considering the amount needed for running complete tests.

Since krypton and sulfur hexafluoride have similar detection levels, the gas quantity required for a test is the same for both tracers. Thus, the cost of a test directly depends on the price of the gas. At the time this work was carried out, the price of Kr was 2 times that of  $SF_6$ . However, the price difference between the two gases is narrowing considerably.

5) The tracer gas must have no significant effect on health and the environment.

As normative tests are often carried out on human subjects, e.g., for RPD certification, the tracer gas must not present any danger to health. Krypton is harmless to health, the only danger common to all gases coming from anoxia at high concentrations. Krypton is not dangerous for the environment either, and is non-flammable and non-explosive, which makes it easy for transport, handling and storage.

6) The tracer gas should not affect or be affected by physical or chemical parameters of the test environment.

This last criterion cannot be evaluated completely via literature review, as it directly depends on environment conditions. Still, krypton is an inert gas and is not affected by temperature or relative humidity in classical laboratory conditions. Moreover, laboratory measurements of krypton concentration stability over a long period were performed in a 1 m<sup>3</sup> sealed enclosure containing various materials and objects. No decrease of concentration was identified, which suggests that no sorption on surfaces commonly found in indoor spaces occurred with this gas.

## **RESPIRATORY PROTECTIVE DEVICES**

### Methods

#### Experimental apparatus

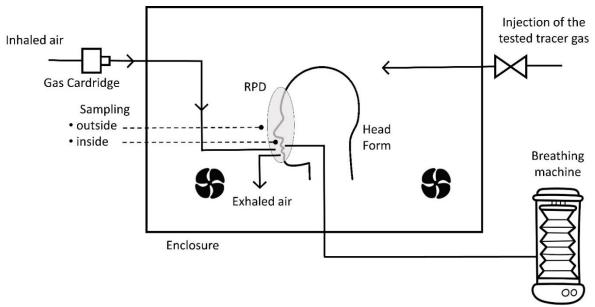


Figure 1 - Experimental set-up for the measurement of inward leakage of respirators

As previously mentioned, the performances of a respirator vary with both the filter efficiency and the inward leakage. However, the latter point directly depends on the respirator's fit on the wearer's face (Clayton and Vaughan 2005). To avoid the consideration of individual characteristics (facial dimensions, energetic metabolism, experience, etc.), the tests were carried out on a headform connected to a breathing machine, in a 1m<sup>3</sup> enclosure. The experimental set-up, based on ISO 16900-1 (2019) for inward leakage measurement is presented in Figure 1.

The chosen headform (i-bodi) corresponds to the large size mentioned in ISO 16900-5 (2016) and based on anthropometric measurements from ISO 16976-2 (2015). The dummy head was connected to the breathing machine simulating sinusoidal breathing cycles to represent human activities. The three breathing rates used during the experiments are described in Table 3 and correspond to light, moderate and heavy work rates according to EN ISO 8996 (2021).

Tuble 5 - Chu	Tuble 5 - Characteristics of the simulated breathing rates						
Breathing	Work rate	Tidal	Frequency	Mean ventilated	Peak inhalation		
rate	work rate	volume (L)	(cycle.min <sup>-1</sup> )	flow (L.min <sup>-1</sup> )	flow (L.min <sup>-1</sup> )		
BR1	Light	1.5	9	13.5	41		
BR2	Moderate	2.1	13	27.3	84		
BR3	Heavy	2.1	21	44.1	141		

Table 3 - Characteristics of the simulated breathing rates

Three models of single cartridge full-face respirators were tested to include different manufacturers, and thus different materials. All respirators are full-face air-purifying respirators certified according to EN 136 (1998) and commercially available. To characterize the respirator fit on the headform, the evaluation of a manikin fit factor was required. Between the two methods mentioned by ISO 16975-3 (2018) to perform a fit test, the Controlled Negative Pressure (CNP) method was adopted. Indeed, this method relies on the measurement of a leak rate, which seems more appropriate than particle counting to study respiratory protection against gases. The manikin fit factor was then determined using a Quantifit (OHD) device with the Redon protocol, recommended by the manufacturer when focusing on full-face masks and meeting Occupational Safety and Health Administration (OSHA) requirements (OSHA 2004). The measurement consists in creating a depression inside the mask and assimilating the amount of air to evacuate to maintain the depression to the leak rate. The fit factor is then calculated as the ratio between the modeled breathing rate, directly defined by the protocol, and the leak rate. The minimum recommended fit factor to consider a successful test and a good fit is of 500 for a full-face respirator, i.e. a leak rate of 100 mL.min<sup>-1</sup>. The very principle of the inward leakage measurement being to evaluate the fit between the respirator and the face of the wearers, several fits were tested to represent various scenarios. Thus, experiments were performed with manikin fit factors of 20, 300, 500 and 3000 to reflect respectively very bad, medium, good and very good fits. These values were notably achieved by redonning and adjusting the respirator several times until reaching the desired manikin fit factor value.

A flexible tube allowed inhaling outside the test chamber to avoid the contamination of the inhaled air by the tracer, while the exhaled air was rejected inside the chamber. Sulfur hexafluoride and krypton were first injected in the closed vessel using a syringe to obtain an ambient concentration between 1000 and 1400 ppmv for each gas. In order to compensate the dilution caused by the introduction of air from outside the enclosure, both gases were permanently injected to maintain the ambient concentration. The flow rate was determined and set according to the tested breathing rate. Two fans were disposed in the chamber to ensure the homogeneity of the atmosphere.

An Omnistar mass spectrometer (model GSD320 O2, Pfeiffer) was used to quantify the two tracer gases. Samples with a capillary tube were taken inside and outside the respirator, in a radius of 30 cm around the mouth (EN 1540 2021). To allow sampling inside the respirator, the capillary tube was introduced between the headform and the face seal. The induced leak was plugged with vaseline. It should be noted that the fit factor measurement was performed after this procedure in order to qualify the fit during the actual test conditions. Concerning krypton measurement, the most abundant stable isotope <sup>84</sup>Kr is monitored by the spectrometer. Sulfur hexafluoride fragments quite easily, inducing that only the SF5+ fragment was measured by the apparatus. In order to convert the intensity of the ionic currents determined by the spectrometer into a concentration value, a calibration of the instrument with both gases was performed and showed great linearity. Preliminary tests performed with each gas separately and with both gases at the same time led to the same inward leakage values. Thus, sulfur hexafluoride and krypton were injected simultaneously during the following experiments, ensuring that the tests were performed in the exact same conditions.

#### Experimental protocol

At the beginning of the test, the headform equipped with the respirator was connected to the breathing machine. The capillary for inner concentration measurement was inserted into the half mask part of the full-face respirator. The manikin fit factor was then measured and the respirator was redonned until reaching the desired value. Once set, five measurements were performed at the beginning and at the end of the test, and the global value was calculated as the harmonic mean of the ten values. It should be noted that the test results were saved only if the respirator fit did not significantly vary during the experiment. The breathing machine was then started and an initial measurement of the concentration inside and outside the RPD is performed to determine the blank. When the ambient concentration  $C_{0,i}$  was measured during at least 5 minutes, followed by the concentration inside the RPD C<sub>i</sub> during at least 10 minutes, before evaluating again the ambient concentration at least 5 minutes. The global ambient concentration was calculated as follows for each gas:

Inward leakage (%) = 
$$\frac{2 C_i}{C_{0,i} + C_{0,f}} \times 100$$
 (1)

Where:

 $C_{0,i}$  = initial ambient concentration in given gas  $C_{0,f}$  = final ambient concentration in given gas  $C_i$  = concentration in the RPD in given gas.

For each respirator, manikin fit factor and breathing rate, the sampling cycle was repeated three times. The global inward leakage was then calculated as the mean value of the three tests and is represented with a 95% confidence interval.

#### **Results and discussion**

The inward leakage was evaluated for each respirator for a fit factor of 500, i.e. the limit value to consider a good fit, and for the three breathing rates. It appears in Figure 2 that the inward leakage values vary between 0.05% and 1.9%. This can seem surprising considering that the respirators are certified according to EN 136 (1998), implying that the inward leakage must be lower than 0.05% when performing different exercises. This can notably be explained by the fact that the respirators were used in previous studies and were donned multiple times, resulting in components stress and impairing the fit. This phenomenon was notably highlighted and discussed by Bergman et al. (2012) for filtering facepiece respirators. Besides, respirators are often tested using an aerosol agent, which tends to lead to lower inward leakage values than with gases (Myers et al. 1990). Moreover, the scattering of the values is due to the fact that, for each respirator, the inward leakage was evaluated for three breathing rate, which impacts the respirator performances (He et al. 2013). For each configuration, Figure 2 then illustrates that similar inward leakage values were obtained with both tracer gases within the margin of uncertainty, regardless of the respirator model and breathing rate. A linear regression can be drawn between the values measured with both tracers, with a coefficient of determination of 0.996, thus showing good consistency between SF<sub>6</sub> and Kr.

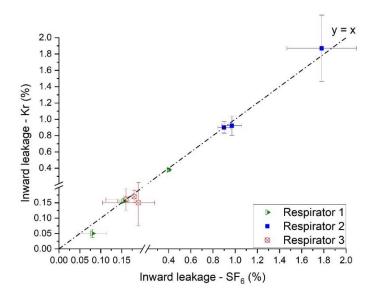


Figure 2 - Comparison between the inward leakage values measured with SF<sub>6</sub> and Kr on three respirators for three breathing rates with a good fit (fit factor of 500)

Since the very principle of inward leakage measurement is to evaluate the leak rate between the facepiece and the face, several fits were tested on respirator 1. Thus, very bad, medium, good and very good fits were achieved, corresponding to fit factors of 20, 300, 500 and 3000 respectively. It then appears on Figure 3 that when increasing the fit factor, the inward leakage logically decreases, i.e. the respirator performances increase (Santandrea and Chazelet 2023). For each of the different configurations, the same inward leakage values were determined with Kr and  $SF_6$  regardless of the respirator fit.

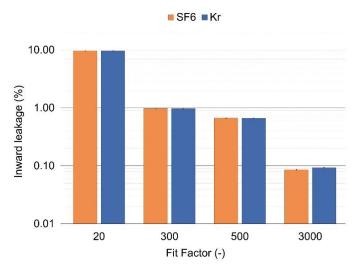


Figure 3 - Evolution of the inward leakage with the fit factor derived from SF<sub>6</sub> and Kr tracer measurements for respirator 1 and moderate breathing rate (BR2)

Experimental tests were then performed on three different models of respirators, three breathing rate (from light to heavy work rate) and four different fits, and showed good consistency between

both tracer gases. In terms of leakage measurement, krypton then appears as a good alternative to sulfur hexafluoride.

### FUME CUPBOARD: RUPTURE OF CONTAINMENT

 $SF_6$  tracing is also commonly used to characterize the efficiency of fume cupboards, and more specifically any containment failures. In order to compare sulfur hexafluoride and krypton as tracers, tests were carried out under transient conditions, artificially creating disturbances similar to those obtained when a person walks past the fume cupboard. The global set-up was designed and validated by Belut et al. (2023). This protocol has the advantage of approaching the european standardized robustness test of fume cupboards described in EN 14175-3 (2019).

#### Methods

#### Experimental apparatus

As a preliminary precaution, in order to avoid external perturbation of the flow, the experimental set-up was placed in a well-controlled test room as illustrated in Figure 4, with dimensions of 6,7  $\times 4.5 \text{ m} \times 3.5 \text{ m}^3$  (length x width x height). This bench was composed of an open cavity that mimics a simplified fume cupboard, a mobile object moving in front of it, a diffusing duct for air compensation, a sampling and analysis system including the mass spectrometer (placed outside the test room) and the injection set-up of the two tracers, sulfur hexafluoride and krypton. The mass spectrometer used in this part is the one used for respirator inward leakage measurement and presented in the previous part of the article. Turbulence and interference from external sources, such as air supply diffusers, were controlled. Thermal convection was managed by low velocity and accurate control of temperature. Doors were blocked and traffic in the room was forbidden during the tests to avoid flow disturbances. Two flows, one of 0.11 m<sup>3</sup>.s<sup>-1</sup> through the tested fume hood and the other of 0.14 m<sup>3</sup>.s<sup>-1</sup> through a ventilation outlet, are extracted from the test room. Meanwhile, a fresh air supply is provided at a flow rate of 0.25 m<sup>3</sup>.s<sup>-1</sup> by a diffusing duct injecting the air at low velocities to avoid disturbances. Local temperatures are monitored using 10 thermocouples distributed over the height to adjust the temperature of the cabin to that of the test hall to ensure the absence of natural convection.

The simplified fume cupboard, presented in Figure 5, was a rectangular enclosure with interior dimensions of 0.8 x 0.34 x 0,4 m<sup>3</sup> (width x height x depth). The ceiling box was connected to a blower through an exhaust rectangular pipe. The airflow velocity through the opening plane of the enclosure was maintained to 0.4 m.s<sup>-1</sup>. The front face of the enclosure was kept wide open. A homogeneous injection of 99.6 NL.min<sup>-1</sup> of air and 0.215 NL.min<sup>-1</sup> of each of the tracer gases was performed through a perforated plate (0,76 x 0.165 m<sup>2</sup>) installed inside the fume cupboard, near the front edge of the surface. Sampling points were located on the opening surface of the enclosure, as illustrated in Figure 5, to allow gas concentration measurement using the mass spectrometer. The rupture of the containment of the fume cupboard is ensured by the passage of a mobile object with dimensions representative of a human (a 1.65 x 0.4 x 0.2 m<sup>3</sup> parallelepiped). The translation of the object, which is positioned at a distance of 0.4 m from the open surface of the enclosure, is

carried out over 2.66 m parallel to the opening with a maximum speed of 1 m.s<sup>-1</sup>.

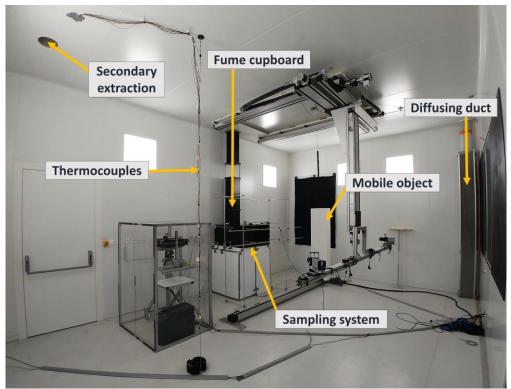


Figure 4 - Complete set-up for the characterization of fume cupboard efficiency

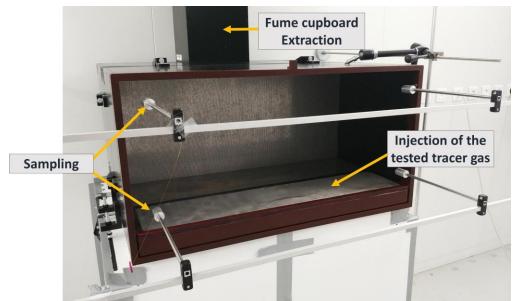


Figure 5 - Injection and sampling locations of tracer for the characterization of fume cupboard efficiency

#### Experimental protocol

At the beginning of the test, the ventilation and temperature controls of the test chamber were switched on, and thermal equilibrium was usually reached after 2 hours. After performing a oneminute blank measurement, the tracer gases were generated. After 5 minutes, the moving object passed by the fume cupboard, and the generation was stopped 2 minutes later. The moving object then returned to its starting position and 35 minutes are waited for the airflow to stabilize in the cabin before performing another test. A total of 10 tests was carried out. The interest of this experiment for tracer testing is that a transient curve is obtained, with enough strength to test the robustness of both tracer responses (SF<sub>6</sub> and Kr) comparison.

Concerning the data analysis of the mass spectrometer measurements, the same calibration curves than for the previous part were used. A special attention was paid to the time lapse of the recordings to synchronize  $SF_6$  and Kr values. Additional treatment of the data was performed in order to compare the tracers released during the rupture of containment. For a continuous source, the ratio of the concentration of tracer ( $SF_6$  or Kr) to the flow of tracer injection (C/Q) is usually calculated to quantify the dispersion of the contaminant outside of the fume cupboard.

#### **Results and discussion**

A typical example of the ratio between the concentration of tracer (noted C) and the flow of tracer injection (noted Q) during the rupture of containment of the fume cupboard is presented in Figure 6. For each test, an increase in the tracer concentration is observed in the opening plane of the ventilated chamber a few seconds after the passage of the mobile object. This concentration then decreases until reaching the initial value before the disturbance, inducing that the enclosure containment is operational again. As illustrated in Figure 6, the curves obtained with  $SF_6$  and Kr are identical, proving that both tracers provide similar response to the rupture of containment of the fume cupboard.

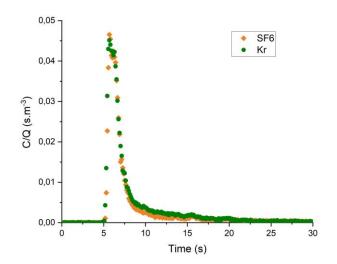


Figure 6 - Tracer responses of Kr and SF<sub>6</sub> injections during a rupture of containment of a fume cupboard

For each of the 10 experiments, from 230 to 280 values were measured, resulting in more than 2500 data. The whole set of values obtained with Kr is presented in Figure 7 against the data from SF<sub>6</sub> measurements. It appears that, for each test, similar results are obtained with both tracers, with most of the values included within a margin of  $\pm$  10%. A linear regression can be plotted between the values measured with the two gases, with a coefficient of determination of 1.04, which demonstrates the remarkable agreement between the two tracers.

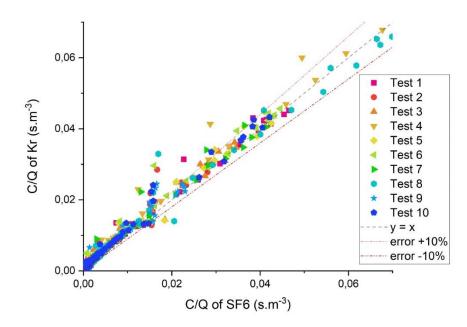


Figure 7 - Comparison between tracer dispersion of SF<sub>6</sub> and Kr during the whole set of data of rupture of containment of a fume cupboard

On a standardization approach, EN 14175-3 (2019) specifies to calculate the mean value of the tracer concentration during the rupture of containment. It is thus very important to ensure that the proposed new tracer has the same behavior during the entire process.

#### CONCLUSION

Sulfur hexafluoride has been used for years as a tracer to characterize the flow or to highlight defects (leaks, short circuit...) in many sectors of activity. It has become a reference tracer gas in many standards. However, due to its strong greenhouse effect, its use was gradually reduced and is probably going to be completely prohibited in the near future. The search for a substitute tracer gas requires meeting many criteria, including having a similar behavior than the former tracer. Tracer experiments are widely used in the field of chemical engineering to characterize the flow behavior of complex process. It is well known from chemical engineers that the tracer and both the injection and detection systems have to be carefully chosen to obtain reliable information about the studied system. Although the validation of the tracer could be done just based on the criteria in order to guarantee the reliability of the standards, comparative tests between  $SF_6$  and a new selected tracer, Kr, have been done to characterize the inward leakage of respiratory protective devices and

the rupture of containment of fume cupboards under different operating conditions. For both applications, the obtained results are similar, proving that Kr is a good candidate to replace  $SF_6$  for standardization tests of collective and individual protection systems. The injection procedure for krypton is similar to that of  $SF_6$  in standards, and this gas can be easily measured by a mass spectrometer. The cost of the tests are of the main order for the two tracers. These results have been presented at standard commission in order to adapt the test procedure to the  $SF_6$  restrictions and use Kr for standardized tests.

### REFERENCES

ANSI/ASHRAE Standard 110-2016. 2016. Methods of Testing Performance of Laboratory Fume Hoods.

Belut E, Lechêne S, Trouette B, Vincent S, Halim Atallah G. 2023. Flow and contaminant transfer at the interface of an aerodynamic containment barrier subjected to the wake of a moving obstacle. Build Environ. 241:110465. https://doi.org/10.1016/j.buildenv.2023.110465

Bergman MS, Viscusi DJ, Zhuang Z, Palmiero AJ, Powell JB, Shaffer RE. 2012. Impact of multiple consecutive donnings on filtering facepiece respirator fit. Am J Infect Control. 40(4):375–380. https://doi.org/10.1016/j.ajic.2011.05.003

Bittner M, Moirandat C. 2022. Rapid Robot-Aided On-site Testing of Fume Cupboards. Ann Work Expo Health. 66(8):1086–1090. https://doi.org/10.1093/annweh/wxac043

Burke M, Wong L, Gonzales BA, Knutson G. 2014. Nitrous Oxide as a Tracer Gas in the ASHRAE110-1995Standard.JOccupEnvironHyg.11(1):32–39.https://doi.org/10.1080/15459624.2013.839881

Clayton M, Vaughan N. 2005. Fit for purpose? The role of fit testing in respiratory protection. Ann Occup Hyg. 49(7):545–548. https://doi.org/10.1093/annhyg/mei046

Clemons CA, Coleman AI, Saltzman BE. 1968. Concentration and ultrasensitive chromatographic determination of sulfur hexafluoride for application to meteorological tracing. Environ Sci Technol. 2(7):551–556. https://doi.org/10.1021/es60019a005

Danckwerts PV. 1953. Continuous flow systems. Chem Eng Sci. 2(1):1–13. https://doi.org/10.1016/0009-2509(53)80001-1

EN 136. 1998. Respiratory protective devices Full face masks Requirements, testing, marking.

EN 140. 1998. Respiratory protective devices - Half masks and quarter masks - Requirements, testing, marking.

EN 941-1+A1. 2019. Protective clothing against dangerous solid, liquid and gaseous chemicals, including liquid and solid aerosols — Part 1: Performance requirements for Type 1 (gas-tight) chemical protective suits.

EN 1540. 2021. Workplace exposure - Terminology.

EN 12327. 2012. Gas infrastructure — Pressure testing, commissioning and decommissioning procedures — Functional requirements.

EN 12599. 2012. Ventilation for buildings — Test procedures and measurement methods to hand over air conditioning and ventilation systems.

EN 13274-1. 2001. Respiratory protective devices - Methods of test - Part 1: determination of inward leakage and total inward leakage.

EN 13311-3. 2001. Biotechnology — Performance criteria for vessels — Part 3: Glass pressure vessels.

EN 14175-3. 2019. Fume cupboards — Part 3: Type test methods.

EN ISO 8996. 2021. Ergonomics of the thermal environment - Determination of metabolic rate.

EN ISO 12807. 2021. Safe transport of radioactive materials — Leakage testing on packages.

Grinshpun SA, Haruta H, Eninger RM, Reponen T, McKay RT, Lee S-A. 2009. Performance of an N95 Filtering Facepiece Particulate Respirator and a Surgical Mask During Human Breathing: Two Pathways for Particle Penetration. J Occup Environ Hyg. 6(10):593–603. https://doi.org/10.1080/15459620903120086

He X, Grinshpun SA, Reponen T, McKay R, Bergman MS, Zhuang Z. 2013. Effects of Breathing Frequency and Flow Rate on the Total Inward Leakage of an Elastomeric Half-Mask Donned on an Advanced Manikin Headform. Ann Occup Hyg. 58(2):182–194. https://doi.org/10.1093/annhyg/met053

Hitchin ER, Wilson CB. 1967. A review of experimental techniques for the investigation of natural ventilation in buildings. Build Sci. 2(1):59–82. https://doi.org/10.1016/0007-3628(67)90007-2

ISO 16000-8. 2012. Indoor air - Part 8 : Determination of local mean ages of air in buildings for characterizing ventilation conditions.

ISO 16900-1. 2019. Respiratory protective devices - Methods of test and test equipment - Part 1 : determination of inward leakage.

ISO 16900-5. 2016. Respiratory protective devices - Methods of test and test equipment - Part 5: Breathing machine, metabolic simulator, RPD headforms and torso, tools and verification tools.

ISO 16975-3. 2018. Respiratory protective devices - Selection, use and maintenance - Part 3: Fit-testing procedures.

ISO 16976-2. 2015. Respiratory protective devices - Human factors - Part 2: Anthropometrics.

ISO 17491-2. 2012. Protective clothing — Test methods for clothing providing protection against chemicals — Part 2: Determination of resistance to inward leakage of aerosols and gases (inward leakage test).

Lester D, Greenberg LA. 1950. The toxicity of sulfur hexafluoride. Arch Ind Hyg Occup Med. 2(3):348–9.

Myers WR, Kim H, Kadrichu N. 1990. Effect of particle size on respirator faceseal leakage. [place unknown]: West Virginia University, Department of Industrial Engineering.

Myhre G, Shindell D, Bréon FW, Collins W, Fuglestvedt J, Huang J, Koch D, Lamarque J-F, Lee D, Mendoza B. 2013. Anthropogenic and Natural Radiative Forcing. In: Clim Change 2013 – Phys Sci Basis [Internet]. 1st ed. [place unknown]: Cambridge University Press; [accessed 2023 Jul 10]; p. 659–740. https://doi.org/10.1017/CBO9781107415324.018

Niemala R, Lefevre A, Muller JP, Aubertin G. 1991. Comparison of three tracer gases for determining ventilation effectiveness and capture efficiency. Ann Occup Hyg. 35(4):405–417. https://doi.org/10.1093/annhyg/35.4.405

OSHA. 2004. Controlled Negative Pressure REDON Fit Testing Protocol. Washington, DC:

Occupational Safety and Health Administration.

Paralovo SL, Stranger M, Lazarov B, Spruyt M, Laverge J. 2022. Adapted tracer gas test for passive measurement of total air change rates using alternative tracer substance. Sci Technol Built Environ. 28(7):792–806. https://doi.org/10.1080/23744731.2021.1997061

Ravishankara AR, Solomon S, Turnipseed AA, Warren RF. 1993. Atmospheric Lifetimes of Long-Lived Halogenated Species. Science. 259(5092):194–199.

Remion G, Moujalled B, El Mankibi M. 2019. Review of tracer gas-based methods for the characterization of natural ventilation performance: Comparative analysis of their accuracy. Build Environ. 160:106180. https://doi.org/10.1016/j.buildenv.2019.106180

Santandrea A, Chazelet S. 2023. Respiratory protective device: one size to fit them all? J Occup Environ Hyg.:1–24. https://doi.org/10.1080/15459624.2023.2205466

Sherman MH. 1990. Tracer-gas techniques for measuring ventilation in a single zone. Build Environ. 25(4):365–374. https://doi.org/10.1016/0360-1323(90)90010-O