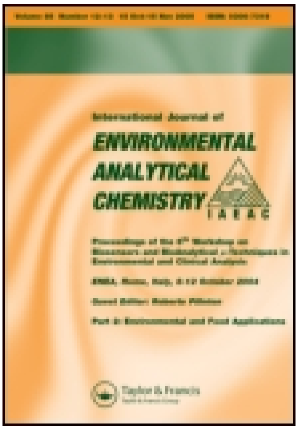

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The effect of the wind velocity on the uptake rates of various diffusive samplers

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This paper examines the results of experiments carried out in an exposure chamber to determine the wind effects on the performance of various diffusive sampler types commonly used for measuring gaseous pollutants in air. The resistance to wind of six diffusive samplers, two Palmes tubes, a badge with diffusion membrane, the EMD sampler and two radial diffusive samplers for different pollutants was compared in a range of velocities from 0 to 300 $cm\,s^{-1}$. For all diffusive samplers tested, an increase in uptake rate was observed with increased air velocity usually following a logarithmic function. The consequences are an underestimation in the concentration measured by the diffusive samplers for low wind velocities below 30 $cm\,s^{-1}$ and conversely an overestimation from 60 $cm\,s^{-1}$. The magnitude of wind effects depends on diffusive sampler type and exceeds an uptake rate variation of ±20% for the axial diffusion tubes and the EMD sampler. With regard to the characteristics of each diffusive sampler, the dependence of uptake rate on wind velocity was analysed and discussed. The radial diffusive samplers for benzene and particularly the ones having a large and thick porous membrane appear to be the most effective design to minimise the influence of air velocity on passive sampling.

Keywords: passive sampler; diffusive sampler; wind velocity; exposure chamber; membrane

1. Introduction

Diffusive samplers are nowadays widely used in determining the concentration of various air pollutants like nitrogen dioxide [1–3], ozone [4–6], sulphur dioxide [7] and volatile organic compounds [8,9] with applications in the domains of indoor air, personal exposure and ambient atmospheric monitoring. In the last thirty years, efforts have been undertaken to improve the accuracy of measurements by these passive sampling methods. The main improvements were made in the means of sampling with the use of more efficient trapping media [10] and the development of new designs intended for limiting the influence of environmental factors on passive sampling [11,12]. The current way is to equip the diffusive sampler with a porous membrane in order to isolate the molecular diffusion path from air movements. Another solution to reduce the wind effects is to use shelters [13].

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However, many studies reported varying degrees of bias because of the effects of wind. Several works [14–18] suggested that wind velocity has no detectable or minor effect on the diffusive samplers. On the contrary, other studies [13,19–22] reported that wind is a major factor affecting the measurements with the same types of diffusive samplers. These results are difficult to compare because the tests were carried out in different ranges of wind velocity.

The wind can affect the mass transfer occurring by molecular diffusion in two manners. At low wind speeds, a stagnant layer of air can occur at the entrance of the sampler increasing the effective diffusion length. This layer leads to increase the transfer resistance, to decrease in uptake rate and consequently, the concentration is underestimated by the diffusive sampler.

Conversely, when the wind speed is high enough, eddies occurring at the diffusion surface can reach the inside the sampler and tend to reduce the diffusion length. The effect is an increase in uptake rate leading to an overestimation of airborne concentration.

The magnitude of these effects depends on the sampler design. Palmes and Lindenboom [23] showed that there was a strong similarity between Ohm's law of electrical current and Fick's law of diffusion. The dependence of uptake rate with wind velocity could be satisfactorily modelled by equations derived from fluid mechanics for the passive samplers having an axial diffusion path as in the Palmes tubes and badges [24,25]. On the other hand, only empiric equations that correlate the sampling rate with wind velocity are given in the literature [16,17] concerning the radial diffusion sampler recently developed by Cocheo *et al.* [11].

In this paper, the effect of wind speed on the uptake rates of six diffusive samplers used for different pollutants was investigated by an experimental approach carrying out a series of tests in an exposure chamber. The aims of this study were to compare the uptake rate of diffusive samplers at various wind velocities and to find the most effective design to minimise the influence of air velocity on passive sampling.

2. Experimental

2.1 *Description of diffusive samplers*

2.1.1 *Diffusion tubes*

The first diffusive tube tested in this study was of the axial tube type (Figure 1a) that was initially developed by Palmes for NO_2 measurements [14]. This sampler (Gradko International, Winchester, UK) consists of an acrylic tube (of 71.16 ± 0.20 mm long and 10.91 ± 0.15 mm inner diameter) open at one end and three stainless steel meshes coated with triethanolamine (TEA, Acros Organics, Noisy Le Grand, France) at the closed end. A removable cap is used to close the open end of the tube before and after exposure. NO_2 diffuses through the air in the tube and is trapped as nitrite ion on TEA. The quantity of NO_2^- ions is extracted within the sampler body with deionised water and analysed by ion chromatography. More details about the sample preparation and analytical method have been given in a previous paper [13].

Passam tube (Passam AG, Mannedorf, Switzerland), another type of diffusion tubes proposed by Hangartner [26], was also tested in these experiments. It is very similar to the Palmes tube but with slightly different dimensions of the tube (of 73.45 ± 0.05 mm long and 9.79 ± 0.20 mm inner diameter). Its uptake rate is lower than that of the Palmes tube owing to a longer length and a smaller inner diameter. NO_2 is sampled on three stainless

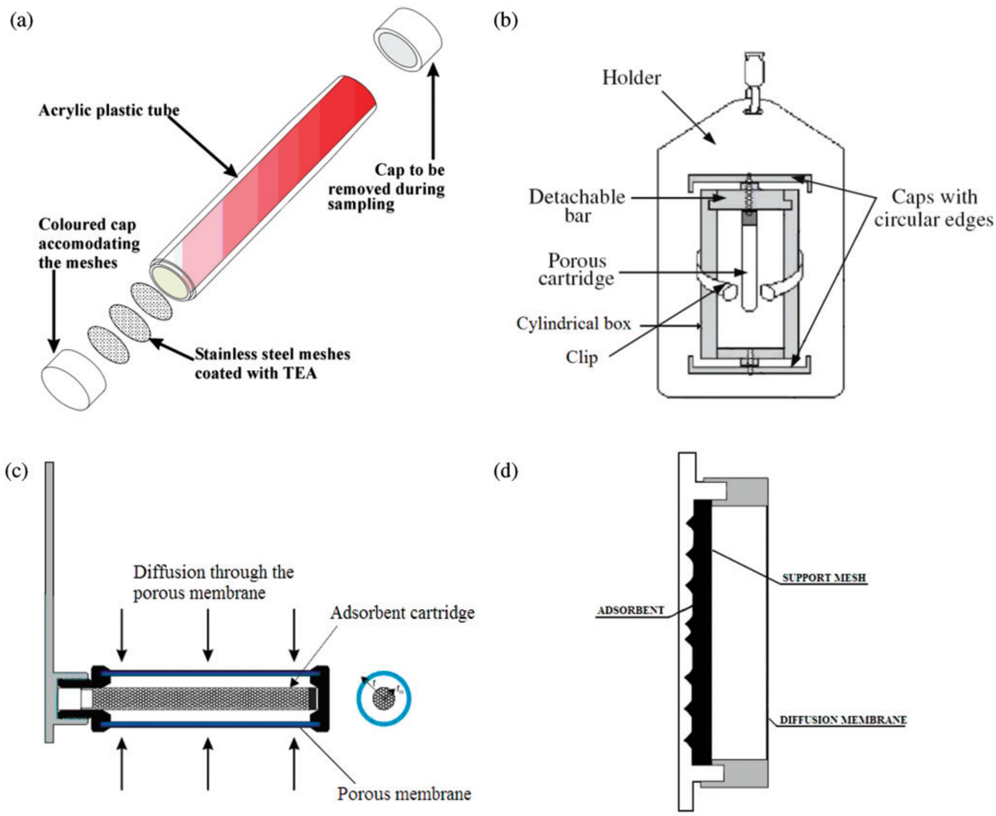

Figure 1. Description of diffusive samplers: (a) Diffusion tube; (b) EMD sampler; (c) Radiello sampler; (d) Badge with a diffusion membrane.

steel mesh discs coated with TEA placed at the closed end of the tube. The samplers were extracted and analysed with the Griess-Saltzman colorimetric method described by Atkins *et al*. [15].

2.1.2 *EMD sampler*

The EMD sampler (Ecole des Mines, Douai, France) was developed for nitrogen dioxide measurements (Figure 1b). It is composed of a porous cartridge impregnated with the TEA solution fitted in a cylindrical protective box equipped with caps at its extremities. The large sampling area (cartridge surface) and the two circular openings provide a high uptake rate to this passive sampler. The extraction was achieved in a test tube by plunging the cartridge into 5 mL of ultrapure water and vortex mixing. Ion chromatography is preferred over the commonly used spectrophotometry method on account of a better sensitivity [27–28]. Tests in an exposure chamber and in field conditions showed that the EMD sampler is suitable for very short sampling durations. Its detection limit is 11 $\mu g\, m^{-3}$ for one hour sampling. All the validation tests are shown elsewhere [28].

2.1.3 *Radial diffusive samplers*

This diffusive sampler is of the radial type (Figure 1c) and suitable for VOC measurements. Only the results on benzene are considered in this study. This passive sampler consists of a stainless steel net cartridge (60 mm long, 4.8 mm in diameter, 100 mesh hole size) filled with 350 mg of 40–60 mesh Carbograph 4 (a graphitised carbon) and inserted into a microporous polyethylene cylinder of 16 mm in diameter, 50 mm long (47 mm available for the diffusion). Two kinds of microporous membrane were tested, the one of 1.75 mm wall thickness and 20–30 μm pore size (white membrane) and the other one of 5 mm wall thickness and 10 μm pore size (yellow membrane). For exposure, the Radiello sampler is screwed on a plane cellulose acetate equilateral triangle equipped with an attaching clip. All ready-to-use radial diffusive sampler components are commercially available from Fondatione Salvatore Maugeri (Padoue, Italia). The molecules of benzene diffuse through the cylindrical membrane towards the cartridge and are adsorbed by Carbograph 4. As the diffusion path is perpendicular to the axis of the cylinder, it can be considered that diffusion occurs with radial symmetry. After sampling, the cartridge is housed into a stainless-steel tube 89 mm long × 6.3 mm OD × 5 mm ID to be analysed with a thermal desorber (TD) (Turbomatrix, Perkin-Elmer, Courtaboeuf, France) interfaced with a gas chromatograph (GC) (Thermo Trace GC 2000, Thermo Electron, Villebon sur Yvette, France) and a flame ionisation detector (FID). The analytical conditions and calibration procedure used for the analysis of these cartridges were described in three previous papers [20,29–30].

2.1.4 *Badge with a diffusion membrane*

This diffusive sampler was a protopype of the axial badge type (Figure 1d) designed by Perez-Ballesta *et al.* [24] for toluene measurements. It consists of a Teflon cover in which the adsorbent is deposited, a stainless steel wire mesh support of 0.1-mm aperture size for compacting the layer of adsorbent and a diffusion membrane of 8.04 cm^3 placed at a specified distance (7 mm) from the adsorbent layer. The diffusion membrane is a stainless steel wire mesh with regular pores of 1 μm size whose thickness is under 0.1 mm. The toluene molecules go through the membrane and diffuse in the badge following one axis perpendicular to the sampling surface. Toluene is desorbed with carbon disulfide and analysed by gas chromatography in accordance with the NIOSH method [31].

2.2 *Expression of uptake rate*

The passive sampling is controlled by the molecular diffusion of molecules along a diffusion path induced by the gradient of concentration setting up between the ambient air and air close to the sampling area where the concentration tends to zero.

An uptake rate ($cm^3\,min^{-1}$) of diffusive sampler for a compound is calculated by applying Equation (1) derived from Fick's first law:

$$UR = \frac{m}{C \times t} \times 10^6 \tag{1}$$

where C is the mean concentration of the compound in air ($\mu g\,m^{-3}$), t the sampling time (min) and m the mass sampled by the diffusive sampler (μg).

2.3 *Experiments in an exposure chamber*

The laboratory experiments were performed using two similar dynamic exposure chambers in which sets of 6 diffusive samplers were simultaneously exposed to controlled atmospheres. The concentration of contaminant, temperature, humidity and wind velocity are controlled (see Figure 2). A detailed description of these exposure devices has already appeared elsewhere [13,24]. Briefly, each test atmosphere was produced by diluting bulk gas mixtures coming from a vaporisation system for toluene and compressed gas cylinders for other compounds. The dilution air is produced by a compressor, chemically filtered in an air purifier (AZ 2020 manufactured by Claind, Wasquehal, France, purity: total hydrocarbons < 0.1 ppm) and supplies two air flows, the first one containing the target compound and the second passes through a bubbler to humidify air. These two air flows are regulated by mass flow controllers (MKS, Le Bourget, France). They are mixed at the chamber inlet, generating an air flow controlled in contaminant concentration and humidity. The open airflow can be regulated between 1 and $5\,L\,min^{-1}$. A humidity in a wide range from 0 to 90% can be achieved. The exposure chamber has a capacity of 35 L. It contains a glass plate separating the lower part, devoted to the generation of air movements by means of three axial fans regulated by a potentiometer, from the higher part where the passive samplers are exposed on the glass plate into the upper part of the chamber. The cylindric form of the chamber favours the recirculation currents of air that allowed to reach 5 to $300\,cm\,s^{-1}$. The exposure chamber is put in a thermostatic enclosure maintaining a constant temperature between 0 and 40°C. Temperature, relative humidity

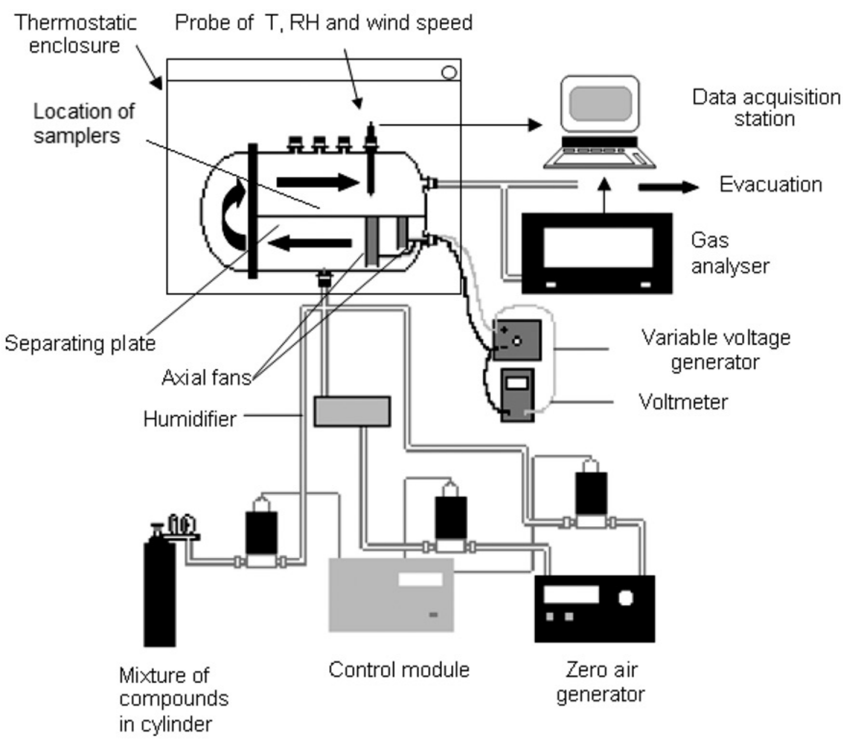

Figure 2. Scheme of the exposure chamber system.

and wind velocity are controlled and continuously recorded by Testo term multifunction probe (Ref: 0635.1045, Testo, Forbach, France) inserted through an upper entrance of the chamber. The sensor is connected to a data logger (Testo term 452, Testo, Forbach, France) for continuous monitoring of three parameters.

For the tests of two NO_2 diffusion tubes and the EMD sampler, the concentration was continuously measured in the exposure chamber by a chemiluminescent NOx analyser (Seres NOx 2000, Seres, France). This monitor is periodically checked with an NO content in N_2 cylinder, certified against the national reference standard of Laboratoire National d'Essais (gravimetric dilution method). For the tests of two Radiello samplers, a benzene monitoring was carried out with a BTEX analyser (VOC 71M, Environment SA, Poissy, France). This analyser was calibrated beforehand and every week from a cylinder certified at the laboratory by a procedure described by Badol *et al.* [32].

The tests of badge with a diffusion membrane were made by Perez-Ballesta *et al.* [24] in a dynamic exposure chamber similar to the one described previously. It has a capacity of 14.8 L and is equipped with an axial fan permitting the generation of recirculation currents which can reach 2–200 cm s^{-1}. The test atmospheres were produced by diluting bulk gas mixtures coming from a vaporisation system. Humidity and temperature were regulated by means of a thermostated gas-liquid contactor and an internal electrical resistance, respectively. The exposure chamber was connected on-line to a gas chromatograph in order to follow the toluene concentration during the experiments.

At first, the uptake rates are determined to the standard conditions defined as follows: a relative humidity of 40–50%, a temperature of 20°C and a wind velocity of 50 cm s^{-1}. Two sets of 6 diffusive samplers are exposed in the exposure chamber under these standard conditions. These uptake rates are taken as reference values for the calculation of uptake rate dependence with wind velocity.

To estimate the uptake rates at various wind velocities, batches of 6 diffusive samplers were placed in the dynamic exposure chamber. They were positioned perpendicular to the direction of the airflow on a glass plate in the upper part of exposure chamber. The environmental conditions of tests are presented in Table 1. Each diffusive sampler was tested under at least five wind velocities between 5 and 300 cm s^{-1}. All the other parameters (temperature, relative humidity, sampling time and compound concentration) were fixed for all the tests.

One unexposed sampler is analysed before each analysis series to check the conditioning system and the analysis.

3. Results and discussion

3.1 *Determination of uptake rates retained as reference values*

The results are given in Table 2. The diffusive samplers can be classified into two groups: the first one consists of high uptake rate samplers (Radiello and EMD samplers and badges) preferably designed for indoor and personal exposures (with short sampling times of some hours) and the second (one) regroups the two diffusion tubes with low uptake rates (close to 1 cm^3 min^{-1}) used for ambient air monitoring (with long sampling times of some days). These uptake rates are taken as reference values for the calculation of uptake rate dependence with wind velocity. For each passive sampler, the limit of detection defined as three times the standard deviation of a blank series are also

Table 1. Environmental conditions for the tests in the exposure chambers.

Diffusive sampler type	Compound	Concentration ($\mu g\,m^{-3}$)	Sampling time (hour)	Temperature ($^{\circ}C$)	Relative humidity (%)	Wind speed range ($cm\,s^{-1}$)
Radiello with a white membrane	Benzene	5	24	20 ± 2	50 ± 4	5–300
Radiello with a yellow membrane	Benzene	5	24	20 ± 2	50 ± 4	5–300
Badge with a membrane	Toluene	380	6	20 ± 2	40 ± 4	9–170
EMD sampler	NO_2	50	4	20 ± 2	50 ± 4	8–140
Palmes diffusion tube	NO_2	200	24	20 ± 2	50 ± 4	15–230
Passam diffusion tube	NO_2	200	24	20 ± 2	50 ± 4	20–230

Table 2. Uptake rates (UR) ($cm^3 min^{-1}$) determined under standard conditions (a relative humidity of 40–50%, a temperature of 20°C, a wind velocity of $50 cm s^{-1}$ and a concentration given in Table 1) and limits of detection.

Diffusive sampler type	Compound	Sampling time (hour)	Experimental UR ± standard deviation	Limit of detection ($\mu g m^{-3}$)
Radiello with a white membrane	Benzene	24	57.7 ± 2.9	0.05
Radiello with a yellow membrane	Benzene	24	28.8 ± 2.2	0.1
Badge with a membrane*	Toluene	6	49.4 ± 1.8	ND
EMD sampler	NO_2	4	57 ± 8	3
Palmes diffusion tube	NO_2	24	1.39 ± 0.07	20
Passam diffusion tube	NO_2	24	1.09 ± 0.04	12

*Data supplied by Perez-Ballesta *et al*. [24].
ND: No data reported in the paper of Perez-Ballesta *et al*. [24].

determined using the uptake rate found under standard conditions and reported as additional data in Table 2.

3.2 *Influences of air velocity on the uptake rates*

The variation of the uptake rate by wind velocity is expressed as the ratio of its value at a certain wind speed to the one found under standard conditions. The comparison of the behaviour of different passive sampler geometries with wind velocity tested with different pollutants is possible by means of the normalised variable defined as the variation of uptake rate. The mean variations (biases) induced by wind velocities for each diffusive sampler are reported in Figure 3. The standard deviations of these mean values do not exceed 14%. Wind velocity has an influence on the uptake rates of all diffusive samplers tested. An increase in uptake rate is systematically observed with increased air velocity. According to Yanagisawa *et al*. [25], the evolution follows a model like $A + B \times \ln$ (wv) where wv is the wind velocity and A and B are constants. That is the case for all samplers tested, as indicated in Figure 3.

For high wind velocities, the arising of uptake rate can be explained by shortening the length of the effective diffusion path [25,33]. Close to $200 cm s^{-1}$, an increase of uptake rates between +40 and +50% is found for the two diffusion tubes without any diffusion membrane. Other authors [13,19] reported tests in an exposure chamber a similar magnitude for Palmes tube. Buzica *et al*. [19] confirmed that the increasing of uptake rate is high about 40% from 1 to $2.8 m s^{-1}$. Yanagisawa *et al*. [25] found a magnitude about 60% over the wind velocity range of 0 to $7 m s^{-1}$. An effective and practical way for reducing the effect of air turbulence and improving the precision of measurements by diffusion tubes is the use of a cylindrical protective box as demonstrated by Plaisance *et al*. [13].

On the contrary, this deviation at high velocities is more reduced (from +8 to +18%) for the samplers with a diffusion membrane like the badge and Radiello samplers. The porous membrane tends to limit the effect of shortening diffusion path induced by high wind velocities. The most resistance to wind is found for the Radiello sampler with a

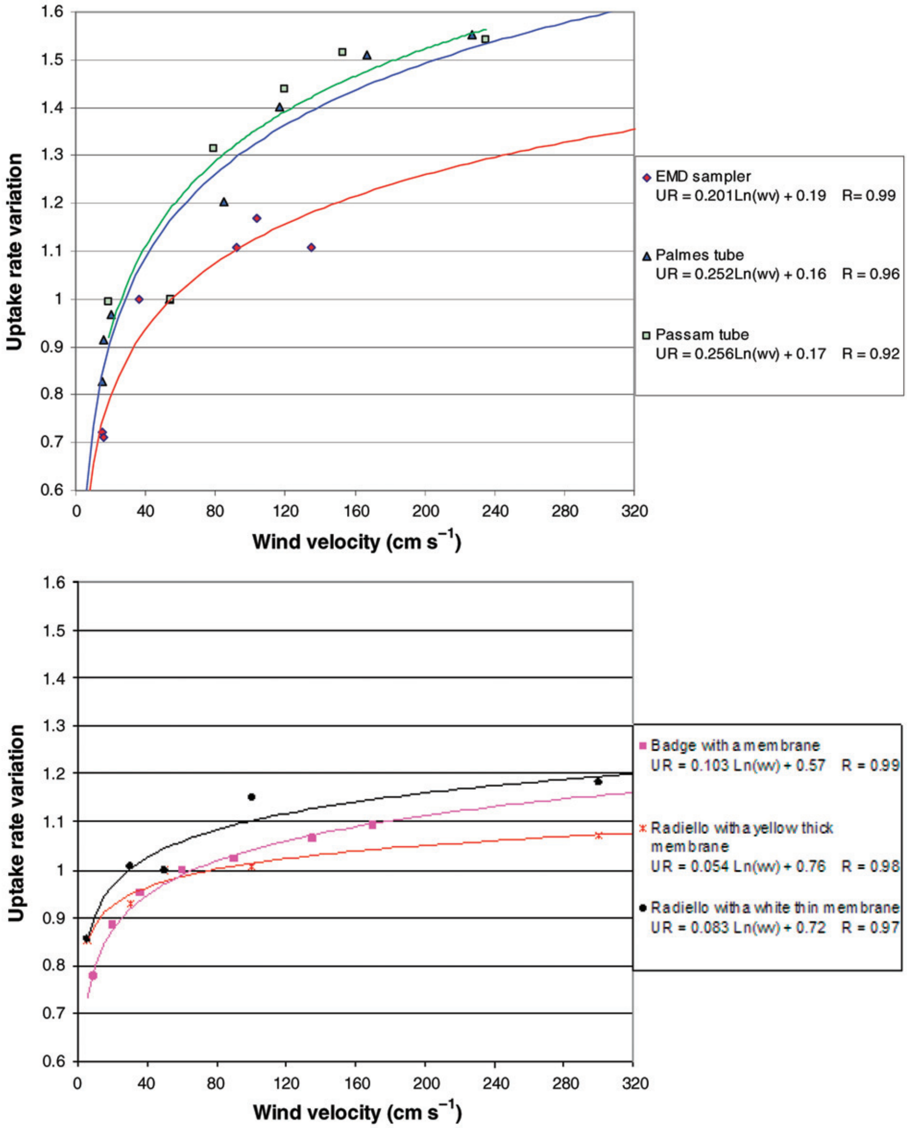

Figure 3. Uptake rate variations of diffusive samplers versus wind velocity (cm s^{-1}).

thick membrane. Then, this resistance also seems to depend on the thickness of membrane. For the EMD sampler, its deviation at high velocities (from +10 to +20%) is intermediate to those of two other types of samplers, although this sampler has not been tested beyond 140 cm s^{-1}. The tortuousness of diffusion path could explain the resistance of this diffusive sampler to high wind velocities.

For weak wind velocities close to 0 cm s^{-1}, a decrease of uptake rates between −13 and −30% compared to the uptake rates found under standard conditions is observed. The increasing of effective diffusion length associated to the setting up of a stagnant air layer at low wind speeds is not higher for the samplers with a membrane than for the

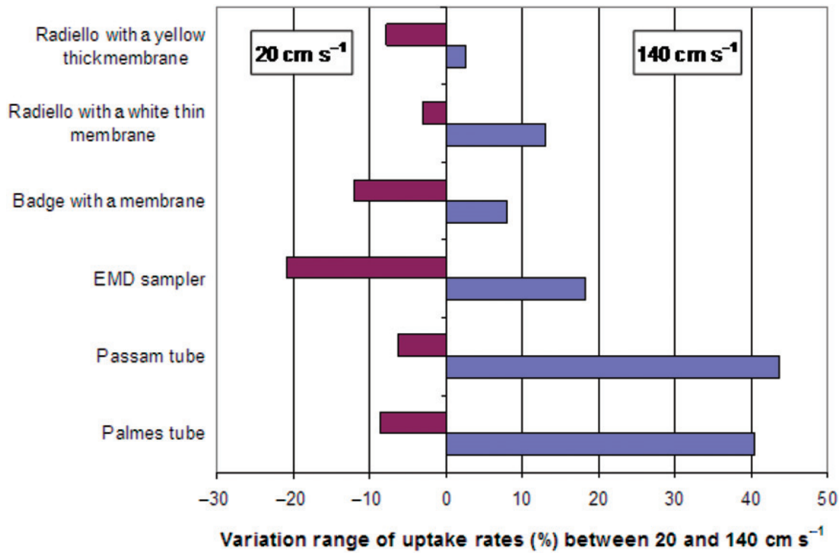

Figure 4. Variation range of uptake rates of six passive samplers between 10 and 200 cm s^{-1}.

samplers without membrane. On the contrary, a low effect is found for the two Radiello samplers (−14%). The radial symmetry of these samplers could be a geometry which favours the upholding of uptake rates at low wind velocities. The stagnation of air which occurs near the sampler at low wind velocities would be more limited in the case of Radiello samplers.

To complete this comparison, the ranges of uptake rates are calculated at 20 and 140 cm s^{-1} by applying the logarithmic regression equations previously found (Figure 3) and reported in Figure 4. These two wind velocities (20 and 140 cm s^{-1}) are chosen because they delimit a domain in which all the samplers were tested. The results confirm that the samplers with a diffusion membrane have a better resistance to wind than the EMD sampler and diffusion tubes. For the last three samplers, the magnitude of wind effects exceeds an uptake rate variation of ±20%. On the other hand, for the Radiello sampler with a thick yellow membrane, the effects of wind speed on the uptake rate tend to be negligible (deviation $< \pm 10\%$). It seems that the use of a thick membrane is a way to privilege to limit the influence of wind on diffusive sampling.

4. Conclusions

The influence of wind velocity on the uptake rates of six diffusive samplers was studied. A progressive increase in the uptake rates was observed with increased air velocity usually following a logarithmic function. The magnitude of wind effects depends on the design of a sampler and is found below ±15% for the samplers equipped with a diffusion membrane. The best resistance to wind is obtained for a radial diffusive sampler equipped with a thick membrane.

The use of a thick porous membrane should be the subject of future investigations for the reduction of air velocity effect on the different types of diffusive samplers.

Furthermore, this research shows that the diffusion tubes are particularly affected by the high wind velocities. By fitting a membrane at the open end of the tube, Gerboles *et al*. [34] demonstrated that the diffusion tube become very little sensible to wind speed. Similar results are obtained in this study for the samplers equipped with a diffusion membrane. To grant the accuracy of these tubes under all the field conditions, the use of membranes should be spread.

These results can help to define new geometries of diffusive samplers which are insensitive to the environmental factors in order to improve the accuracy of the measurements.

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