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Mass Transfer of Diffusive Species with Nonconstant In-Flight Formation and Removal in Laminar Tube Flow. Application to Unattached Short-Lived Radon Daughters

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ABSTRACT. This article deals with convective-diffusive aerosol transport with in-flight formation and removal and is applied to the unattached fraction of short-lived radon decay products. Two novel contributions to previous studies are given in this numerical and experimental work: on the one hand, we solve the mass-transport equations for all the short-lived radon daughters; on the other hand, we include the ^{218}Po neutralization into the mass-transport equation of the first radon decay product. Concerning the mass-transfer of all short-lived radon daughters, numerical calculations lead to the development of simple correlations for the ^{214}Pb and ^{214}Bi penetration fractions. Those correlations can be used to determine the diffusion coefficient of ^{214}Pb and ^{214}Bi using the 2-filter method. In our experiments, a diffusion coefficient equal to 5×10^{-6} m^2 s^{-1} is found for the ^{214}Pb. Concerning the ^{218}Po neutralization, better agreement is observed between our numerical and experimental results when ^{218}Po neutralization is taken into account. These results confirm the neutralization rates found by Howard and Strange (1994).

INTRODUCTION

Mass-transfer is a general engineering topic and describes various technological processes occurring in chemical, nuclear, and environmental applications. The description of transport characteristics is important for understanding the macroscopic behavior of binary mixtures such as solute-solvent, chemical reactions, or fluid-particles systems. For the latter case, convective-diffusive mass transport occurs for fine and ultra-fine aerosols and is of particular interest for aerosol deposition assessments. Particle deposition losses in tubes by random Brownian motion have actually long been considered as a problem in

aerosol sampling devices and in human airways. Considerable research has been carried out to calculate and minimize them. Furthermore, the diffusion coefficient of these particles is an essential property which can be measured by means of tube flow experiment.

To study the convective-diffusive mass-transport, the following general equation can be written (for steady-state transport):

$$\nabla \cdot \left(\vec{v} c_i\right) = D_i \Delta c_i + q_i - p_i, \qquad (1)$$

where c_i and D_i are the concentration (atoms per unit of volume) and the diffusion coefficient of the i-species, v is the gas velocity, and q_i and p_i are the formation and removal rates of the i-constituent.

The fluid flow velocity is obtained using the Navier–Stokes equations: mass and momentum conservation equations for a newtonian incompressible fluid with the condition of adherence at the tube wall.

Equation (1) has already been studied for laminar tube flows, making the following assumptions:

- established fluid velocity profile (parabolic profile), no formation ($q = 0$), and no removal ($p = 0$) of aerosol due to other phenomena than deposition: many authors (Towsend 1898; Gormley and Kennedy 1949; Thomas 1967; Davies 1973; Ingham 1975a; Bowen et al. 1976; Holman 1972) have studied this case and given an analytical solution of the penetration fraction in the tube;
- established fluid velocity profile, constant rate of aerosol formation ($q = cst.$), and no aerosol removal ($p = 0$): this case has already been studied by Berezhnoi and Kirichenko (1964), Tan (1969), and Ingham (1975b);
- constant rate of formation of aerosol ($q = cst.$) and nonconstant rate of removal of aerosol: $p = -\lambda c_i$, where λ is a typical constant of the sink process given per unit of time; Sasse et al. (1994) studied this case with the assumption that the removal process is the radioactive decay of ^{218}Po, the first radon daughter.

In our work, the species studied are the short-lived radon daughters (^{218}Po, ^{214}Pb, ^{214}Bi, ^{214}Po). They are considered to be aerosols of nanometric size, since they have the same property as ultra-fine particles in that they can, unlike a gas, adhere to a surface. These radionuclides are of particular interest because their inhalation causes a radiation dose to lung tissue and is associated with an increased risk of lung cancer.

The aim of our work is to extend the previous studies, especially the one of Sasse et al. (1994). This extension is done at 2 levels:

- solving the mass-transport equations for all the short-lived radon daughters, and not only for the first one, as done previously;
- considering that the ^{218}Po is charged at its birth, ^{218}Po neutralization is included in the model, leading to 2 mass-transport equations for both the neutral and charged fractions of the ^{218}Po aerosol.

Furthermore, in this article, numerical study and experiments specifically designed for this work are carried out simultaneously.

THEORETICAL MODEL AND APPLICATIONS

Classical Model Description

The steady-state mass-transport equations for the ^{218}Po are usually written as

$$\nabla \cdot \left(\vec{v} c_1\right) = D_1 \Delta c_1 + \lambda_0^d c_0 - \lambda_1^d c_1, \qquad (2)$$

where λ^d is a radioactive decay constant (radon ^{222}Rn: $\lambda_0^d = 2.1 \times 10^{-6} \mathrm{s}^{-1}$, ^{218}Po: $\lambda_1^d = 3.8 \times 10^{-3} \mathrm{s}^{-1}$). The second term of the right hand of Equation (2) represents the production of the ^{218}Po due to radon decay.

The third term is the removal by radioactive decay of the ^{218}Po.

For concentration calculations, the following boundary conditions are to be imposed with Equation (2):

$$c_1(r=R, z)=0, \qquad c_1(r, z=0)=0, \qquad (3)$$

where r and z are, respectively, the radial and the axial distance and R is the tube radius. The boundary condition ($r=R$) assures the aerosol collection at the tube wall. The inlet condition ($z=0$) models a filter which is set up at the tube entrance in order to retain all the aerosols that could be produced upstream of the tube, but it lets the radon gas penetrate into the tube.

The influence of the formation and removal processes can be studied by calculating the ^{218}Po penetration fraction through the tube, which is defined as

$$f_{pen_1}=\frac{\phi_{pen_1}}{\phi_{prod_1}}, \qquad (4)$$

where ϕ_{pen_1} and ϕ_{prod_1} are, respectively, the convective flux at the tube outlet and the total rate of production in the whole tube of the ^{218}Po.
ϕ_{pen_1} is obtained by

$$\phi_{pen_1}=\int_0^R v(r, z=L)\cdot c_1(r, z=L)\cdot 2\pi r\, dr. \qquad (5)$$

The total rate of production of ^{218}Po can be calculated analytically by the following expression:

$$\phi_{prod_1}=\int_0^R\int_0^L \lambda_0^d c_0\, dr\, dz=\lambda_0^d c_0 \pi R^2 L. \qquad (6)$$

The main application of those equations concerns the 2-filter method (Fontan 1964; Thomas and LeClare 1970; Frey et al. 1981). This method consists in determining the apparent ^{218}Po diffusion coefficient by comparing the measured ^{218}Po penetration fraction to the theoretical penetration fraction. The conclusion of the numerical work of Sasse et al. (1994), who solve the equations described above, was that the apparent ^{218}Po diffusion coefficient obtained by the 2-filter method using analytical theories (Berezhnoi and Kirichenko 1964; Tan 1969; Ingham 1975b), rather than numerical calculations, is typically 10 to 20% too high.

^{218}Po Classical Model Extended to All Short-Lived Radon Daughters

Following the work of Sasse et al. (1994), using a numerical code we not only studied the ^{218}Po mass transport, but also the ^{214}Pb and ^{214}Bi mass transport. In the stationary case, the coupled mass-transport equations for all short-lived radon daughters are written as

$$\nabla\cdot(\vec{v}c_i)=D_i\Delta c_i+\lambda_{i-1}^d c_{i-1}-\lambda_i^d c_i \quad \text{for } i\geq 1, \qquad (7)$$

where λ_n^d is a radioactive decay constant ($n=0$ stands for radon; $n=1$, 2, 3 stands for ^{218}Po, ^{214}Pb, and ^{214}Bi; $\lambda_0^d=2.1\times 10^{-6}\,s^{-1}$; $\lambda_1^d=3.8\times 10^{-3}\,s^{-1}$; $\lambda_2^d=4.3\times 10^{-4}\,s^{-1}$; $\lambda_3^d=5.9\times 10^{-4}\,s^{-1}$). The second term of the right hand of Equation (7) represents the production of the i-radon daughter due to $i-1$ radon daughter decay. The third term is the removal by radioactive decay of the i element.

This equation is a quite common way to express the coupled dynamic of all the short-lived radon daughters, but the solving of these coupled equations requires a numerical code. Equations (4) and (5) for the penetration fraction and for the convective flux can be generalized for all the radon daughters. For radon daughters other than ^{218}Po, the production rate is not constant, so Equation (6) cannot be used. The total rate of production ϕ_{prod_i} is thus calculated using mass balance: the number of atoms marked i that decay per unit of time is equal to the total number of produced

atoms marked $(i+1)$. This total number of produced atoms $(i+1)$ is equal to the sum of the total number of deposited atoms i and the total number of penetrated atoms i through the tube. To be sure that the total number of atoms is conserved, these summations are done for the 5 first radon daughters (the fifth radon daughter has a half-life of 22.3 years, so it can be considered that its radioactive decay is negligible compared to residence times often used in tubes). The produced flux is then written

$$\phi_{prod_i} = \sum_{j=i}^{5} \left(\phi_{pen_j} + \phi_{dep_j}\right), \tag{8}$$

where the deposited flux ϕ_{dep_j} is defined as

$$\phi_{dep_j} = \int_{z=0}^{z=L} D_j \cdot (\nabla c_j)_{(r=R,\,z)} \cdot 2\pi R\, dz. \tag{9}$$

By only calculating the deposited flux on the tube surface and the penetrated flux on the outlet section, we determine the penetration fraction of each i species.

The penetration fractions of the second and third radon daughter can be used to determine the diffusion coefficients of these species using the 2-filter method, a method usually devoted for the ^{218}Po diffusivity determination. As a matter of fact, the diffusion coefficient of each radon progeny might not be the same because of their different physical and chemical behavior. Raghunath and Kotrappa (1979) have observed a decrease of the diffusion coefficient from parent to daughter. However, the application of the 2-filter method to all the radon daughters can only be done if the penetration fractions are calculated numerically.

Novel Contribution to the ^{218}Po Model

The dynamic of the ^{218}Po depends not only on its radioactive decay, but also on its neutralization and its aggregation with other molecules. It is generally accepted that 88% of the first radon daughter are charged at their birth and only 12% are neutral (Renoux 1965; Porstendörfer and Mercer 1979). It is also admitted that the radon decay products are never present as single atoms, but are always clusters of atoms or molecules (Goldstein and Hopke 1985; Hopke 1996). Several studies have been carried out to characterize and to understand these processes (Raabe and Wrenn 1969; Busigin et al. 1981; Chu and Hopke 1988; Shi and Hopke 1991; Howard and Strange 1992, 1994). It has been found that these 2 processes lead to a variation of the ^{218}Po aerosol apparent diffusion coefficient with the ^{218}Po "age" (which corresponds to a residence time): with ^{218}Po aggregation, the ^{218}Po apparent diffusion coefficient decreases; with ^{218}Po neutralization, the ^{218}Po apparent diffusion coefficient increases (the neutral species has a higher diffusion coefficient than the charged one). As we will see later in this article, we suppose that the increase of the apparent diffusion coefficient with residence time that we observed in our experiments is due to neutralization phenomena. Thus, we choose to take into account, in the mass-transport equations, this mechanism. The different neutralization processes are modelled using a global neutralization rate (λ_1^n). The new set of equations including ^{218}Po neutralization can thus be written as follows:

for the neutral ^{218}Po,

$$\nabla \cdot \left(\vec{v} \cdot c_1^n\right) = D_1^n \Delta c_1^n + 0.12\,\lambda_0^d c_0 - \lambda_1^d c_1^n + \lambda_1^n c_1^c; \tag{10}$$

for the charged ^{218}Po,

$$\nabla \cdot \left(\vec{v} \cdot c_1^c\right) = D_1^c \Delta c_1^c + 0.88\,\lambda_0^d c_0 - \lambda_1^d c_1^c - \lambda_1^n c_1^c, \tag{11}$$

where D_1^c and D_1^n are, respectively, the neutral and charged ^{218}Po diffusion coefficients. The neutralization rate is not well

known and varies with experimental conditions, depending on concentrations of various ambient molecules (Raabe and Wrenn 1969; Busigin et al. 1981; Chu and Hopke 1988; Leung and Philips 1988; Shi and Hopke 1991; Howard and Strange 1992, 1994). In this study, the recent results of Howard and Strange (1994) will be used. They found, in purified air, a neutralization rate defined by $\lambda_1^n = 0.0015\sqrt{\lambda_0^d c_0} + 0.083\,s^{-1}$, leading to values between 0.12 and 0.44 s^{-1} for radon volume activities between 0.5 and 57 $kBq.m^{-3}$.

It should be underlined that the solving of Equations (10) and (11) has not been studied so far for application to radon daughters convective-diffusive mass-transport in laminar tube flow.

In our experimental study, only the global outlet concentration of the ^{218}Po is measured, without separating the neutral and charged fractions. Thus, in order to compare it with our experimental results, the global ^{218}Po penetration fraction is calculated from the neutral and charged concentrations.

OUR NUMERICAL METHOD

The concentration and velocity fields are calculated by solving the complete set of equations described above: Navier–Stokes and mass-transport equations for each species. These calculations have been done using a finite-element method, integrated in a numerical code called TRIO EF, developed in the French Atomic Energy Commission by Magnaud and Goldstein (1989). The numerical methods for flow and mass equations are both implicit decentered resolution methods.

The space discretization has a higher radial density near the tube surface than in the tube center. This radial density is adjusted until it has no influence on the resulting calculations. The finest and the largest radial meshes are, respectively, 0.5 mm and 2.5 mm. The axial meshes are 5 mm wide.

Various validations of this numerical model on previous works concerning laminar tube flows (Gormley and Kennedy 1949; Berezhnoi and Kirichenko 1964; Sasse et al. 1994) have been carried out before our specific study and are all presented in Malet (1997a).

EXPERIMENTAL SET-UP

The experimental set-up is shown in Figure 1.

The air is first purified with silicagel and molecular sieve (13 X) to avoid chemical reactions between radon daughters and air impurities. The concentration of various molecules in this purified air was measured with infra-red spectrometry. This air was found to contain < 1 ppm of N_2O, CO, SO_2, NO, NO_2, C_2H_4, C_2H_6, 1.5 ppm of CH_4, < 2 ppm of HNO_2, HNO_3, C_2H_5OH, < 10 ppm of CO_2, C_6H_6, and 610 ppm of H_2O.

This purified air is injected into a dry radium ^{226}Ra source (Pylon Electronics Inc., Ottawa, Ontario, Canada). The radium decays to radon, which can be mixed with a by-pass dilution air to obtain radon volume activities in the range of 10 and 300 $kBq\,m^{-3}$. Flow-rates are regulated and measured with mass-flow meters (Hastings Instruments, Hampton, VA). A silicon detector (BARASOL, Algade, Limoges, France) is used to measure the radon concentration. This detector was compared with an ionization chamber (AlphaGUARD-Genitron-Genrich 1994) and scintillation cells (Pradel and Billard 1959). The agreement between the different methods was good.

The radon is first injected in a thoron (^{220}Rn) decaying volume. Thoron is a radon isotope with a short radioactive half-life (55

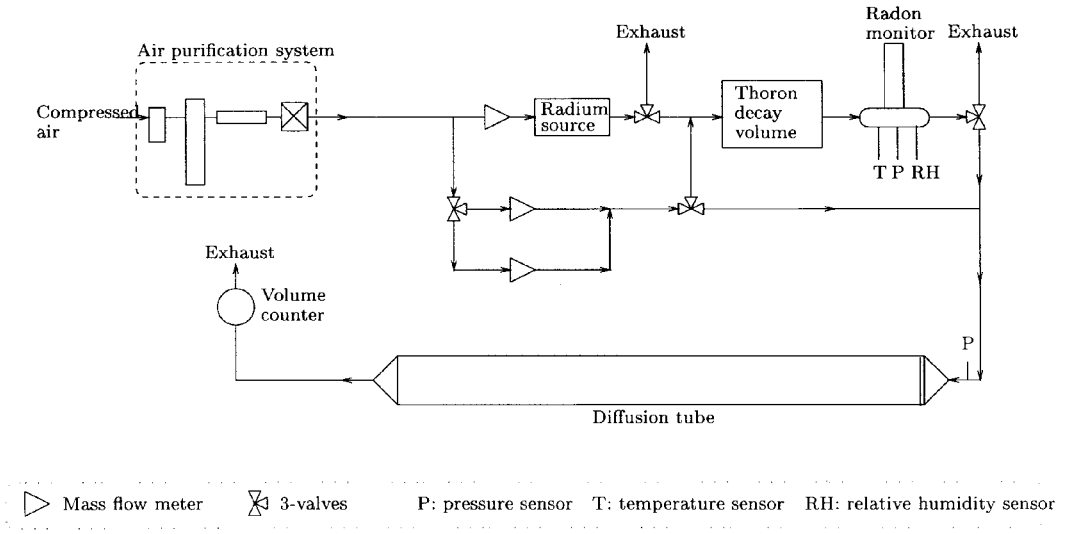

FIGURE 1. Experimental set-up.

s), which can be produced from impurities that can be found in the radium source. This decaying volume guarantees that thoron, and also thoron daughters, cannot be counted as radon or radon daughters with the global α-counting methods used in our experiments. We have also verified by α-spectrometry that there were no thoron daughters in our experiments (Malet 1997b).

The radon is then introduced into the diffusion tube (0.08 m inner diameter and 2 m length, smooth surface of 0.05 μm average height of the protrusions). The tube is thermally insulated to avoid flow perturbation due to temperature differences in the laboratory. Flow-rates in the tube are between 2 and 45 $1\,min^{-1}$, so that residence times are between 13 and 300 s, and Reynolds numbers based on the tube diameter between 40 and 835.

A high efficiency filter is placed on each end of the tube. The upstream filter is used to be sure that the inlet concentration of aerosols in the tube is equal to 0. We have verified that there were no radon daughters passing through this filter by measuring the concentration on a second filter placed just behind the first one. The downstream filter is used to sample the aerosols. The sampling volume is determined with a gas volume counter (Schlumberger, Montrouge, France) which is installed downstream of the tube. This counter was calibrated on a standard volume counter. The filter activity is then measured using gross α-counting methods (Nazaroff 1984) with 1 h counting time. For a given flow-rate ($15\,l\,min^{-1}$), 24 measurements of the ^{218}Po activity on the tube outlet were performed to verify the repeatability of the experiments, which was found to be very good (for a mean ^{218}Po activity of 2584 $Bq\,m^{-3}$, an experimental standard deviation of 20 $Bq\,m^{-3}$ was obtained). For the other flow-rates, each activity was measured at least 3 times.

^{218}Po size-distribution measurements are also performed. This is done using a grid

diffusion battery with cut-off diameters between 1 and 7 nm. The theory used for grid penetration is that of Cheng and Yeh (1980). The inversion data algorithm used here is the Expectation-Maximization algorithm developed by Maher and Laird (1985). More details about these experiments can be found in Malet et al. (1997). A new device, an annular diffusion channel developed by Kerouanton et al. (1996), is also used for the same purpose.

RESULTS AND DISCUSSION

Numerical Calculations

In our numerical work, using the classical model for the ^{218}Po mass-transport equation, i.e., using the same assumptions as Sasse et al. (1994), we come to the same general conclusions on the determination of the ^{218}Po apparent diffusion coefficient (Malet 1997a).

Penetration Fraction of ^{214}Pb and ^{214}Bi. We then apply the classical ^{218}Po mass-transport model on the second and third radon daughters. The numerical calculations show that the first 3 short-lived radon daughters do not always have the same concentration profiles and thus the same penetration fractions. A comparison of radial concentration profiles is given in Figure 2. The non-paraboloidal profiles can be explained as follows: because of the parabolic velocity profile in a laminar tube flow, the air resi-

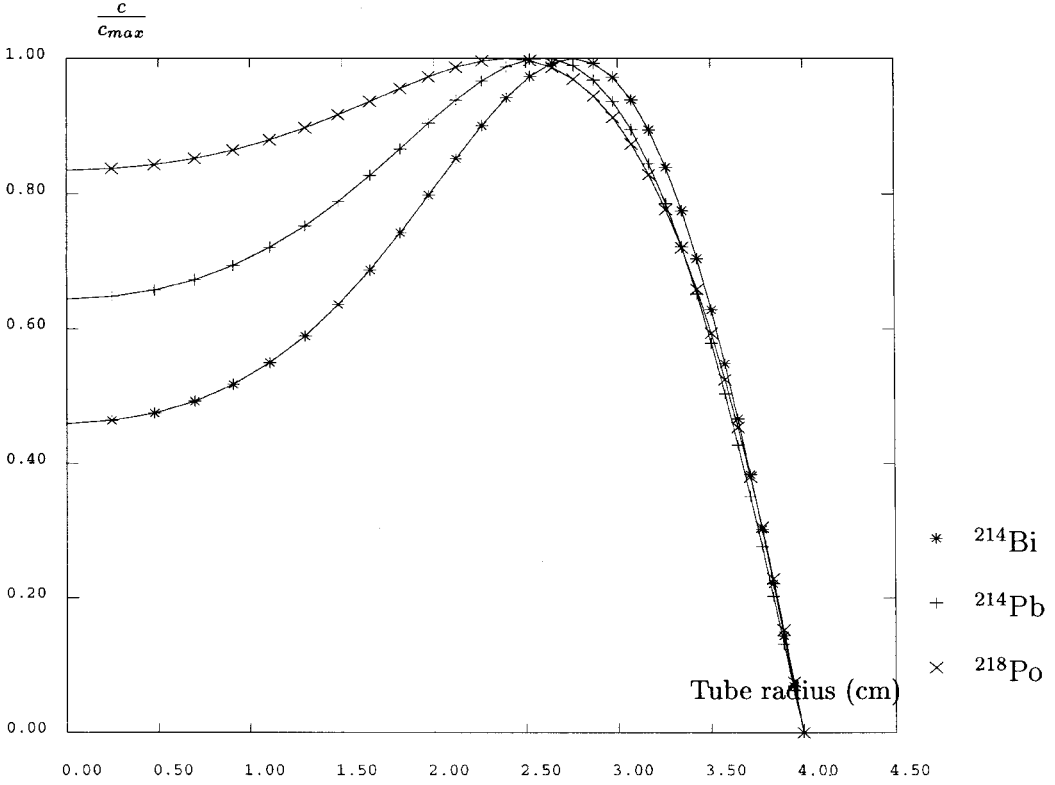

FIGURE 2. Numerical calculation of the radial concentration profiles of the first 3 radon daughters; $z = 2$ m; $R = 0.04$ m; $Re = 500$; $Sc = 3$; $Q/D > 50$ m.

dence time in the center of the tube is shorter than near the walls. The in-flight formation of aerosols is lower in the middle of the tube than near the surfaces. In the case where the convection is much greater than the diffusion process, the aerosol does not diffuse to the tube center and its concentration is reduced in this region. Furthermore, the different concentration profiles for each short-lived radon daughter can be explained by the axial concentration profiles. Due to their different radioactive decays, i.e., to their different disappearance rates, the first 3 radon daughters are not formed at the same time in the tube, as can be seen in Figure 3: for example, at the half length of the tube ($z = 1$ m here), 54% of the ^{218}Po is already formed, where only 32% and 19% of the ^{214}Pb and the ^{214}Bi are produced.

As a result, the first 3 short-lived radon daughters do not have the same penetration fractions. For the second and third radon daughters, these penetration fractions can be calculated only numerically. We calculate the penetration fractions versus the diffusional parameter $\alpha = \frac{\pi DL}{Q}$ (see Figure 4). In order to use these results to determine the diffusion coefficients of the ^{214}Pb and ^{214}Bi with the 2-filter method,

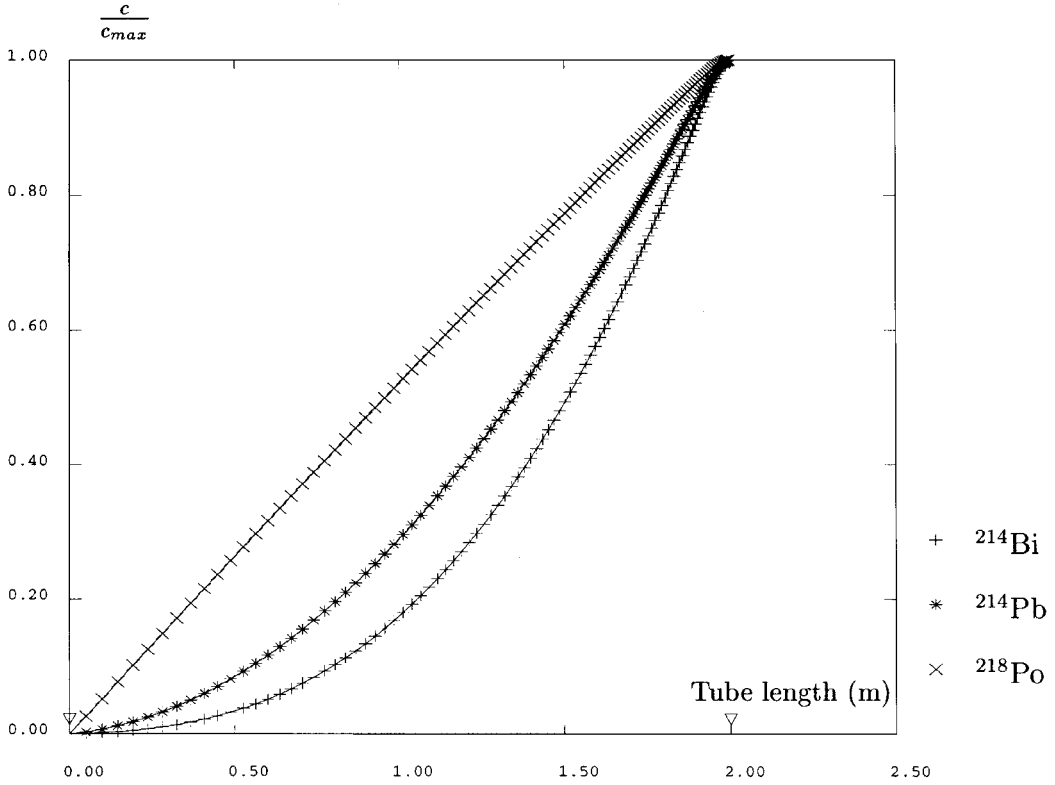

FIGURE 3. Numerical calculation of the axial concentration profiles of the first 3 radon daughters at $r = 0.03$ m; $L = 2$ m; $R = 0.04$ m; $Re = 500$; $Sc = 3$.

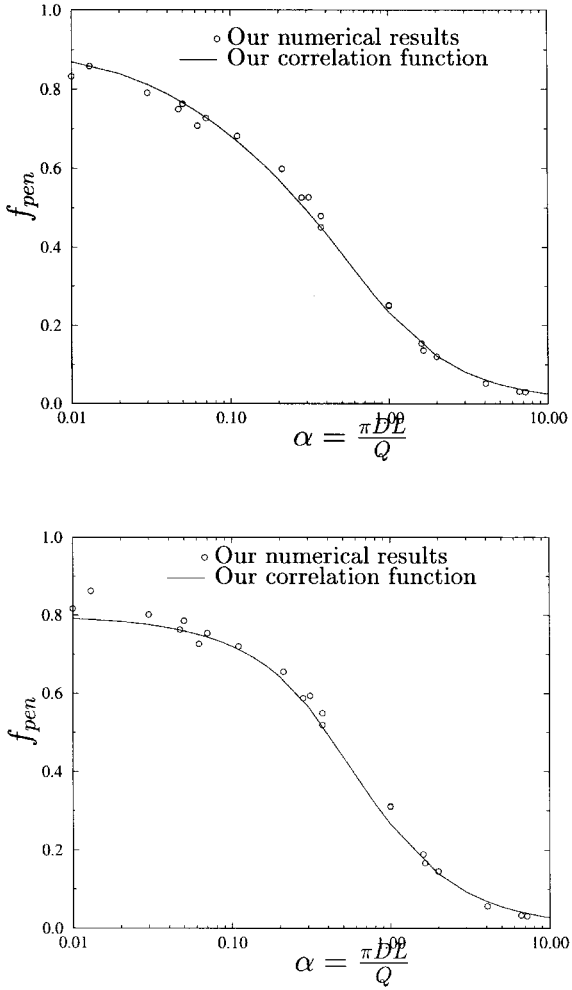

FIGURE 4. Penetration fraction of ^{214}Pb (top) ^{214}Bi (bottom) in laminar tube flow from our numerical calculations.

one would need simple expressions of the penetration fractions. Fitting curves to these results are given as follows.

The ^{214}Pb penetration fraction is given by the following:

For $0.01 \leq \alpha = \frac{\pi DL}{Q} < 10$,

$$f_{pen_{^{214}Pb}} = \frac{5.7}{\alpha}[0.04184(1-\exp^{-3\alpha}) + 0.0012(1-\exp^{-27.5\alpha})], \quad (12)$$

where, in α, D is the diffusion coefficient, L is the tube length, and Q is the tube flow-rate.

For the ^{214}Bi penetration fraction, we obtain the following:

—for $0.4 < \alpha = \frac{\pi DL}{Q} < 10$,

$$f_{pen_{^{214}Bi}} = \frac{5.7}{\alpha}[0.04184(1-\exp^{-3\alpha}) + 0.0012(1-\exp^{-27.5\alpha})]; \quad (13)$$

—for $0.01 \leq \alpha = \frac{\pi DL}{Q} \leq 0.4$,

$$f_{pen_{^{214}Bi}} = 0.8(1-\arctan\alpha). \quad (14)$$

These calculations were done by taking the same diffusion coefficient for all radon daughters and by neglecting the neutralization of ^{218}Po. They are given for 0.5 m $< L <$ 2 m, 0.01 m $< R <$ 0.04 m, 2.10^{-6} m^2 s^{-1} $< D < 9.10^{-6}$ m^2 s^{-1} ($1.5 < Sc < 7$), $Re < 1200$.

Neutralization Importance. As described in the theoretical part, the ^{218}Po mass-transport model is modified in order to include the ^{218}Po neutralization. In order to explain the importance of the neutralization, we plot, in Figure 5, simulated results of (1) the global ^{218}Po penetration fractions obtained with our model for 2 different values of the neutralization rate ($\lambda_1^n = 0.005$ and 0.5 s^{-1}) and (2) the ^{218}Po penetration fractions obtained with the classical model, without taking into account the neutralization process, and for different ^{218}Po diffusion coefficients currently found in the literature: $D = 2.4 \times 10^{-6}$ m^2 s^{-1}, $D = 6.6 \times 10^{-6}$ m^2 s^{-1}, and $D = 5.5 \times 10^{-6}$ m^2 s^{-1} (the first two are currently considered as, respectively, lower and higher values and the last one as a mean value). For $\lambda_1^n =$ 0.005 s^{-1}, the neutralization process is quite slow compared to the residence times in the tube. The aerosol is mainly charged, and the global penetration fraction obtained with our model (including neutralization) is close to that obtained for the case calculated with the classical model (no neutralization) for a lower diffusion coefficient, which could correspond to the charged species ($D = 2.4 \times 10^{-6}$ m^2 s^{-1}). For $\lambda_1^n = 0.5$ s^{-1}, the neutralization process is very fast and the aerosol is neutralized rapidly. The global penetration fraction obtained with our model (including neutral-

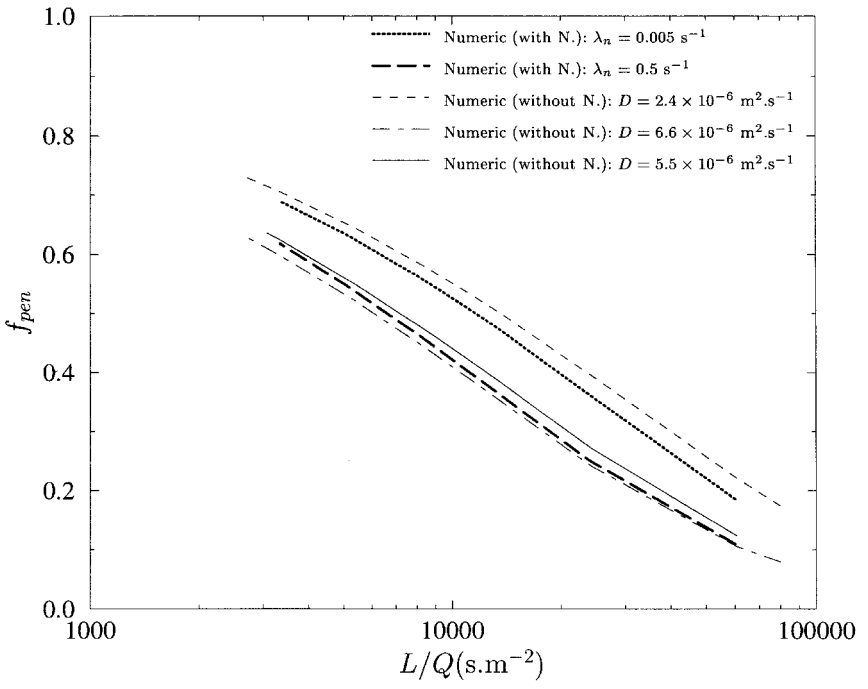

FIGURE 5. **^{218}Po penetration fraction in laminar tube flow. Influence of the ^{218}Po neutralization (numerical results). $D_1^c = 2.4 \times 10^{-6}$ m^2 s^{-1}, $D_1^n = 6.6 \times 10^{-6}$ m^2 s^{-1} (0.6 nm diameter aerosol) (N.: ^{218}Po neutralization).**

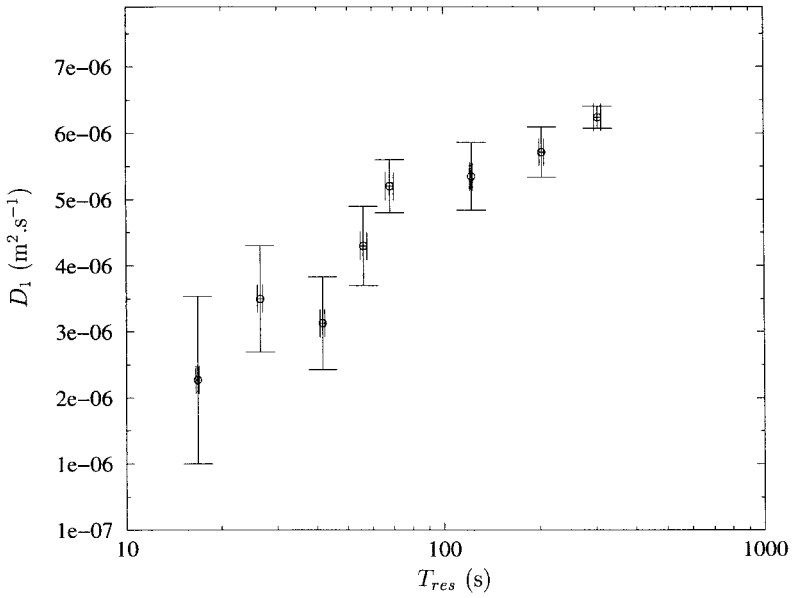

FIGURE 6. Apparent ^{218}Po diffusion coefficients D_1 for different tube residence times T_{res}. Results obtained with the 2-filter method using our numerical calculations. Errors calculated for 95% confidence interval.

ization) is close to that obtained for the case calculated with the classical model (no neutralization) for a higher diffusion coefficient, which could correspond to the neutral species ($D = 6.6 \times 10^{-6}$ m^2 s^{-1}). In this case, the neutralization process can be neglected and the aerosol can be considered as neutral. Between those 2 extreme cases ($\lambda_1^n = 0.005$ s^{-1} and $\lambda_1^n = 0.5$ s^{-1}), the neutralization cannot be neglected.

Furthermore, neglecting this process can lead to some misunderstanding. In Figure 5, it should also be observed that if the neutralization process is taken into account in the mass-transport equations of the neutral and charged ^{218}Po, the global ^{218}Po penetration curve is close to that obtained for the classical model (no neutralization) for an aerosol having a diffusion coefficient of $D = 5.5 \times 10^{-6}$ m^2 s^{-1} (a commonly found value in the literature for the apparent diffusion coefficient of the ^{218}Po). This is why we should call this last value an "apparent diffusion coefficient." This remark can explain the different diffusion coefficients of the global ^{218}Po obtained in former studies on the 2-filter method: in some experiments, the apparent diffusion coefficient that was measured could correspond to a mainly charged fraction and in others, to a mainly neutral fraction. In each case where the neutral and charged fractions exist, the mass-transport equations for both species should be computed to obtain an accurate diffusion coefficient value.

Experiments

The 2-filter method is applied to determine the apparent diffusion coefficients of the first three radon daughters in our experiments.

^{218}Po Diffusion Coefficient. ^{218}Po diffusion coefficients versus the mean residence times in the tube are plotted in Figure 6. An increase in the apparent ^{218}Po diffusion

coefficient with the tube residence time is observed. One can make the hypothesis that this variation can mainly be due to neutralization, since the diffusion coefficients of neutral species are higher than the one of charged species. These experimental results lead us to apply the new set of equations for the mass-transport of ^{218}Po developed in the theoretical section, taking into account this ^{218}Po neutralization mechanism.

^{218}Po Diffusion Coefficients Considering ^{218}Po Neutralization. Considering the ^{218}Po neutralization for the numerical calculations in Equations (10) and (11), 3 parameters are unknown: the diffusion coefficients of both the charged and the neutral ^{218}Po and the neutralization rate of the ^{218}Po. In this paper, as described below, the neutral ^{218}Po diffusion coefficient is determined experimentally, the charged ^{218}Po diffusion coefficient is calculated, and the neutralization rate is obtained using the Howard and Strange (1994) formula as previously described.

The neutral diffusion coefficient is determined with the grid diffusion battery and annular diffusion channel by assuming that for long residence times (300 s), the aerosol should be mainly neutralized. This leads for our experiments to a mean diffusion coefficient of about 6×10^{-6} m^2 s^{-1}, which is also the approximate value found with the 2-filter method for a long residence time (see Figure 6). The corresponding aerosol diameter was obtained using the Ramamurthi (1989) relation. From this measured neutral diameter and using the Chapmann and Cowling (1958) theory, the diffusion coefficient of the charged ^{218}Po was calculated.

In Figure 7, our numerical results for the ^{218}Po penetration considering the neutralization process are compared to our experimental results. It should be observed that the comparison between numerical and experimental results is very good. This comfirms the accuracy of the neutralization rate found by Howard and Strange (1994).

These results could lead to another application of the 2-filter method. In the case where the real diffusion coefficients of the charged and neutral ^{218}Po aerosols will be determined with grid diffusion battery, electrical mobility analyzer, or hypersonic impactor, the unknown parameter of the mass-transport equations will be the neutralization rate. This parameter could be determined by fitting the numerical global penetration fraction to the experimental one. This method will not pretend to give a very precise value of the neutralization rate, but it will be simple to carry out and will be a good indicator of the range of neutralization rates in an experiment.

Second and Third Radon Daughters. The main problem in using the 2-filter method for the second and third radon daughters is due to the experimental errors in their measured concentrations and in the lack of simple expressions of their penetration functions (derived in this paper). The experimental errors are due to the counting methods that are usually used (Thomas 1972; Nazaroff 1984), which are much more precise for the first radon daughter than for the second and the third, especially for cases where there is much more ^{218}Po than ^{214}Pb and ^{214}Bi (cases of the 2-filter method here). Nevertheless, using a high radon activity and longer residence times in the tube, the 2-filter method can also be used for the second and third radon daughters. In our experiments, results with $< 30\%$ relative error in the concentration were kept, so that results on the ^{214}Bi were not precise enough to be presented in this study. The results for the ^{214}Pb are presented in Figure 8. The experimental results of the ^{214}Pb are compared to the numerical results for different diffusion coefficients of the radon daughters. A value around 5×10^{-6} m^2 s^{-1}

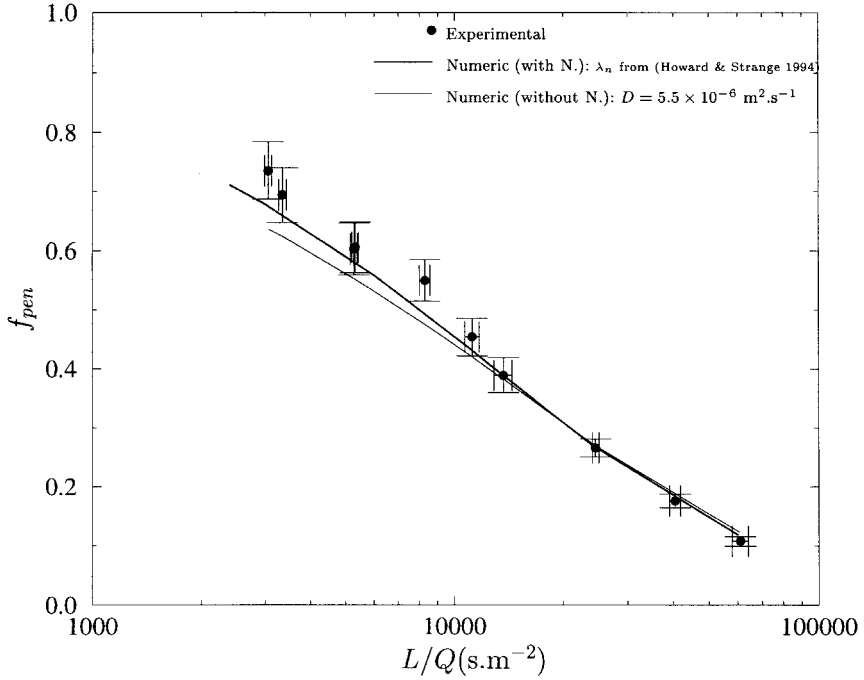

FIGURE 7. ^{218}Po penetration fraction in laminar tube flow. Comparison between our numerical and our experimental results. $D_i^c = 2.4 \times 10^{-6}$ m^2 s^{-1}, $D_i^n = 6.6 \times 10^{-6}$ m^2 s^{-1} (0.6 nm diameter aerosol); errors calculated for 95% confidence interval (N.: ^{218}Po neutralization).

was found to be an accurate value for the diffusion coefficient of the ^{214}Pb. It has to be underlined that the ^{214}Pb diffusion coefficient investigated here is an apparent diffusion coefficient (the chemical and physical state of this species is not studied here).

CONCLUSION

In this work, we study experimentally and numerically the convective-diffusive transport of aerosols with in-flight formation and sink in laminar tube flow. Such study has already been applied to the transport of the first short-lived radon daughter, the ^{218}Po (Sasse et al. 1994), and is here extended, on the one hand, to all the short-lived radon daughters and, on the other hand, to the neutral and charged ^{218}Po.

Generalization of the classical ^{218}Po mass-transport model to all the short-lived radon daughters leads to simple expressions (Equations (12), (13), (14)) of the penetration fractions of these radionuclides. These expressions can then be used for the ^{214}Pb and ^{214}Bi diffusion coefficients determination using the 2-filter method. These simple expressions avoid the use of numerical calculations of Navier–Stokes and mass-transport equations. In our experiments, a value around 5×10^{-6} m^2 s^{-1} is found for the apparent diffusion coefficient of the ^{214}Pb.

Taking the same assumptions as Sasse et al. (1994) and applying the classical 2-filter method, a variation of the apparent diffusion coefficient with the tube residence time is observed. This variation can be explained with neutralization. Using our novel contribution to the classical model, i.e., rewriting

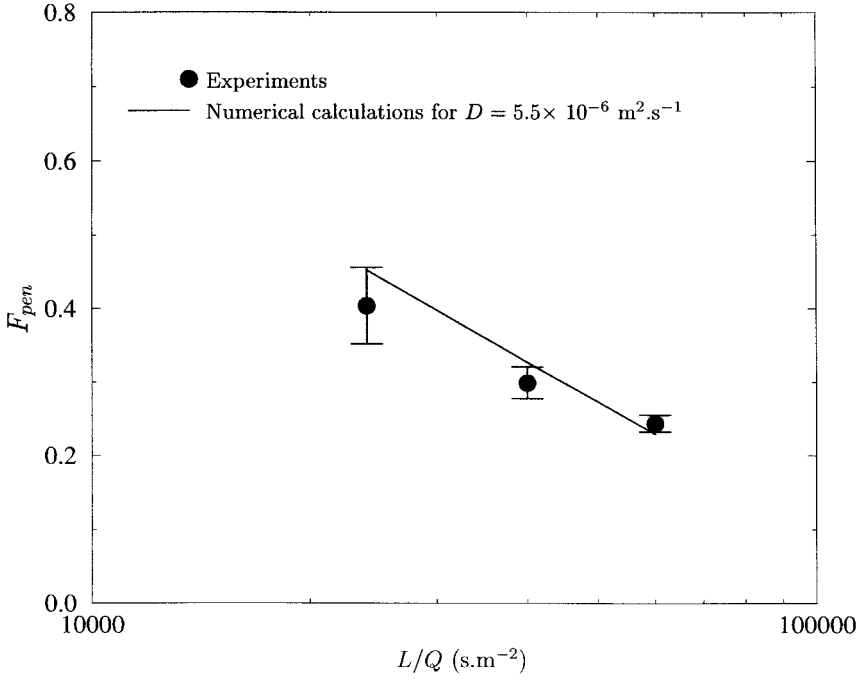

FIGURE 8. Numerical and experimental results for the penetration fraction of the second radon daughter (^{214}Pb).

the usual mass-transport equation for ^{218}Po used by Sasse et al. (1994) as 2 mass-transport equations for the charged and the neutral species, a good agreement was found between our numerical and our experimental results on the global ^{218}Po penetration fraction, using a neutralization rate between 0.08 and 0.39 s^{-1}. This confirms the neutralization rates found by Howard and Strange (1994). It is also concluded that, for that range of ^{218}Po neutralization rates, the ^{218}Po neutralization has to be taken into account in the mass-transport equation and can modify the global ^{218}Po penetration fraction, i.e., the ^{218}Po diffusion coefficient if the 2-filter method is used.

References

Berezhnoi, V. M., and Kirichenko, V. V. (1964). Theory of Diffusive Deposition of Decay Products of Inert Gases in Circular and Flat Channel, *Atomnaya Energiya* 17:1031–1033, translation.

Bowen, B. D., Levine, S., and Epstein, N. (1976). Fine Particle Deposition in Laminar Flow through Parallel Plate and Cylindrical Channels, *J. Colloid Interface Sci.* 54:375–390.

Busigin, A., Van der Vooren, A. W., Babcock, J. C., and Philips, C. R. (1981). The Nature of Unattached RaA (^{218}Po) Particles, *Health Phys.* 40:333–343.

Chapmann, S., and Cowling, T. G. (1958). *The Mathematical Theory of Non-uniform Gases*, Cambridge University Press, Cambridge, England.

Cheng, Y. S., and Yeh, H. C. (1980). Theory of a Screen-Type Diffusion Battery, *J. Aerosol Sci.* 11:313–320.

Chu, K. D., and Hopke, P. K. (1988). Neutralization Kinetics for Polonium-218, *Envir. Sci. Techn.* 22:711–717.

Davies, C. N. (1973). Diffusion and Sedimentation of Aerosol Particles from Poiseuille Flow in Pipes, *J. Aerosol Sci.* 4:317–328.

Fontan, J. (1964). *Dosage des radioéléments gazeux donnat des produits radioactifs de filiation*, Rapport de Thèse 218, Université de Toulouse, France.

Frey, G., Hopke, P., and Stukel, J. J. (1981). Effects of Trace Gases and Water Vapor on the Diffusion Coefficient of ^{218}Po, *Science* 211:480–481.

Genrich, V. (1994). *International Calibration Experiment*, Rapport d'étude, Genitron Instruments, Frankfurt, Germany.

Goldstein, S. D., and Hopke, P. K. (1985). Environmental Neutralization of Polonium-218, *Envir. Sci. Techn.* 19:146–150.

Gormley, P. G., and Kennedy, M. (1949). Diffusion from a Stream Flowing through a Cylindrical Tube, *Proc. Roy. Irish Aca.* 52A: 163–169.

Holman (1972). *Heat Transfer*, McGraw–Hill, New York.

Hopke, P. K. (1996). The Initial Atmospheric Behavior of Radon Decay Products, *J. Radioanalytical and Nuclear Chemistry* 203:353–376.

Howard, A. J., and Strange, W. P. (1992). Measurement of $^{218}_{84}Po^{+}$ Neutralization Rates in Gases, *Nucl. Instr. Meth. Phys. Res. A* 311:378–385.

Howard, A. J., and Strange, W. P. (1994). Determination of $^{218}Po^{+}$ Ionic Neutralization Rates in Gases, *Nucl. Instr. Meth. Phys. Res. A* 349:481–488.

Ingham, D. B. (1975a). Diffusion of Aerosols from a Stream Flowing through a Cylindrical Tube, *J. Aerosol Sci* 6:125–132.

Ingham, D. B. (1975b). Diffusion of Disintegration Products of Radioactive Gases in Circular and Flat Channels *J. Aerosol Sci.* 6:395–402.

Kerouanton, D., Tymen, G., and Boulaud, D. (1996). Small Particle Diffusion Penetration of an Annular Duct Compared to other Geometries, *J. Aerosol Sci.* 27:345–349.

Leung, H. M. Y., and Philips, C. R. (1988). The Electrical and Diffusive Properties of the Unattached ^{218}Po in Air Systems, *Radiat. Prot. Dosim.* 22:97–105.

Magnaud, J.-P., and Goldstein, S. (1989). *The Finite Element Version of the TRIO Code*, 7th International Conference in Fluid Mechanics, Huntsville, AL.

Maher, E. F., and Laird, N. M. (1985). EM Algorithm Reconstruction of Particle Size Distributions from Diffusion Battery Data, *J. Aerosol Sci.* 16:557–570.

Malet, J. (1997a). *Transport et dépôt des aérosols nanométriques. Application à la fraction libre des descendants à vie courte du radon*, Thèse de doctorat, Université Paris XII, Créteil, France.

Malet, J. (1997b). *Transport et dépôt des aérosols nanométriques. Application à la fraction libre des descendants à vie courte du radon*, Rapport CEA-R-5776, Commissariat à l'Energie Atomique, Gif-sur-Yvette, France.

Malet, J., Michielsen, N., Kerouanton, D., Huet, C., Boulaud, D., Renoux, A., and Tymen, G. (1997). Comparison Between Different Methods for ^{218}Po Diffusion Coefficient Measurements, *J. Aerosol Sci.* 28:S709–710.

Nazaroff, W. W. (1984). Optimizing the Total Alpha Three Count Technique for Measuring Concentrations of Radon Progeny in Residences, *Health Phys.* 46:395–405.

Porstendörfer, J., and Mercer, T. T. (1979). Influence of Electric Charge and Humidity upon the Diffusion Coefficient of Radon Decay Products, *Health Phys.* 37:191–199.

Pradel, J., and Billard, F. (1959). *Le thoron et les risques associés dans la manipulation des composés du thorium*, Rapport CEA-R-1165, Commissariat à l'Energie Atomique, Gif-sur-Yvette, France.

Raabe, O. G., and Wrenn, M. E. (1969). Analysis of the Activity of Radon Daughter Samples by Weighted Least Squares, *Health Phys.* 17:593–605.

Raghunath, B., and Kotrappa, P. (1979). Diffusion Coefficients of Decay Products of Radon and Thoron, *J. Aerosol Sci.* 10:133–138.

Ramamurthi, M. (1989). *The Detection and Measurement of the Activity Size Distributions (dp > 0,5 nm) Associated with Radon Decay Products in Indoor Air*, Ph.D. thesis, University of Illinois, Urbana, IL.

Renoux, A. (1965). *Etudes des ions radioactifs de l'atmosphère*, Thèse de doctorat, Faculté des Sciences de Paris, Rapport CEA R-2771, France.

Sasse, A. G. B. M., Gadgil, A. J., and Nazaroff, W. W. (1994). On the Measurement of ^{218}Po Diffusivity using the Two-Filter Method, *J. Aerosol Sci.* 25:689–697.

Shi, B., and Hopke, P. K. (1991). Study of Neutralization of ^{218}Po Ions by Small Ion Recombination in O_2, Ar, and N_2, *Health Phys.* 61:209–214.

Tan, C. W. (1969). Diffusion of Disintegration Products of Inert Gases in Cylindrical Tubes, *Int. J. Heat Mass Transfer*. 12:471–478.

Thomas, J. W. (1967). Particle Loss in Sampling Conduits. In *Symp. Proc. Assessment of Airborne Radioactivity*, Vienna, pp. 701–712.

Thomas, J. W. (1972). Measurements of Radon Daughters in Air, *Health Phys.* 23:783–789.

Thomas, J. W., and LeClare, P. C. (1970). A Study of the Two-Filter Method for ^{222}Rn, *Health Phys.* 18:113–122.

Towsend, J. (1898). *Phil. Trans*. 193A:129.

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