RADIOISOTOPE INVESTIGATION OF IMPURITY TRANSPORT IN AIR FLOWS

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The use of the radioactive tracer method of studying impurity transport in air is investigated. An expression relating the counting rate and the tracer concentration in the gas mixture is obtained. Certain experimental results are presented.

In view of the intensified use of new chemical reagents the question of the transport of harmful impurities by air flows has acquired special importance. The propagation of smokes and toxic gases ejected into the atmosphere by factories and transport vehicles, the motion of gases and vapors in industrial plants, and mass transfer in ventilated and air-conditioned buildings are typical problems of this type.

As an example it is possible to point to the harmful effects of impurities present in the atmosphere of industrial buildings. Thus, toxic gases, aerosols, and vapors present in excessive amounts are responsible for more than 95% of cases of occupational diseases and poisoning [1]. The impurities either leak into the atmosphere or are evaporated from the free surface of the product in the presence of either forced or natural convection. By investigating the transport of these impurities it is possible to organize the air flow in the building with the object of improving the atmosphere and providing healthier working conditions.

As experience has shown, many problems can be successfully solved by using labeled atoms (radioactive tracers). This method is characterized by its sensitivity, accuracy, speed, andeconomy, and by the convenience with which the instrument readings can be automatically recorded.

The tracer method is now being used for investigating the turbulent diffusion of dust in mines [2], for studying the interaction of thermal currents and circulation flows in heated rooms [3], for investigating the behavior of gaseous impurities in air ducts [4], in determining the efficiency of ventilating systems [5], for investigating the distribution of air flows in structures of various kinds [6-8], for studying a series of atmospheric turbulence problems [9], etc. Especially promising is the use of radioactive tracers in model investigations of impurity transport based on the theory of similarity.

In radioisotope studies the tracer gas is mixed in negligibly small amount (10^{-6}-10^{-3}%) with the impurity, whose concentration distribution in the air is to be studied, and transported together with the latter. In turbulent transport the role of molecular processes is insignificant and the impurities are passively entrained by the air flow. The impurity concentration distribution is almost the same for gases with different molecular weights [10] and, accordingly, in studying turbulent flows it is possible to employ radioactive gases with μ as much as several times different from the μ of the nonradioactive impurity. The tracer concentration field is similar to the concentration field of the investigated impurity. Knowing the relation between the amount of impurity introduced into the flow and the activity of the tracer, and having determined the concentration field of the radioactive gas, we can find the field of the investigated impurity.

The author's investigations [11] have shown that Kr-85 and Xe-133 are ideal for solving such problems. Their characteristics are given in [12].

The principal component of the experimental apparatus is usually an end-window counter sensitive to soft β-radiation. The counter may either be located in a fixed volume, into which a sample of the radioactive mixture is drawn from the air flow, or may be placed directly in the labeled air flow. In both cases it may be assumed that the inlet window of the counter occupies the center of a hemisphere filled with

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labeled air. Information on the tracer concentration is given by the pulse counting rate N, which is related with q by the expression

$$N = Kq. \tag{1}$$

Usually, K is determined experimentally for a specific counter and radioactive isotope by crushing in a certain sealed volume an ampul containing a tracer of given activity and measuring the corresponding counting rate.

Except for certain special cases, we lack an analytic expression of the form $K = f(E, \rho, \rho_1, \sigma, \sigma_1, h, r_0, G)$, owing to the difficulty of taking into account all the factors affecting the detection of the radiation. At the same time, a solution of the problem is urgently required. This would make it possible, without resorting to special experiments, to determine the tracer concentration for a variety of radiation detectors and radioactive gases by measuring only the pulse counting rate. Using appropriate formulas, it would be possible to calculate the optimal activity of the tracer required for measuring N with predetermined accuracy before starting the experiment. Finally, the formulas obtained could be widely used in monitoring radioactive gases.

Below we derive an equation from which it is possible to calculate the coefficients K for a case frequently encountered in practice – an end-window radiation detector in a volume of β-active gas.*

Let the counter window of radius r_0 be located in a hemisphere of radius R occupied by an air–radioactive mixture with concentration q (Fig. 1). The coordinate origin is taken at the center of the lower face of the counter window. It is required to calculate the number of β-particles N registered by the counter per unit time.

The elementary β-particle flux through an area element of the counter window emitted by a volume element of the radiating gas

$$d\Phi = \frac{q \cos\alpha \exp\left(-\sigma r_1 - \frac{\sigma_1 h}{\cos\alpha}\right) dV dS}{4\pi r_1^2}. \tag{2}$$

The β-flux entering the counter through the inlet window can be found by integration

$$\Phi = \frac{q}{4\pi} \iiint_V \iint_S \frac{\cos\alpha \exp\left(-\sigma r_1 - \frac{\sigma_1 h}{\cos\alpha}\right) dV dS}{r_1^2}. \tag{3}$$

Since

$$dV = R^2 \sin\varphi dR d\varphi d\psi;\ dS = r dr d\omega;\ \cos\alpha = \frac{R\cos\varphi}{r_1};$$

$$r_1^2 = R^2 + r^2 - 2Rr\cos\gamma;\ \cos\gamma = \sin\varphi\cos(\psi - \omega), \tag{4}$$

relation (3) can be reduced to the following form

$$\Phi = \frac{q}{4\pi} \int_0^{\varphi_0} \int_{h'}^{R_0} \int_0^{2\pi} \int_0^{r_0} \int_0^{2\pi} R^3 r \cos\varphi \sin\varphi \times \exp\left[-\left(\sigma + \frac{\sigma_1 h}{R\cos\varphi}\right) \sqrt{R^2 + r^2 - 2Rr\sin\varphi\cos(\psi - \omega)}\right] \times d\varphi dR d\psi dr d\omega \left\{[R^2 + r^2 - 2Rr\sin\varphi\cos(\psi - \omega)]^{3/2}\right\}^{-1}. \tag{5}$$

Here, it is necessary to make certain remarks concerning the upper limits of integration with respect to φ and R.

Since the β-particles have a finite range, they can enter the counter only from a volume whose characteristic geometric dimension does not exceed the maximum range in the medium in question. If the counter is located in an infinite air medium, then, strictly speaking, the volume from which β-emission is detected will be bounded not by a hemisphere of radius R_{0m} but by the surface of a certain body of

* Most radioactive gases are β-emitters.

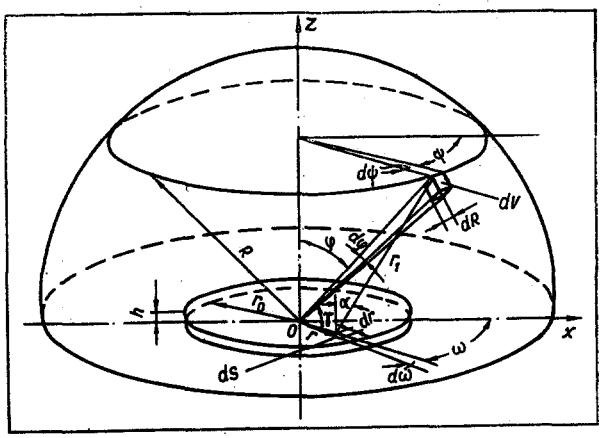

Fig. 1. Diagram illustrating the derivation of Eq. (9).

revolution, at each point on which R_{0m} has a perfectly definite value that is a function of the angle φ (Fig. 2a)

$$R_{0m} = \frac{P_m - \frac{\rho_1 h}{\cos\varphi}}{\rho}. \tag{6}$$

The limiting value of φ is found from the condition $R_0 = h'$; then since $\rho_1 \gg \rho$, we have

$$\varphi_0 = \arccos\frac{\rho_1 h}{P_m}. \tag{7}$$

The relation between the radiation flux and the counting rate is given by

$$N = \varepsilon\lambda\eta\Phi. \tag{8}$$

From Eqs. (1) and (8), using the relation (5), we find the coefficient K. Setting $K = K_1$, we can write

$$K_1 = \frac{\varepsilon\lambda\eta}{4\pi}\int_0^{\varphi_0}\int_{h'}^{R_0}\int_0^{2\pi}\int_0^{r_0}\int_0^{2\pi} R^3 r\cos\varphi\sin\varphi \times \exp\left[-\left(\sigma + \frac{\sigma_1 h}{R\cos\varphi}\right)\sqrt{R^2 + r^2 - 2Rr\sin\varphi\cos(\psi - \omega)}\right] \times d\varphi dR d\psi dr d\omega \left\{[R^2 + r^2 - 2Rr\sin\varphi\cos(\psi - \omega)]^{3/2}\right\}^{-1}. \tag{9}$$

In the general case Eq. (9) cannot be expressed in elementary and tabulated functions; it was solved by numerical integration on a computer. Certain values of K_1, obtained as a result of these calculations, are presented in Table 1.

If we assume that $R \gg r$, $\varphi_0 = \pi/2$ ("point" counter) and that R_{0m} does not depend on φ, we can reduce Eq. (9) to the following form, denoting K for this case by K_2,

$$K_2 = \frac{\varepsilon\lambda\eta S}{2}\int_0^{\frac{\pi}{2}}\int_{h'}^{R_0}\cos\varphi\sin\varphi\exp\left(-\sigma R - \frac{\sigma_1 h}{\cos\varphi}\right)d\varphi dR. \tag{10}$$

After integrating we find

$$K_2 = \frac{\varepsilon\lambda\eta S}{2\sigma}[1 - \exp(-\sigma R_0)]\,E_3(\sigma_1 h). \tag{11}$$

The expression obtained has been proposed for use in connection with the investigation of air flows by the labeled atom method [6] and the monitoring of radioactive gases [13]. As we have shown, it can be derived as a particular case of Eq. (9). The authors of [6] and [13] do not give data on the limits of applicability of Eq. (11).

TABLE 1. Values of K Characteristic of End-Window Counters ($\varepsilon = 1$, $\lambda = 1$, $\eta = 1$)

E, MeV	ρ_1, h, kg/m²							
	0,01	0,03	0,01	0,03	0,03	0,05	0,03	0,05
—	r_0=0,25 cm		r_0=0,9 cm		r_0=1,5 cm		r_0=2,5 cm	
0,155	0,132	0,0692	1,89	1,07	6,47	3,56	15,5	8,80
0,345	0,448	0,343	5,86	4,49	12,7	9,98	37,1	29,5
0,470	0,910	0,784	11,8	10,2	28,3	24,7	79,6	69,7
0,670	1,67	1,53	21,7	19,9	55,2	50,8	154	142
0,897	2,55	2,40	33,0	31,0	86,3	81,4	241	227

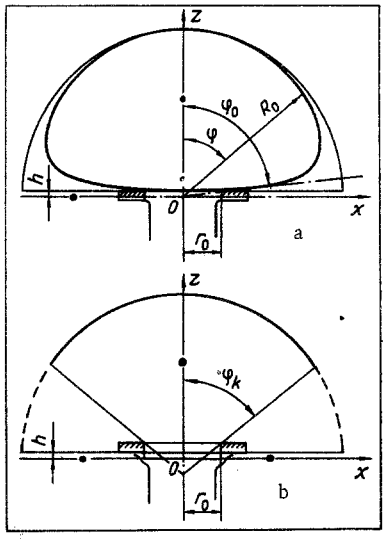

Fig. 2

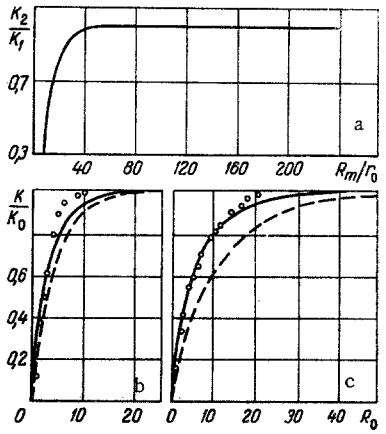

Fig. 3

Fig. 2. Geometry of end-window counter measurements: a) ideal ($\varphi = 0$-φ_0); b) actual ($\varphi = 0$-φ_k; $\varphi_k < \varphi_0$).

Fig. 3. Graphs illustrating the investigation of Eqs. (9) and (11) and the comparison of the calculated and experimental data: a) the dependence $K_2/K_1 = f(R_m/r_0)$, $R_0 = R_{0m}$; b) the dependence $K/K_0 = f(R_0)$ for $C^{14}O_2$; c) the dependence $K/K_0 = f(R_0)$ for Xe-133; the continuous curve corresponds to $K = K_1$, the dashed curve to $K = K_2$; the points represent the experimental data.

The results obtained using relation (11) can be evaluated by comparing them with the data derived from general equation (9). Such a comparison has been made in Fig. 3a. Clearly, at small R_m/r_0 the values of K_1 and K_2 differ by as much as several times. As might have been expected, with increase in R_m/r_0 the difference between K_1 and K_2 decreases: at $R_m/r_0 \geq 25$ it is already less than 10%, and starting from $R_m/r_0 = 50$ K_1 and K_2 are practically equal.

R_m is a function of P_m, which, in its turn, depends on E. For the counters usually employed $r_0 = 2.5$ -30 mm, and $\rho_1 h = 0.01$-0.05 kg/m^2. Thus, it can be established that in the general case Eq. (11) is suitable for use in radiometric calculations only at $E \geq 0.3$ MeV. For softer β-emitters Eq. (11) leads to satisfactory results only at $r_0 < 25$ mm and $\rho_1 h < 0.05$ kg/m^2. For example, for radioactive gases based on C-14 ($E = 0.16$ MeV) this equation is unsuitable for use at $r_0 > 10$ mm and $\rho_1 h > 0.03$ kg/m^2.

When Eq. (11) is inappropriate, it is possible to employ the data of Table 1 or graphs obtained on the basis of those data.*

On being placed in a labeled air flow or a hemispherical chamber, into which a sample of the flow is drawn, the counter detects the β-emission from a certain volume with radius determined by P_m. It is interesting to consider the contributions to the counting rate of the individual layers of labeled air. This would make it possible to select the optimal sampler volume and estimate the actual volume from which β-particles enter the counter. Except for a curve of the form $K_1/K_0 = 1 - \exp(-\sigma R_0)$, following from Eq. (11), nothing has been published on this point.

In order to solve the problem we numerically integrated Eq. (9) on a computer for various values of the limit R_0 ($R_0 < R_{0m}$). Moreover, we conducted a series of experiments with radioactive tracer gases. The relations obtained are compared in Fig. 3a, b. The principal contribution to the counting rate is made by the layers of air nearest to the counter window. As R_0 increases, the growth of the counting rate slows and at $R_0 \geq R_m/3$ practically vanishes. The experimental points lie quite close to the curves obtained from Eq. (9). A slight deviation is observed only at large R_0. The investigation showed that this agreement holds up to $E = 0.5$ MeV; end-window counters are usually employed precisely in this region of maximum

* In deriving Eqs. (9) and (11) it was assumed that the tracer is uniformly distributed in the air volume surrounding the counter. It has been shown [14] that a nonuniform distribution has little effect on the results of the K measurements.

TABLE 2. Corrections for Measurement Geometry

Counter	Isotope	E, MeV	φk, deg	η
SBT-9	Xe-133	0,345	33	0,32
SBT-7	Xe-133	0,345	37	0,42
SBT-9	Kr-85	0,670	33	0,31
SBT-7	Kr-85	0,670	37	0,38

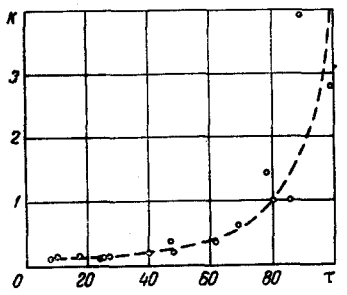

Fig. 4. Experimental dependence of the coefficient K (cm^3) as a function of the time τ (days) elapsed since preparation of the Xe-133 (SBT-9 counter; $R_0 = R_{0m}$).

β-particle energies. The significant deviation of the calculated from the experimental data at large R_0 and E may be attributable to the fact that the exponential law, always employed to describe beta absorption, is not sufficiently rigorous [15].

The agreement with the experimental results of the data obtained from Eq. (11) is much less satisfactory; consequently, it scarcely makes sense to use relation (11) in calculations relating to the dependence of the counting rate on the labeled air volume.

As mentioned above, the corrections ε, λ, and η are introduced into the calculations. The first of these is the probability of detection of a particle entering the counter (for β-emission $\varepsilon = 1$); the second is determined by the number of β-particles emitted per disintegration of the tracer nucleus (found from its radioactive decay scheme); finally, the third correction takes into account the geometry of the measurements and its physical significance deserves closer scrutiny.

In ordinary counters because of design considerations the inlet window is depressed as compared with the ideal window, for which the calculations have been made. Moreover, the sensitive volume of the counter begins not from the inner face of the inlet window, but at a distance of several millimeters from the latter [16]. Accordingly, under actual conditions the β-particles enter the counter from an air volume bounded not by the angle $2\varphi_0$, but by an angle $2\varphi_k$ that depends on the type of counter (Fig. 2b).

An approximate value of η can be obtained from the expression

$$\eta = \frac{E_3(\sigma_1 h) - \cos^2\varphi_k E_3\left(\frac{\sigma_1 h}{\cos\varphi_k}\right)}{E_3(\sigma_1 h)}. \quad (12)$$

Table 2 gives values of η calculated from relation (12) for the optimal tracers Xe-133 and Kr-85 and Soviet end-window counters in common use.

Special experiments showed that the values of N obtained by calculation using Eqs. (1) and (9) are in full agreement with the values of N determined experimentally. On average the difference was of the order of 15%.

In investigating the propagation of toxic gases, and dusts, and other substances in air flows it is necessary to give serious attention to the long-lived radioactive gas impurities that may be contained in the ampuls of tracer gas. The noxious effects of such impurities are apparent with reference to the example of Xe-133. On the day they are prepared ampuls of the latter still contain Kr-85 (up to 1%) and Xe-131m (up to 5%). The half-lives of these isotopes (10.6 years and 12 days, respectively) are greater than that of Xe-133 itself (5.3 days). With time the fraction of Xe-133 activity in the ampul falls, while the fraction of Kr-85 activity increases. Failure to give proper consideration to this fact may lead to distortion of the results of determinations of Xe-133 concentration in the air, since the K for the gas in the ampul will be essentially different from the K characteristic of Xe-133 (the role of Xe-131m in this effect is only slight since it is a γ-active gas and γ-rays are detected with a probability ε hundreds and even thousands of times less than that for β-particles). As an example, in Fig. 4 we show data on the dependence of K on the time elapsed since the date of preparation of the Xe-133. Clearly, only up to $\tau < 30$ days does the value of K remain practically constant. With further increase in τ the value of K increases sharply. This increase is accompanied by intense scatter of the experimental points: ampuls from different batches may contain different amounts of Kr-85, whose effect is negligible at small τ.

NOTATION

μ	is the molecular weight;
m	is the amount of impurity introduced into the air per unit time;
a	is the activity of the tracer gas mixed with the impurity;
q	is the concentration (specific activity) of the tracer in the air;
N	is the pulse counting rate;
K	is a coefficient that depends on the maximum energy of the particles, the type of counter, and the measurement geometry;
E	is the maximum energy of the β-spectrum;
ρ	is the air density;
ρ_1	is the density of the material of the counter window (usually mica);
h	is the thickness of the layer of counter window material;
σ	is the linear absorption coefficient for β-particles in air;
σ_1	is the same in the material of the counter window;
G	is the geometry factor;
r_0	is the window radius
Φ	is the β-particle flux through the counter window;
V	is the air volume in which the labeled impurity is contained;
S	is the area of the window;
r_1	is the line segment joining dV and dS;
α	is the angle between r_1 and the normal to dS;
R	is the radius vector of dV;
φ	is the angle between R and the z axis;
ψ	is the polar angle defining dV;
r	is the radius vector of dS;
ω	is the polar angle defining dS;
γ	is the angle opposite r_1 in the triangle formed by R, r, and r_1; $h' = h/\cos\varphi$;
R_0	is the radius of the hemispherical volume in which the counter is located, cm;
P_m	is the maximum range of the β-particles;
φ_0	is the limiting value of the angle φ;
R_{0m}	is the limiting value of R_0 for a given angle φ;
R_m	is the maximum value of R_{0m} [$R_m = (P_m - \rho_1 h)/\rho$];
ε	is the counter efficiency;
λ	is the β-particle yield per disintegration;
η	is the geometry correction;
K_0	is the value of K at R_{0m};
K_1	is the value of K calculated from Eq. (9);
K_2	is the value of K calculated from Eq. (11);
φ_k	is the maximum value of φ obtainable in practice;
$E_3(\sigma_1 h)$ and $E_3(\sigma_1 h/\cos\varphi_k)$	are respectively the tabulated integral functions

$$\sigma_1^2 h^2 \int_{\sigma_1 h}^{\infty} \frac{\exp(-t)dt}{t^3} \text{ and } \frac{\sigma_1^2 h^2}{\cos^2\varphi_k} \int_{\frac{\sigma_1 h}{\cos\varphi_k}}^{\infty} \frac{\exp(-t)\,dt}{t^3}.$$

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