

Residence time of atmospheric pollutants as dependent on source characteristics, atmospheric diffusion processes and sink mechanisms

By BERT BOLIN, GÖRAN ASPLING and CHRISTER PERSSON, *Department of Meteorology, University of Stockholm, Sweden*¹

(Manuscript received September 28; revised version November 11, 1973)

ABSTRACT

A brief discussion of the sink mechanisms of atmospheric pollutants is given and particular emphasis is given to the relative importance of rain-out (wash-out) and dry deposition. The turbulent transfer processes that determine the latter are analyzed in the light of the classical theory for the atmospheric surface boundary layer. The transfer rates to the free atmosphere (where rain-out and wash-out are the sink mechanisms) and to the earth's surface (when dry deposition occurs) are computed as dependent on height of emission, roughness of the earth's surface, deposition velocity (or accommodation efficiency) at the earth's surface and the intensity of the turbulent processes (as dependent on wind velocity). On the basis of such computations the residence time of atmospheric pollutants are computed and the article concludes with a discussion of the relative importance of rain-out (wash-out) and dry deposition in this regard.

1. Introduction

In air pollution studies the main concern usually is the determination of the concentration of the pollutant in the air as a result of source characteristics and the turbulent diffusion and dispersion by atmospheric motions. As a result of work during several decades we now know reasonably well the diffusive capability of the atmosphere under various meteorological conditions in the neighbourhood of a point source and also within and in the vicinity of a source cluster such as a city or an industrial area.

However, pollutants do not stay in the atmosphere for ever. As emissions grow we are also gradually becoming more and more concerned with the final fate of the pollutants, since damage may not only be the result of high concentrations in the air, but also depend on the accumulation of pollutants on the ground (cf. Munn & Bolin, 1971; Sweden's case study to the UN Conference on the Human Environment, 1971). The deposition pattern largely

depends on the characteristic residence time or transit time of the compound in the atmosphere and the way this time is determined by the interplay between the different sink mechanisms (cf. Bolin & Rodhe, 1973). This residence time also is an important parameter in discussions of the natural cycles of such compounds and is therefore of major importance whenever we wish to consider the atmosphere as a link in geochemical and ecological systems.

In the present paper we shall explore in some detail the way in which the residence time of an atmospheric compound is dependent on (a) source characteristics (height of emission), (b) atmospheric diffusion (horizontal and vertical transfer), and (c) sink mechanisms (essentially rainout and dry deposition).

We are of course basically concerned with the three-dimensional dispersion of matter, and in reality the complexity of atmospheric motions often does not allow of a simplification of the description in space of the relevant processes to one dimension (the vertical), as will be done below. This is obviously the reason for developing numerical models to permit a reasonably adequate description of the dispersion process

¹ Contribution no. 276.

both horizontally and vertically. It is still of some interest to discuss generally the vertical transfer process without regard to horizontal transfer or the occurrence of horizontal inhomogeneities of the dispersion characteristics of the underlying surface. The vertical transfer determines the relative importance of, on the one hand, rain-out (and wash-out) being dependent on processes in the free atmosphere and, on the other, dry deposition at the earth's surface. The results may be of value in the subsequent development of the more sophisticated three-dimensional models of atmospheric dispersion of matter on a regional scale.

2. Sink mechanisms for atmospheric constituents

2.1. General survey

We shall not discuss all possible sink mechanisms in detail, since they depend to a large extent on the compound being considered, but will merely refer to basic text-books (cf. Mason, 1971). The main mechanisms are:

(1) *Sedimentation*, which is of significance only for particles with radii larger than 5–10 μm .

(2) *Rain-out*, due to (a) the consumption of condensation nuclei (aerosols) during the formation of cloud elements, (b) the subsequent attachment of aerosol particles to cloud elements as a result of Brownian motion, and (c) the adsorption of trace gases to cloud elements.

(3) *Wash-out*, due to (a) the collection of aerosol particles on falling precipitation, (b) the adsorption and fixation of trace gases to falling precipitation.

(4) *Chemical reactions*, which need to be considered whenever we do not merely wish to study the cycles of basic elements (sulfur, carbon, etc.) but also the transformation of chemical compounds (e.g. SO_2 , H_2SO_3 , H_2SO_4 , etc.).

(5) *Dry deposition*, which implies the impingement of particles on to roughness elements of the earth's surface as well as the absorption of gaseous compounds.

For most compounds rain-out (2) and dry deposition (5) are the dominant sink processes. (The wash-out mechanism, furthermore, has many things in common with rain-out.) In the following we shall restrict ourselves to a study of their relative importance under varying meteorological conditions.

2.2. Rain-out

The efficiency of the rain-out mechanism depends on (a) the efficiency of the micro-physical processes in the clouds and (b) the frequency of precipitation.

The consumption of aerosol particles by the cloud drops varies greatly with particle radius and the adsorption of gases to cloud elements depends on the chemical composition of the cloud drops. Much more work is required here to understand these processes quantitatively, but it is interesting to note, for example, that there is evidence of a rather rapid transformation of sulfur dioxide into sulfuric acid in clouds as long as the pH of the cloud drop water is significantly higher than 4 (cf. Brosset, 1973). It is quite clear, however, that even so, the most important factor in determining the efficiency of the rain-out mechanism for removal of pollutants from the atmosphere is the frequency of rains (Rodhe & Grandell, 1973). On the basis of rain statistics from Stockholm they showed that even in the case of a very effective transfer of the pollutants into the cloud drops and ultimately into the rain drops, the average residence time for a pollutant in the atmosphere would be about 40 hours in winter and 90 hours in summer, if rain-out were the only sink mechanism. The values are of course only approximate and would certainly be different in another climatic region. Rodhe & Grandell (l.c.) also derived the distribution function for the probability of rain-out of a pollutant released at an arbitrary instant (cf. also Bolin & Rodhe, 1973). A more precise description of this kind might well be important for the present problem, but here we shall merely assume that the removal by precipitation may be described by a characteristic mean residence time τ_0 and that removal due to this process at any one place is statistically proportional to the concentration in the air (see below).

2.3. Dry deposition

The turbulent transfer of pollutants towards the surface of the earth and deposition on the ground, vegetation, the sea surface, etc., is a complicated process that is only partly understood. It is generally accepted that the transfer of momentum, heat, and matter is accomplished in about the same manner in the surface boundary layer except close to the earth's surface.

In other words the coefficients of eddy viscosity, eddy heat conductivity and eddy diffusivity are about the same, but they all depend critically on the wind and temperature distributions. The fundamental problem of atmospheric turbulence in the surface boundary layer is to determine the implicit relations between the structure of turbulence, the distribution of wind and temperature and the magnitude of the various transfers that occur. Even though this problem has not been finally solved, our present knowledge is adequate for some simple computations.

As the surface of deposition (or absorption) is approached, the similarity between transfer of momentum, heat and matter ceases to be valid. This is due to the fact that molecular processes come into play, since the intensity of the turbulent motions necessarily tends towards zero close to the surface. Molecular viscosity and heat conductivity are dependent on different aspects of the molecular processes in the boundary layer next to the surface and the transfer of gases and particles, for example, depend on the molecular weight of the gas or the size of the particles under consideration. Furthermore, the surface may not be a perfect sink and retain each impinging molecule or particle, while obviously the transfer of momentum and energy is not dependent on the "efficiency" of the surface as a sink in the same manner. Considerable progress has been made in recent years with regard to improving our knowledge of the deposition process on smooth surfaces (cf. Sehmel, 1970), but the extension of these results to natural surfaces is not clear. Thus it is useful first to review briefly the concept of deposition velocity, and its inverse, the transfer resistance, and their relation to the basic dynamic processes that describe the turbulence in the surface layer (cf. Chamberlain, 1966).

Let F be the vertical flux of a trace substance, $q(z)$ the concentration in the air of this substance and ρ the air density. The definition of the deposition velocity between the two levels z_1 and z_2 , $v(z_1, z_2)$, is then given by

$$v(z_1, z_2) = \frac{F}{\rho[q(z_1) - q(z_2)]} \quad (2.1)$$

Since $v(z_1, z_2)$ refers to the transfer between the two levels z_1 and z_2 , "transfer velocity" would

be a more appropriate term than deposition velocity. The transfer resistance between these two levels is then defined by

$$r(z_1, z_2) = \frac{1}{v(z_1, z_2)} = \frac{q(z_1) - q(z_2)}{F/\rho} \quad (2.2)$$

The expression (2.2) defines the distribution of the resistance through the layer in analogy with electrical resistance being given by the ratio of electrical potential difference and current. The flux F is assumed to be constant through the surface layer and the resistance is proportional to the concentration difference across the layer. The resistances are additive.

In the lowest part of the dynamic sublayer (cf. Monin, 1970) we may consider the transfer of momentum to be independent of stratification. Thus

$$u(z) = \frac{u_*}{k} \ln \frac{z}{z_0} \quad (2.3)$$

where z is measured from the so-called "displacement level", $u(z)$ is the horizontal wind velocity at z , $u_*^2 = \tau/\rho$ is the friction velocity, τ is the stress or momentum flux, $k = 0.4$ is the von Karman's constant and z_0 is the roughness length of the underlying surface. At the level $z = z_0$, $u(z) = 0$. In analogy with the definition (2.1) we may define a "momentum transfer velocity" $v_M(z_1, z_2)$

$$v_M(z_1, z_2) = \frac{\tau}{\rho[u(z_1) - u(z_2)]} = \frac{u_*^2}{u(z_1) - u(z_2)} \quad (2.4)$$

For the layer down to $z = z_0$ we get

$$v_M(z_1, z_0) = \frac{u_*^2}{u(z_1)} \quad (2.5)$$

Thus, the resistance through this layer (z_1, z_0) is

$$r(z_1, z_0) = \frac{u(z_1)}{u_*^2} = \frac{1}{ku_*} \ln \frac{z_1}{z_0} \quad (2.6)$$

As was stated above it is reasonable to assume that the turbulent transfer of matter through this layer is described by an identical mechanism, i.e. $D = K_M = ku_*z$ where D is the turbulent diffusivity. We may thus consider (2.6) as a general expression for the resistance down to

the level z_0 , provided of course that the simple logarithmic wind law is valid. Otherwise a more general expression involving also the effect of the stratification may be derived in the way given by Monin & Obukhov.

The final transfer of matter to the surface requires other mechanisms than for transfer of momentum. Even in the case where the surface is a perfect sink, i.e. each impinging molecule or particle stays there, $q(z) \geq 0$ because of the resistance for transfer from the level $z = z_0$ to the surface. It follows from (2.1) and (2.6) that

$$q(z_1) - q(z_0) = \frac{F}{\rho k u_*} \ln \frac{z_1}{z_0} \quad (2.7)$$

Chamberlain (1966) has studied this problem in a wind tunnel. By formally extending the integration to a level $z = z_v$, where $q(z_v) = 0$, in the case where the surface is a perfect sink, he defines a roughness length for mass transfer and we may accordingly write

$$r(z_1, z_v) = r(z_1, z_0) + r(z_0, z_v) = r(z_1, z_0) + \frac{1}{k u_*} \ln \frac{z_0}{z_v} \quad (2.8)$$

Chamberlain's experiments show that z_v is a function of the character of the surface but also of the turbulence in the layer above, i.e. u_* , and the trace substance, e.g. molecular weight in the case of a gas and presumably particle size in the case of an aerosol.

If the surface is not a perfect sink, an additional resistance exists for the final transfer of the tracer to the surface, since only a fraction of the impinging molecules or particles stick to the surface. We may formally describe this situation by replacing the boundary condition at $z = z_v$ for a perfect sink, i.e. $q(z_v) = 0$, by

$$v_s q(z_v) = k u_* z_v \left(\frac{\partial q}{\partial z} \right)_{z=z_v} \quad (2.9)$$

If $v_s \rightarrow \infty$, $q(z_v) \rightarrow 0$ and we have the case of a perfect sink. If $v_s \rightarrow 0$, $(\partial q / \partial z)_{z=z_v} \rightarrow 0$, and no transfer to the surface occurs. Thus v_s , which has the dimension of a velocity, may be viewed as the deposition velocity from level $z = z_v$ to the surface. Its inverse represents the resistance for transfer across to the surface.

$$r(z_v, 0) = v_s^{-1} \quad (2.10)$$

It should be stressed, however, that eq. (2.9) applies to the level $z = z_v$, if v_s is to express only the way the accommodation of the pollutant on the surface influences the transfer velocity or resistance.

The resistance for transfer of matter from the free atmosphere ($z = z_1$) to a natural surface may thus be looked upon as the sum of three parts:

(1) $r(z_1, z_0) = [v_M(z_1, z_0)]^{-1}$, i.e. the resistance down to the level z_0 , which is equal to the resistance for momentum transfer to the surface.

(2) $r(z_0, z_v) = (k u_*)^{-1} \ln (z_0 / z_v)$, i.e. the resistance for transfer of matter from the level z_0 to the surface in the case where the surface is a perfect sink. This resistance depends upon the compound being considered (i.e. molecular weight for a gas) and also depends on, for example, u_* (cf. Chamberlain, 1966).

(3) $r(z_v, 0) = v_s^{-1}$, which expresses the accommodation of the compound on the surface; $v_s \rightarrow \infty$ and $r(z_v, 0) \rightarrow 0$ in the case of a perfect sink.

Let us next consider the relative importance of these three resistances. Field measurements of air concentrations at about 2 m elevation and determination of the rate of deposition on the ground have yielded values of a transfer velocity for this whole layer varying from fractions of a cm sec^{-1} to several cm sec^{-1} . It is possible to compute the transfer velocity, v_M (or resistance) from the level of concentration measurements (z_1) to z_0 (or z_v) with the aid of eq. (2.5) provided z_0 (or z_v) and u_* or $u(z_1)$ are known. Eq. (2.5) is readily transformed with the aid of (2.3) into

$$v_M(z_1, z_0) = r(z_1, z_0)^{-1} = \frac{k^2 u(z_1)}{(\ln z_1 / z_0)^2} \quad (2.11)$$

Fig. 1 shows the values of v_M as a function of $u(z_1)$ and z_0 for the range of values that normally occur. It is obvious that *as long as the resistance for transfer from a few meters height to the earth's surface is less than about 1 cm sec⁻¹, by far the major part of the resistance is due to the final transfer through the viscous boundary layer surrounding the objects on to which deposition occurs.* This fact will be discussed further in the following sections.

In reality it is often difficult to determine from experiments to what extent the resistance for transfer from the atmosphere to a natural

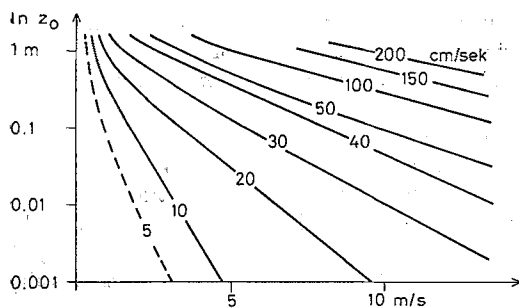


Fig. 1. Transfer velocity, $v_M(z_1, z_0)$ as a function of z_0 , roughness length, and $u(z_1)$, z_1 being assumed to be 2 m.

surface depends on (2) or (3) above. To understand the physical processes well such a distinction is important, even though for practical purposes we may simply try to determine the total resistance, or transfer velocity, from the level $z = z_0$ to the surface, i.e. $r(z_0, 0) = r(z_0, z_v) + r(z_v, 0)$. In analogy to (2.9) and (2.10) we may then define a transfer velocity v_s and have

$$r(z_0, 0) = (v_s')^{-1} \quad (2.12)$$

This may often be the only quantity that can be determined from observations.

The treatment above has been based on the assumption of neutral stratification. This is obviously often not the case in reality. The derivation of an expression for the resistance for transfer down to the level $z = z_0$ must then obviously include the dependence of the wind profile and thus the resistance on the heat flux. Even though the stratification plays no direct role in the transfer from the level $z = z_0$ to the surface, its magnitude depends on a proper determination of u_* in the layer above. At present we shall restrict ourselves to these few remarks.

The formalism given above should provide a reasonable basis for the formulation of experiments in the field to determine the rate of deposition. Measurements of SO_2 deposition are under way and will be reported separately. In the following sections we shall present some model computations that illustrate the relative importance of the different sink mechanisms as the emission height and the intensity of the turbulent processes are varied and apply these results in some specific cases of interest in the field of atmospheric chemistry.

3. Concentration distribution and transfer rates as dependent on source and sink distributions

3.1. Model assumptions

The combined effect of the processes as described in section 2 may be studied with the aid of the equation

$$\frac{\partial}{\partial z} \left(D \frac{\partial q}{\partial z} \right) + Q(z) - S(z) = 0 \quad (3.1)$$

where

$$\begin{cases} D = ku_* z & z_v \leq z \leq H \\ D = ku_* H & z > H \end{cases} \quad (3.2)$$

and the boundary condition

$$\begin{cases} v_s q = ku_* z \frac{\partial q}{\partial z} & z = z_v \\ q = 0 & z \rightarrow \infty \end{cases} \quad (3.3)$$

Here, $Q(z)$ and $S(z)$ denote the source and sink distributions as a function of the vertical (see below). The distribution of D as a function of the vertical at some distance from the surface is of course only described in a very approximate way by the expression (3.2). Particularly its dependence on the static stability has not been explicitly included, which is necessary when evaluating profile measurements. We may, however, vary the range of u_* and H to describe roughly the variations in reality. As mentioned before, z_v is often not known. We may then apply the lower boundary condition to the level $z = z_0$ and choose a value for the transfer velocity from z_0 to the surface, v_s' , that takes this circumstance into consideration.

Since we have reduced our problem to one dimension in space, we have implicitly assumed that horizontal variations are small in some sense. Obviously, horizontal transfer must be small compared with the vertical transfer. Since horizontal winds are 10^2 to 10^3 times the transfer velocities with which we are concerned, horizontal variations in concentrations over a distance of about 10 km should be smaller than the vertical variations over 10 to 100 m in the lowest part of the atmosphere.

The roughness length, z_0 , may vary from about 10^{-4} m for sand, ice and snow surfaces to about 1 m for forests and cities, etc. Cham-

berlain (1966) determined z_v for transfer of thorium and water vapour to grass-like surfaces and sand. He found generally that z_v is 0.1 to 0.3 times z_0 and that z_v decreases with increasing stress, i.e. u_* . It is unclear what the concept z_v means in a forest (or a city) and no measurements are available to clarify the difference in resistance for momentum transfer and transfer of matter under such conditions. One might expect, however, that if such resistance were to be formally expressed by a roughness length for mass transfer, z_v , we would have $z_v < z_0$ at least to the extent that this is so for grass-like surfaces. Measurements to clarify this are desirable.

The friction velocity, u_* , varies from values less than 0.1 m/s for light winds to a few m/s for strong winds. Thus we shall let $D_v = ku_* z_v$ vary between $10^{-5} \text{ m}^2 \text{ s}^{-1}$ and $10^{-1} \text{ m}^2 \text{ s}^{-1}$, and $D_H = ku_* H$ between 1 and $10 \text{ m}^2 \text{ s}^{-1}$.

The source $Q(z)$ should of course be distributed homogeneously in the horizontal plane to permit the simplification to one space dimension. If concerned with the transfer of ozone from the stratosphere and troposphere for destruction at the ground, or the production and vertical transfer of sea salt nuclei over the sea, such an assumption is plausible. It becomes more questionable when we are concerned with anthropogenic sources, the distribution of which is often far from homogeneous. The following results may only be considered as indicative of problems of that kind. To simplify the following discussion we shall assume that $Q(z)$ is a (continuous) point source at the level h of intensity Q_0 , i.e.

$$\int_{h-\delta}^{h+\delta} Q(z) dz = Q_0 \quad (3.4)$$

where 2δ is a small height interval, i.e. $\delta \ll h$.

In the following we shall limit ourselves to a consideration of the relative importance of the two major sink mechanisms, i.e. rain-out by precipitation, and dry deposition. The latter is taken care of by the lower boundary condition as formulated above and in the preceding section. We shall let v_s , as an expression for the surface accommodation, vary between zero and infinity to include both the case of the surface being a perfect sink and the opposite extreme when rain-out represents the only sink mechanism. Of course, the rate of rain-out varies

widely according to climatic conditions. We shall here use the results of Rodhe & Grandell (1972) and assign a characteristic rate of elimination of matter, τ_0 , above the level H , that is equivalent to a residence time of $3 \times 10^5 \text{ sec}$, which corresponds to about 83 hours.

The solution of eq. (3.1) for various values of the parameters u_* , H , z_0 (or z_v), v'_s (or v_s) and h yields a set of concentration distributions $q(z)$.

If we let these parameters take on all possible values, i.e.

u_*	0.01 ms ⁻¹ – 1.0 ms ⁻¹
z_0 (or z_v)	0.001 m – 1.0 m
v'_s (or v_s)	0 – ∞
h	0.1 m – 300 m

a great variety of distributions $q(z)$ may be obtained. It is not very helpful to present all such possibilities, but a few will be given, that illustrate the most important dependence of $q(z)$ on some of these parameters. It is in this context preferable to use $u(z_1)$ (z_1 , being the level at which concentration measurements are usually performed) as a parameter rather than u_* . $u(z_1)$ is uniquely determined from a specification of z_0 and u_* in accordance with eq. (2.3). We shall use the values $u_1 = 1 \text{ m/s}$ (light winds), $u_1 = 10 \text{ m/s}$ moderate to strong winds in our computations.

As was stressed in the introduction the residence time of a pollutant in the atmosphere is of basic importance in order to assess the horizontal dispersion and the characteristic deposition pattern. It is therefore of interest to summarize our results also with regard to how the different parameters determine the residence time. Again particular attention will be given to the determination of the relative importance of the two major sink mechanisms, rain-out and dry deposition.

4. Results

4.1. Concentration distributions

We first compare the vertical distribution of the concentration, when emission occurs at 20 m and 300 m, assuming the wind at the 2 m elevation, u_1 , to be 1, 4 and 10 m/s and the deposition velocity to vary between 0 and ∞ while the roughness length is kept constant, $z_0 = 10^{-1} \text{ m}$ (Fig. 2).

We note the marked difference in the con-

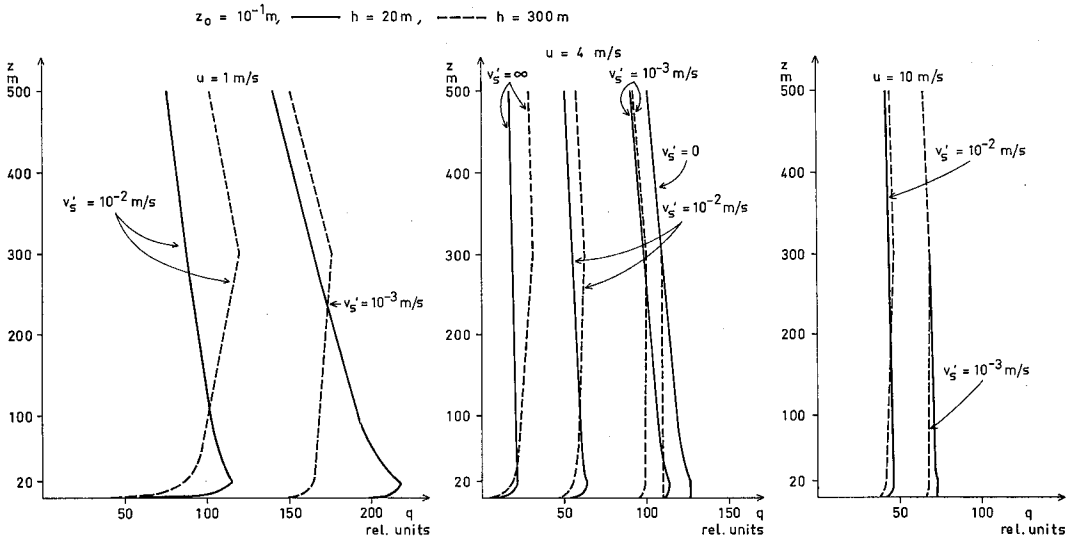


Fig. 2. Concentration of pollutant as a function of elevation, when emission occurs at 20 m (solid lines) and 300 m (dashed lines) and a roughness length (z_0) of 10 cm. The transfer velocity (v'_s) is assumed to be 0, 10^{-3} , 10^{-2} m s $^{-1}$ and the wind velocity at 2 m, 1 m s $^{-1}$, 4 m s $^{-1}$ and 10 m s $^{-1}$. Mean residence time before rain-out is 83 h .

centration values obtained as a function of wind velocity. For high winds (c) the vertical transfer both to the surface and to the levels aloft,

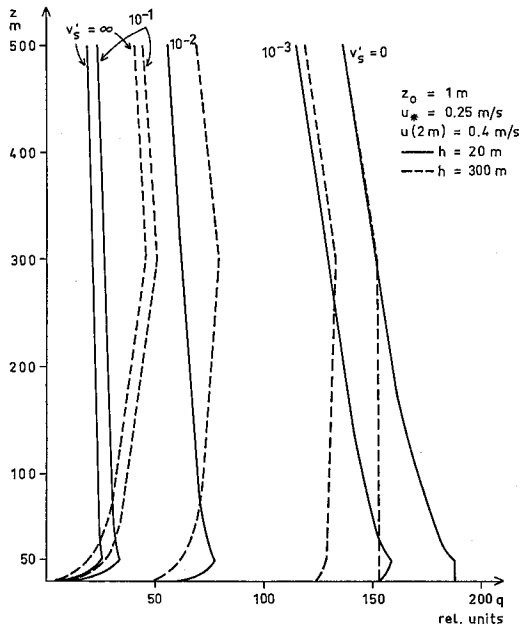


Fig. 3. Concentration of pollutant as a function of elevation (compare Fig. 1) for a very rough surface ($z_0 = 1$ m) and weak winds.

where rain-out takes place, is effective and reduces the turnover time very much in comparison with what is the case, when the wind is weak. Also the value of the deposition velocity, v'_s , has a pronounced influence on the concentrations observed. We also note a change in the vertical profile as we approach the surface of the earth. When $v'_s \rightarrow \infty$, the value of $q(0) \rightarrow 0$ and a marked vertical gradient is observed close to the surface. For more realistic values, 10^{-2} or 10^{-3} m s $^{-1}$, this gradient is quite weak and thus difficult to observe if the wind is moderate or strong but clearly distinguishable when the wind is weak. This is more clearly shown in Fig. 3, where a very large value of $z_0 = 1$ m has been chosen, corresponding to a forest, and a wind velocity $u(2\text{ m}) = 0.4$ m s $^{-1}$. As long as the transfer velocity v'_s is larger than a few mm s $^{-1}$ a vertical gradient is noticeable in the lowest layers of the atmosphere.

4.2. Residence times

As indicated before, it is of particular interest to deduce the dependence of the residence time of an atmospheric pollutant on the parameters at disposal in our computations. If we keep $u_* = 0.25$ m s $^{-1}$ constant, but let v'_s and z_0 (and thus correspondingly $u(z_1)$ according to (2.3))

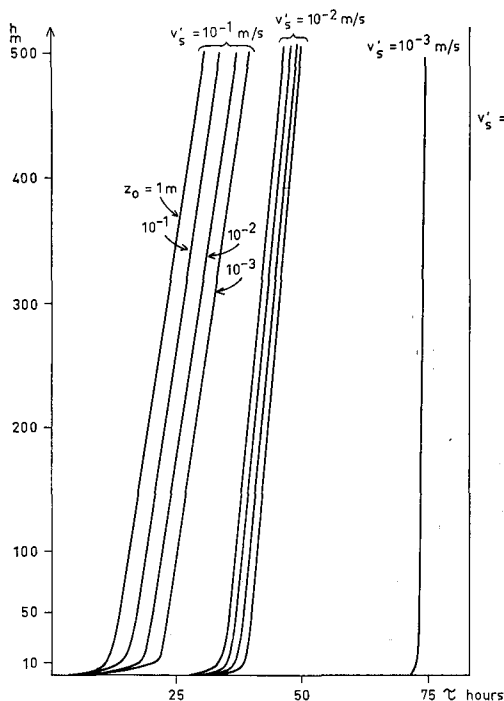


Fig. 4.

vary, we find that the residence time τ_0 depends on the level of emission, as shown in Fig. 4. For values of $v'_s = 10^{-2} \text{ m s}^{-1}$, which is probably a typical value over land for many pollutants, τ_0 is about half the value obtained in the case $v'_s = 0$ in which case rain-out is the only removal mechanism. As the emission height decreases, the residence time decreases, due to greater dry

Fig. 4. Residence time as function of height of emission for various assumptions regarding the deposition velocity v'_s ($v'_s = 0, 10^{-3}, 10^{-2}, 10^{-1} \text{ m s}^{-1}$) and roughness length of the earth's surface ($z_0 = 10^{-3}, 10^{-2}, 10^{-1}$ and 1 m). Residence time in case of no dry deposition is assumed to be 83^h .

Fig. 5. Residence time as function of deposition velocity (v'_s) and roughness length of the earth's surface (z_0) for various values of wind velocity at 2 m elevation (1, 4 and 10 m/s). Height of emission is $h = 300 \text{ m}$ and the thickness H of the layer of increasing eddy diffusivity according to eq. (3.2) is 100 m . Residence time in case of no dry deposition is 83^h . According to (2.3) and (3.2) the values for z_0 correspond to values of D at levels $z \geq H$, which are also given along the vertical coordinate.

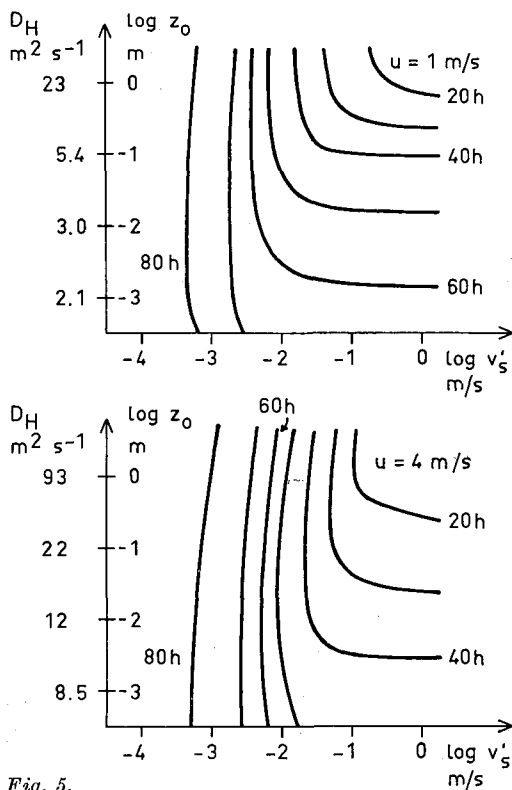
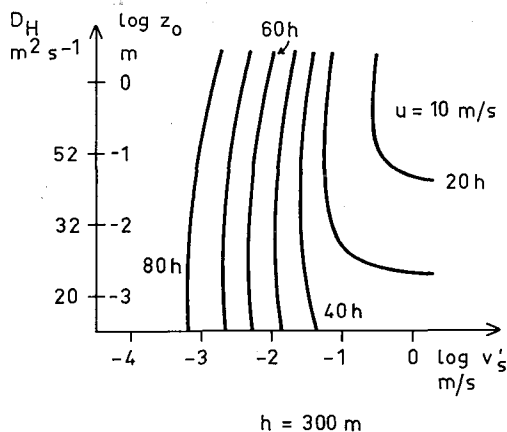


Fig. 5.



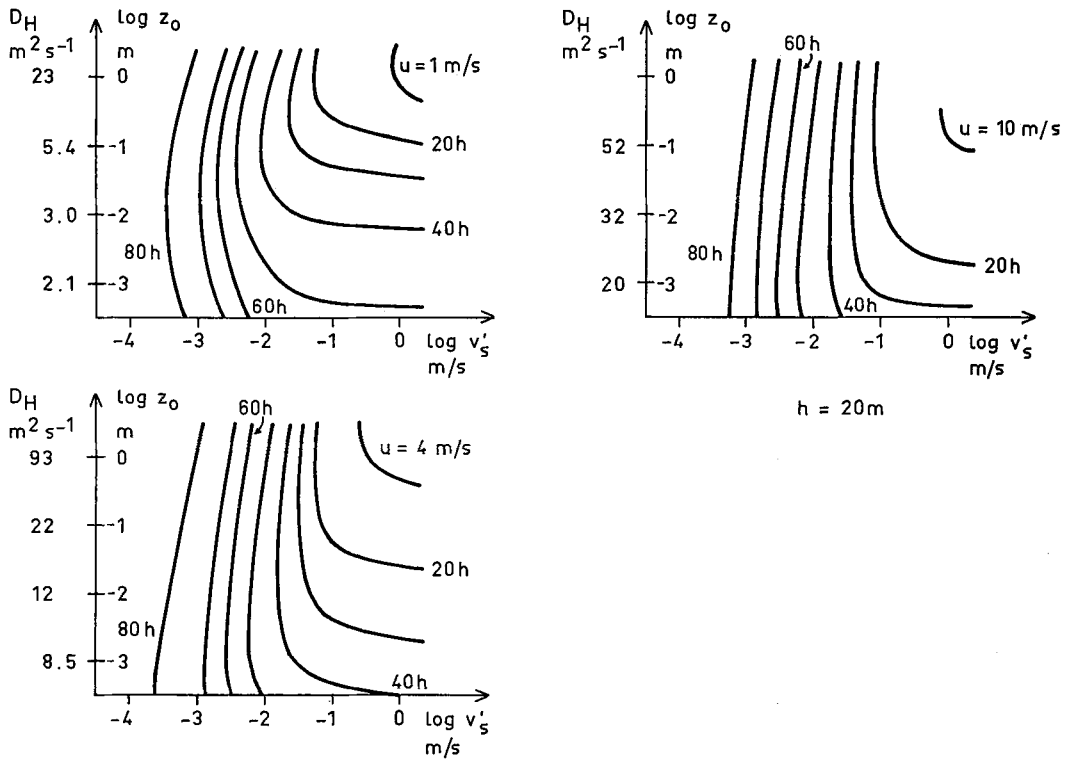


Fig. 6. The same as Fig. 5, but for an emission height of $h = 20 \text{ m}$.

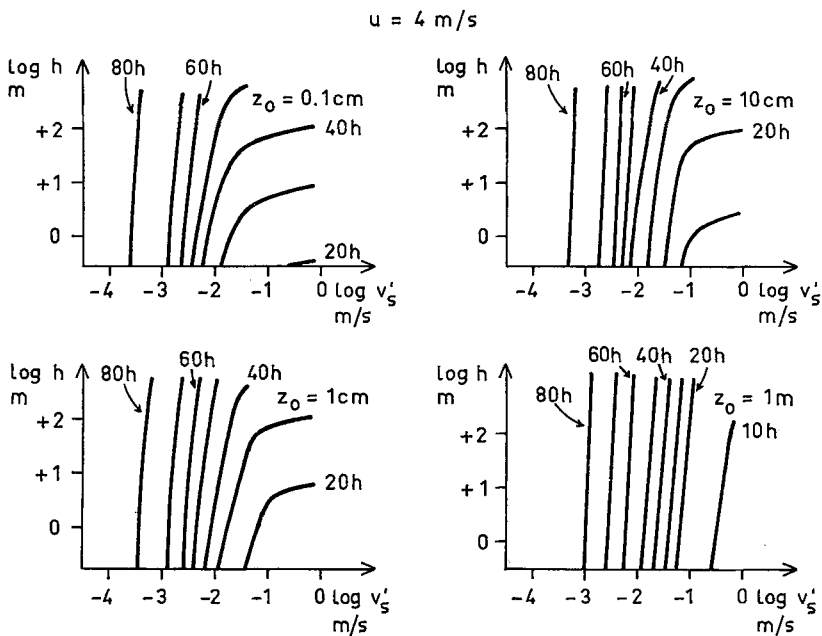


Fig. 7. Residence time as function of deposition velocity (v'_s) and height of emission (h) for various values of the roughness length and for a wind velocity of 4 m s^{-1} at 2 m elevation. See further Fig. 5.

deposition, but a rapid decrease does not occur until $h < 10$ m. Furthermore, we see that the result is rather independent of the value of the roughness length z_0 .

We summarize in Figs. 5, 6 and 7 the way the residence time depends on the decisive parameters: height of emission, h , roughness length, z_0 , wind velocity at 2 m elevation, u (2 m) and deposition velocity, v'_s .

4.3. Discussion

It is interesting to note that the value for the residence time is closely dependent both on the character of the earth's surface (z_0 and v'_s) and the meteorological conditions. If $h < 10$ –20 m the height of emission is also an important parameter but less so for large values of h . The wind velocity seems to be of least importance, particularly when the level of emissions is above the lowest tens of meters. Obviously the emissions from an elevated source are transported both upwards and downwards in proportions which are rather independent of the wind speed.

Since the deposition velocities for many pollutants and surfaces are probably not much larger than 10^{-2} m s $^{-1}$ the results of the computations indicate that dry deposition is not normally likely to be more important than rain-out, but it may well be just as important.

It is of course not possible to proceed further without measurements aiming at a better determination of the deposition velocity for different pollutants and surfaces. In such investigations the present restriction of the theoretical de-

ductions to a neutrally stratified boundary layer must be removed, which should cause no serious problems, provided simultaneous measurements of temperature, wind, and pollutant profiles are made.

Similar considerations to those presented above have been given by Fabian & Junge (1970) in connection with their discussion of ozone destruction at the earth's surface. Their aim was a more limited one, however, and of course no sinks due to rain-out and wash-out were included. Also the manner of presentation of the results as given here might be more informative and permit their use in a qualitative discussion of the relative efficiency of sink mechanisms for the removal of different pollutants from the atmosphere. It is clear, on the other hand, that a more precise discussion hardly can be carried out without much more careful consideration of the three-dimensional dispersion patterns in the atmosphere and also the geographical distribution of sources and sinks in the area under consideration.

Acknowledgement

This research has been sponsored partly by the Swedish Science Research Council (Contract No. 0223-050) and partly by the Research Committee of the National Swedish Environment Protection Board (Contract No. 7-126/73).

Our thanks are due to Dr Henning Rodhe, who took an active part in the early phase of the work reported here.

REFERENCES

- Bolin, B. & Rodhe, H. 1973. A note on the concepts of age distribution and transit time in natural reservoirs. *Tellus* 25, 58–62.
- Brosset, C. 1973. Air-borne acid. *Ambio* 2 (1–2), 2–9.
- Chamberlain, A. C. 1966. Transport of gases to and from grass and grasslike surfaces. *Proc. Roy. Soc., Ser. A*, 290, 236–265.
- Fabian, P. & Junge, Chr. E. 1970. Global rate of ozone destruction at the earth's surface. *Arch. Met. Geoph. Biokl., Ser. A* 19, 161–172.
- Mason, B. J. 1971. *The physics of clouds*. Clarendon Press, Oxford.
- Monin, A. S. 1970. The atmospheric boundary layer. *Ann. Rev. Fluid Mechanics* 2, 225–250.
- Munn, R. E. & Bolin, B. 1971. Global air pollution—meteorological aspects. A survey. *Atmospheric Environment* 5, 363–402.
- Rodhe, H. & Grandell, J. 1972. On the removal time of aerosol particles from the atmosphere by precipitation scavenging. *Tellus* 24, 442–454.
- Sehmel, G. A. 1970. Particle deposition from turbulent air flow. *J. Geophys. Res.* 75, 1766–1781.
- Sweden's Case study for the United Nations Conference on the human environment 1971: Air pollution across national boundaries. The impact on the environment of sulfur in air and precipitation. Royal Ministry for foreign affairs, Stockholm.

ВРЕМЯ ЖИЗНИ АТМОСФЕРНЫХ ПРИМЕСЕЙ В ЗАВИСИМОСТИ ОТ
ХАРАКТЕРИСТИК ИСТОЧНИКА; АТМОСФЕРНАЯ ДИФФУЗИЯ И МЕХАНИЗМ СТОКА

Кратко обсуждаются механизмы стока атмосферной примеси, причем особое внимание уделяется сравнительному значению процессов вымывания и сухого отложения. Процессы турбулентного переноса, ответственные за сухое отложение, анализируются в свете классической теории атмосферного приземного пограничного слоя. Скорости переноса в свободную атмосферу (где механизмом стока является вымывание) и на поверхность

Земли (где имеет место сухое отложение) вычисляются в зависимости от высоты эмиссии, шероховатости земной поверхности, скорости выпадения (или эффективности поглощения) на поверхности земли и интенсивности турбулентных процессов (зависящих от скорости ветра). На базе этих вычислений находится время жизни атмосферной примеси. Статья заключается дискуссией о сравнительном значении вымывания и сухого отложения.