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THE CONCENTRATION OF PLUTONIUM IN HAIR FOLLOWING INTRAVENOUS INJECTION*

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Abstract—The concentration of plutonium has been determined along the length of the hair of a female subject who received 11 kBq (0.3 μCi) of ^{239}Pu by intravenous injection in 1945. The subject succumbed to her pre-existing illnesses 518 days post injection, and her remains were exhumed in 1973.

The subject's hair, 280 mm in length for the longest strands, was divided into 20-mm long sections and the plutonium concentration in each was determined. The concentration ranged from 22 Bq/kg (0.59 pCi/g) at the distal end to 3 Bq/kg (0.08 pCi/g) at the proximal end (nearest the scalp). The distance of each section from the scalp was then converted to an estimated time in days post injection by applying the growth rate of the hair indicated for this individual.

The plutonium concentration as a function of time could be well-fitted (in the least-squares sense) by either of two functions. The first function was the sum of a single exponential, whose half-time was 81 ± 16 days, plus a constant term. This half-time corresponded to that found for the longest-lived of five components of the plutonium concentration in the blood of other individuals in the same series of injections, i.e. 88 ± 13 days (Du72). The second function 'which fitted the hair data was the sum of two exponentials, whose half-times were 32 ± 18 and 260 ± 80 days, respectively. These values overlap the half-times of 42 and 300 days found for components of the urinary excretion of plutonium by man (Du72). This agreement is expected in view of the correspondence of the hair and blood data, since urine is a plasma filtrate. The possible use of hair as a bioassay material for plutonium in man is suggested.

INTRODUCTION

THE CONTENT and macrodistribution of plutonium have been determined in the remains of a young woman who received 11 kBq (0.3 μCi) of ^{239}Pu (IV) citrate by intravenous injection as part of an excretion study in 1945. This subject, Center for Human Radiobiology case number 40–010, was designated Hp-4 in the early literature (La50). At the time of injection she was suffering from Cushing's disease with hypertension, osteoporosis and nephropathy with uremia. She succumbed to her illnesses 518 days post injection.

Her remains were exhumed by the Center for Human Radiobiology in 1973 with the permission of the next of kin. After selected samples were analyzed for plutonium content and distribution, the remains were re-interred. The results for the skeleton and selected soft tissues have been reported elsewhere (La78); the skeleton contained 54 ± 2% of the injected amount. Because an autopsy had been performed prior to interment, only a few types of soft tissue were available for analysis, and the concentrations

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of plutonium in them were about 100 times lower than those in bone.

The subject's hair, 280 mm in length for its longest strands, appeared to be intact and in good condition. Since the analysis of trace elements in hair is one method of determining exposure to toxic materials (e.g. Ja78), it was decided to measure the plutonium concentration along the length of the hair, and to compare the results with the concentrations of plutonium in blood and urine observed in the injection cases (La50; Du71; Du72).

CHEMICAL ANALYSIS

The subject's hair was removed from the remains with the scalp intact. It was dried at 100°C for 48 hr and then cut away from the scalp as closely as possible. The hair was washed with a detergent solution until microscopic examination showed it to be free of foreign matter; it was then rinsed with water and air-dried. Because of the presence of small bits of tissue in the wash solution it was impossible to determine if any plutonium in the solution had been leached from the hair. However, there is no reason to suspect that the mild detergent used and distilled water could in fact have leached plutonium from the hair strands. The longest strands measured 280 mm and we defined the parameter d as the distance of any portion of the hair from the scalp. The hair strands were cut into 20-mm long sections, beginning at the proximal ends. Sections with the same value of d were combined, weighed and then analyzed as a single sample.

The plutonium content of each section was determined by α-spectrometric isotope dilution. The hair samples were dissolved in nitric acid, a known amount of ^{242}Pu and an excess of 9 M hydrobromic acid were then added and the solution was evaporated to incipient dryness. The procedure ensured isotopic exchange between the ^{239}Pu that was in the hair and the isotopic diluent, ^{242}Pu. The plutonium was separated from the solution with the anion exchange procedure of Larsen and Oldham (La75); it was electrodeposited on a stainless steel planchet with the technique reported by Kressin (Kr77), and the deposit was assayed in an α-spectrometer. The plutonium recovery ranged from 70 to 90%; the resolution (FWHM) of the ^{239}Pu and ^{242}Pu peaks in the series ranged from approx. 10 to 13 fJ (60 to 80 keV).

The results are given in Table 1. The plutonium concentration is expressed as becquerels per kg of hair (1 Bq/kg = 0.027 pCi/g). The plutonium concentration shows a steady increase with distance from the scalp, except for a few fluctuations which are not statically significant, and for the data point nearest the scalp itself. The concentration of plutonium in the scalp was determined to be 210 Bq/kg. The very large value of plutonium concentration in the hair section nearest the scalp may be due either to cross-contamination during the drying and removal of the hair from the scalp, or possibly to adsorption during the 26 yr of interment. Since there is no known physiological reason for the large value, this data point has been omitted from the subsequent analysis.

It must be noted that our choice of d as a parameter requires the assumption that the growth rate of the hair was uniform over the entire scalp. There does not, however, appear to be any smearing of the data as would be expected if the combined sections contained hair that was formed at different times. Consequently the assumption of uniform growth rate over the scalp seems justified.

Table 1. The concentration of plutonium in the sections of the hair of case 40-010. As described in the text, d is the distance from the scalp to the middle of the section and t is the estimated time of formation of the section in days post injection; m is the mass of the section and y is the observed concentration of plutonium in the section

d, mm	t, days	m, grams	y, Bq/kg
10	499	1.35	16.8±0.7
30	462	0.91	3.5±0.2
50	425	0.82	2.9±0.2
70	388	0.74	3.0±0.2
90	351	0.59	5.3±0.4
110	314	0.57	4.2±0.3
130	277	0.48	5.3±0.4
150	240	0.55	5.5±0.3
170	203	0.35	6.6±0.4
190	166	0.26	6.1±0.4
210	129	0.22	8.0±0.5
230	92	0.09	12.1±1.2
250	55	0.02	13.0±1.3
270	18	0.02	21.5±1.9

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5.5±0.3
6.6±0.4
6.1±0.4
8.0±0.5
2.1±1.2
3.0±1.3
1.5±1.9

MATHEMATICAL ANALYSIS

As the first step in relating the concentration of plutonium in hair to that in the blood, we must convert d, a distance parameter, to a time parameter. This requires knowledge of the growth rate of the hair. The average growth rate of hair for females given in ICRP publication 23, Reference Man, is 0.36 mm/day (ICRP75); at this rate, 280 mm would grow in 775 days. However, plutonium was found in the most distal section (d = 260–280 mm) of this subject's hair, which implies that at least some of that section was formed after injection. If we assume that the distal end of that section represents a time close to the injection, then an average growth rate of 280 mm/518 days, or 0.54 mm/d is indicated for this individual. Such a rapid growth rate is consistent with the medical status of this patient, since hypertrichosis is known to be among the consequences of Cushing's disease (Me72).

It should be noted that 0.54 mm/d is a minimum growth rate, since there is no way of knowing if some hair containing plutonium had been cut off before death. It seems unlikely, however, that a significant length of plutonium-laden hair would have grown out from the scalp fast enough to be included in any portion cut. On the other hand, since the hair was divided into 20-mm long sections, it may be possible that there was no plutonium in the most distal few mm, implying that the growth rate could have been a little less than 0.54 mm/d. In the absence of any evidence to the contrary, the hair growth rate for this case is assumed to be 0.54 mm/d. Again, the results of subsequent analyses appear to support this assumption. The estimated time of formation, t, for each section of the hair is then equal to $(280\text{-}d)/0.54$, and is given in column 2 of Table 1.

The next step is relating the plutonium in hair to that in the blood is to obtain a mathematical description of the plutonium concentration in hair as a function of time. Several functions were fitted to the data by the method of weighted least squares, and two of them were reasonable fits. The first was the sum of an exponential and a constant, and the second was the sum of two exponentials. The functions are given below as equations (1) and (2), respectively, where y is the concentration of plutonium in the hair in Bq/kg, and t is the time in days post injection:

$$y = (18.2 \pm 2.8)\exp[-0.693t/(81 \pm 16)] + (2.9 \pm 0.05) \quad (1)$$

and

$$y = (17.5 \pm 7.2)\exp[-0.693t/(32 \pm 18)] + (10.1 \pm 2.7)\exp[-0.693t/(260 \pm 80)]. \quad (2)$$

One S. E. is shown for each parameter. The two functions are plotted with the data in Fig. 1. There is little to choose in terms of goodness-of-fit criteria between equations (1) and (2). For the former, $\chi^2 = 15.9$ with 10 degrees of freedom, while for the latter, $\chi^2 = 13.2$ with 9 degrees of freedom. Either function can be interpreted in terms of other physiological data from the injection cases, as discussed in the following sections.

COMPARISON WITH BLOOD DATA

Durbin has analyzed the plutonium content of the blood as a function of time post injection for eleven of the subjects in

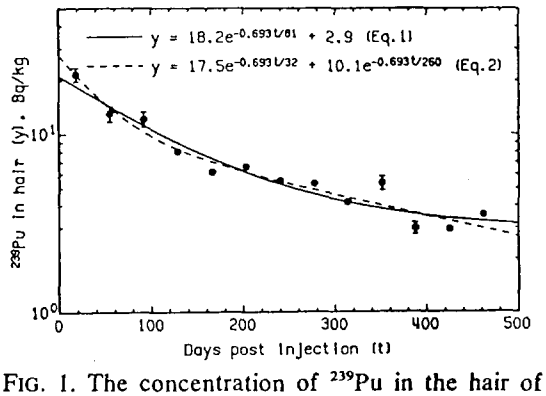

FIG. 1. The concentration of ^{239}Pu in the hair of case 40-010, as a function of time post injection. Errorbars are ommited where they lie within the size of the symbols. The curves are the two functions fitted to the hair data, as described in the text. For each function, y is the concentration of plutonium in hair (Bq/kg) and t is the time post injection in days.

1000180

Langham's study (Du71; Du72; La50). She found that the plutonium in the blood of five subjects with normal circulation exhibited five exponential components whose intercepts and half-times are shown in Table 2. The blood data for case 40–010 (Hp–4) given in (La50) cannot be analyzed into five components because there are not enough values at either early (<1 day) or late (>23 days) times post injection. However, the data can be fitted by a three-component exponential, whose parameters are shown in parentheses in Table 2. Note that the half-time of the longest-lived component observed in the blood of 40–010 (5.5 days) is quite comparable to BT_4 for the normal cases (5.0 days).

In Durbin's analysis, those cases with circulatory impairments, including 40–010, demonstrated higher intercepts and longer half-times for the early ($T_{1/2}<2$ days) components in the blood, but were not significantly different from the normal cases for the longer-lived components. Thus we may assume that case 40–010 would also have exhibited another blood component with a half-time comparable to BT_5, the longest-lived component of the normal blood curve, equal to 88 ± 13 days.

The exponential term of equation (1) above exhibits a half-time of 81 ± 16 days, in substantial agreement with BT_5. The constant term in equation (1) may then be interpreted as either an equilibrium value or as another exponential with a much longer half-time. It must be borne in mind, however, that Durbin derived the value of BT_5 from only two cases, Hp-3, whose last blood sample was taken 23 days post injection, and Hp-12, whose last blood sample was taken 46 days post injection (Du71). Consequently, this component is less well established than the others. Nevertheless, the hair and blood data appear to be mutually consistent, and it may be concluded that the concentration of plutonium in the hair reflects that in the blood, as a function of time.

COMPARISON WITH URINE DATA

Durbin also analyzed the urinary excretion of plutonium by the injection cases (Du71; Du72), and found that a five-component exponential function provided a better fit to the data than did the power function of Langham's equation (La50). The intercepts (percent injected dose excreted per day) and half-times of the five components for those subjects with normal kidney function are also given in Table 2.

The nephropathy suffered by case 40-010 did not appear to have much influence on the

Table 2. The parameters of the blood and the daily urinary excretion curves derived by Durbin for the injection cases (Du71). Each curve has five exponential components; the intercepts are denoted by S (per cent of injected dose) and the half-times by T (days). The parameters of the curves fitted to the actual blood and urine data of case 40-010 (Hp-4 in La50) are shown in parentheses

Component	Blood curve		Urine curve	
i	BS_i(%)	BT_i(days)	US_i(%)	UT_i(days)
1	52.4 (7.0)	0.0139(0.0333)	0.41 (0.50)	1.23(1.06)
2	27.1 (60.4)	0.304 (0.868)	0.12 (0.20)	5.5 (7.0)
3	17.2 (13.0)	1.2 (5.5)	0.013	42
4	3.3	5.0	0.003	300
5	0.44	88	0.0012	4000

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intercepts or half-times of the two components observed in her urinary excretion data (shown in parentheses in Table 2), compared with the same components in cases with normal kidney function (Du71). Therefore her later components, even though not observed in the urine data of 40-010 (last sample collected on day 27 post injection), would also be expected to agree with those for the normal cases in Table 2.

The half-times of the two components fitted to the hair data (equation 2), equal to 32 ± 18 and 260 ± 80 days, overlap UT_3 and UT_4, namely 42 and 300 days, within error. This agreement between the time dependences of the hair and urine data is not surprising in view of the correspondence betwen the hair and blood data, since urine is a plasma filtrate.

However, the relative magnitudes of the two components are quite different in the hair data vs the urine curve. From equation (2), the ratio of the intercepts is 1.7 ± 0.8, while from Table 2, $US_3/US_4 = 4.33$. This disagreement may possibly be explained by the medical status of this individual, involving both nephropathy and hypertrichosis. Since urine and hair are both routes of excretion for plutonium in the blood, the time dependence of each should reflect the time dependence of the blood content, as observed. The relative efficiency of each in removing plutonium from the blood may well have changed in time in this individual, as her disease progressed. Thus the relative magnitude of the two components could vary with excretion route, while the time dependence remains the same.

DISCUSSION

It should be noted that the two major assumptions involved in the analysis of the hair data, namely that the growth rate was constant over the scalp and that the distal portion of the longest hair was formed close to the time of injection, are both supported by the consistency of the hair data with the blood and urine data.

The correspondence of the hair and blood data is further strengthened by consideration of the half-times in equation 2. The value of BT_5, 88 ± 13 days, is presumably a composite of UT_3 and UT_4. If the sum of these components of the urinary excretion curve is analyzed as a single exponential from 0 to 300 days post injection, it exhibits a half-time of 92 days, essentially identical to BT_5. This analysis of two components as one explains the fact that equation (2) is a slightly better fit to the hair data than is equation (1). The constant term in equation (1) then, which does not correspond directly to any component of the blood curve, very probably reflects the appearance of that component of the plutonium concentration in the blood which gives rise to the fifth component of the urine curve, with $UT_5 = 4000$ days.

If we pursue the correspondence between equation (1) and the fifth component of the normal blood curve, the concentration ratio of plutonium in hair to plutonium in blood can be determined by comparing the intercepts of the exponentials. Langham assumed that the total mass of blood was equal to 7.7% of the body mass (La50). For this subject, with a body mass of 55.5 kg, the initial concentration in blood would have been 11 kBq per 4.28 kg of blood, or 2.5 kBq/kg. The concentration for the longest-lived component (BS_5) would have been 0.44% of this, or 11.3 Bq/kg. The fitted concentration in hair was 18.2 Bq/kg, and therefore, the concentration ratio was 1.6.

The concentration ratio between the hair and urine can be determined by comparing the intercepts of equation (2) and US_3 and US_4. $US_3 = 0.013\%$ of the injected dose per day, or 1.43 Bq/day. If the average urinary output is taken as 1000 ml/d with a specific gravity of 1.02 (ICRP75), then the concentration of plutonium in urine was 1.4 Bq/kg. Since the intercept of the first component of equation (2) is 17.5 Bq/kg, the concentration ratio is 12.5. Similarly, for the second component of equation (2) and US_4 the concentration ratio is 31.2. As mentioned above, this change in concentration ratio may be attributed to the illnesses of this subject.

Regardless of the discrepancies in the above analysis, it nevertheless raises the intriguing possibility that human hair may serve as a bioassay material for the determination

1000182

roughness length. It is typically of the order of 1 cm but can be as small as 1 mm over the sea and as large as 1 m above cities and forests.

To integrate (1) and (2) under diabatic conditions ($z/L \neq 0$), the specific form of the ϕ-functions must be known, but it is always permissible to write $\phi \equiv 1 - z\partial\psi/\partial z$, so the solution can still be expressed as a logarithmic function, but this time with the addition of a diabatic correction. For velocity the result is

$$\bar{u} = \frac{u_*}{k}\left(\ln\frac{z}{z_0} - \psi_M\left(\frac{z}{L}\right) + \psi_M\left(\frac{z_0}{L}\right)\right). \quad (4)$$

The ψ-functions have been determined experimentally (see, e.g. Bu73). The last term is always disregarded in practice.

In the same way as (1) and (2), the surface layer concentration gradient of a particular constituent with a stationary horizontally homogeneous distribution is given by

$$\frac{\partial\bar{\chi}}{\partial z} = \frac{\chi_*}{kz}\phi_c\left(\frac{z}{L}\right), \quad (5)$$

where $\bar{\chi}$ is the average concentration at height z, and χ_* is a characteristic scale for turbulent concentration fluctuations. The definition of χ_* is not abritrary; $-\chi_* u_*$ is the vertical flux of the constituent. As above, ϕ_c corrects for diabatic influence. It is generally believed (see, e.g. Mo71, p. 442) that $\phi_c \simeq \phi_H$, both of which are slightly different from ϕ_M for negative values of z/L (unstable conditions). Integration of (5)

$$\bar{\chi} - \chi_0 = \frac{\chi_*}{k}\left(\ln\frac{z}{z_{0c}} - \psi_H\left(\frac{z}{L}\right)\right). \quad (6)$$

Thus $\bar{\chi}$ under these circumstances is a logarithmic function of height, χ_o is the surface concentration (at $z = z_{oc}$), and z_{oc} is of similar magnitude as the roughness length but not necessarily the same. However, $\ln(z/z_{oc})$ can always be written $\ln(z/z_0) + \ln(z_0/z_{0c})$, from which it is seen that the first term dominates at sufficient height of observation, so that differences in z_o and z_{oc} become less significant. In the following we shall assume that this is the case.

The assumption of horizontal homogeneity leading to (6) is also valid for the main part of the deposition taking place from a single plume, except for narrow zones around the plume's interception with the surface, and along the edges of the plume. Using the assumption that local horizontal homogeneity exists if $\partial\bar{\chi}/\partial z \ll \partial\bar{\chi}/\partial x$, where x is a coordinate in a horizontal direction, and furthermore that a typical horizontal concentration gradient is $\bar{\chi}/\sigma_y$, where σ_y is the cross wind plume dimension, it can be shown from a formula given in the next section that $\sigma_y \gg z(\bar{u}/u_*)$ is sufficient condition for horizontal homogenity. Here z is the height at which the deposition velocity is defined.

3. THE DEPOSITION VELOCITY

The deposition velocity v_d is defined by $D = -v_x\bar{\chi}$, where D is the deposition flux (deposited amount per unit time per unit surface area). In the constant flux layer we also have $D = -u_*\chi_*$. Consequently we have from (6)

$$v_d = \frac{ku_*}{\ln\frac{z}{z_0} - \psi_H + k\frac{\chi_0}{\chi^*}}, \quad (7)$$

from which it is seen that v_d, especially close to the ground, is a strong function of height. The term $k\chi_o/\chi_*$ is due to the fact that the surface cannot always absorb everything transported downwards by turbulent diffusion. For fixed values of z and ψ_H, it is seen that v_d is at its maximum for $\chi_0 = 0$ which corresponds to a surface that is a perfect absorber. In this case $\bar{\chi} \to 0$ as $z \to z_0$ with the consequence that $v_d \to \infty$ since the flux is constant. A perfect absorber can, for example, be realized in the case we consider the diffusion of a gas that is highly soluble in the surface medium. The other extreme is realized for $\chi_* = 0$ (no flux), in which case $v_d = 0$. In practice, especially with regard to aerosols, which are most relevant in connection with reactor accidnets, it is very conservative to assume $\chi_0 = 0$. Thus, assuming $\phi_H \sim \phi_M$ we obtain from (7) with use of (4)

1000183

the formula of Chamberlain (Ch53)

$$v_d < u\left(\frac{u_*}{\bar{u}}\right). \tag{8}$$

For typical values ($z_0 = 1$ cm, $\bar{u} = 5$ m/sec, $z = 10$ m) we have from (3) that $u_* \simeq 0.3$ m/sec. According to (8) this means that v_d would be 1.8 cm/sec at the most under these circumstances. In practice, with $\chi_0 \neq 0$ the deposition velocity would be considerably less which is seen by referring back to (7). For other sets of (z_o, u, z) one would of course obtain other limiting values of v_d. But for very small heights (~ 1 m) and very large values of z_o (~ 1 m), one is forced to consider complications arising from $z_o/z_{oc} \neq 1$ and $\chi_0 \neq 0$, which then limits the increasing trend in v_d.

At a given height, with a given constituent concentration, the deposition velocity is also a function of stability. For positive values of z/L, ψ_H is given approximately by $-5z/L$. The above example, with $z_o = 1$ cm, $\bar{u} = 5$ m/sec, and $z = 10$ m, would, with a value of $L = 10$ m, correspond moderately stable conditions (typically nighttime, clear skies). This results in a value of u_* that is only about half of the neutral value and a maximum deposition velocity less than a quarter of the above value of 1.8 cm/sec.

It has now been demonstrated that v_d is a highly variable parameter, which is a well known fact that has been stated often in the literature (We77). Even when the surface plays no role ($\chi_o = 0$), diurnal stability changes cause variability in v_d.

The dependence of height can be minimized by choosing a sufficiently large reference height for the definition of v_d. As the concentration profile is logarithmic, most of the variation takes place near the ground. At a height of 200 z_o (typically 2 m) it would make only a 25% difference in v_d to take 1000 z_0 (typically 10 m) as a reference height. But close to the surface the gradients become steep, and thus the height dependence of v_d is very pronounced.

As most atmospheric deposition experiments are performed in the height range of 2–10 m it should be realized that they pertain to a region where the gradients are small. Hence these measured deposition parameter values are especially well suited for calculations in which the so-called source depletion approach (Sl68) is used. This is because in that approach there are no gradients resulting from the depletion by surface deposition. Instead the material deficit is distributed vertically throughout the diffusing cloud. Of course, the concentration gradients exist close to the ground, but they have no consequence for the validity of this type of parameterization, as it is done at a sufficiently high level for height differences to have a negligible effect. The rapid distribution of deposited material deficit of course assumes that a typical diffusion velocity is large compared to v_d. However, since a typical diffusion velocity is given by the turbulent velocity, u_*, while v_d is at least a factor $u_*/\bar{u}$ less, the above assumption is seen to always be a good one. It is often mentioned that the source depletion model might be satisfactory except for very stable conditions with large deposition velocities. As (7) demonstrates, however, the fact is the contrary: the larger the stability, the smaller v_d is in comparison to u_*, and the closer the source depletion concept is approximated.

4. *K*-MODELS

The source depletion model of dry deposition rests on the assumption that $v_d \ll u_*$. Although this is fulfilled in practice, the method can be regarded as being correct only in the limit of $v_d \to 0$. Attempts to develop physically more correct deposition models have been carried out using eddy diffusivity models (K-models) in which the deposition is treated as a flux boundary condition. Thus

$$D = -K\frac{\partial \bar{\chi}}{\partial z}, \tag{9}$$

where K is the eddy diffusivity. Here we shall discuss some of the problems that occur in the use of such models. We first note that even when they are "refined" to the state where K is taken to be a function of height

1000184

one has to be careful in choosing actual numbers. Using (5) it is seen that to satisfy the flux condition $D = -\chi_* u_*$, K must be given by

$$K = \frac{kzu_*}{\phi_c\left(\frac{z}{L}\right)}. \tag{10}$$

So besides being a function of height and stability it is seen that K is an implicit function of wind speed through its dependence on u_*. For given values of $\bar{u}$ and stability, z/L, neither K nor values of v_d above a certain limit, can be chosen arbitrarily. Thus studies that use a value of K from one source and a value of z_o from another as done, for example by Berkowicz and Prahm (Be78), and then vary v_d and $\bar{u}$ independently are far less realistic than the usual straightforward source depletion approach.

Furthermore it is common in such studies to take v_d at its usual value around the order of 1 cm/sec and then supply it at some arbitrary low grid point level. Thus, in the example of Nordlund *et al.* (No79), it does not seem reasonable to compare a source depletion case using a relevant deposition velocity, with a K-model applying the same v_d at a level as low as the order of the roughness length.

5. CONCLUSION

It is often said that source depletion models are applicable only when the dry deposition velocity is small compared to vertical mixing. As we have seen, however, this is always the case if the deposition velocity is defined at usual observation heights, especially for stable conditions. Examples with large deposition and slow diffusion in the cloud are seen to be completely artificial.

If one instead uses a parameterization of the deposition process which has its reference level very close to the surface, where the concentration gradient is large, one has to take into account the fact that the deposition velocity is a strong function of height in this region. This is usually disregarded in eddy diffusivity or K-models. Using such models one must also regard the interdependence of v_d and K. The two parameters cannot arbitrarily be varied relative to each other.

The only case in which one might regard the deposition velocity as an independent parameter, i.e. not dependent on the turbulent transport processes, is in the case of washout. However, rain falling through a pollution cloud scavenges uniformly throughout the cloud volume, so this is clearly not a case in which one would use a K-model since this is an example of source depletion.

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