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Technical Report



AIR CONCENTRATION AND DEPOSITION RATES FROM UNIFORM AREA SOURCES

Prepared by: MONITORING AND ASSESSMENT RESEARCH CENTRE of the SCIENTIFIC COMMITTEE ON PROBLEMS OF THE ENVIRONMENT. INTERNATIONAL COUNCIL OF SCIENTIFIC UNIONS

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Air concentration and deposition rates from uniform area sources

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Prepared by: Monitoring and Assessment Research Centre of the Scientific Committee on Problems of the Environment, International Council of Scientific Unions

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ABSTRACT

The surface of the earth or the sea may release certain substances to the atmosphere. Often these substances are also classed as pollutants when produced by human activities. This report provides a theory and illustrative charts to predict the natural contribution to the air concentration or deposition. To apply the theory, the natural source strength must be known and, unfortunately, this is often unavailable. The theory has been approximately validated by several natural substances such as radon gas and lead.

1.0 Introduction

The earth's surface (or the sea surface) may sometimes be considered to be a uniform area source for certain substances evolving from the land (or the sea). The atmospheric concentration of such natural substances would continue to grow were there not dry and/or wet removal processes. When the rate of emission from the land (or the sea) exactly balances the loss from the air, a steady-state concentration is achieved in the atmosphere. This is, presumably, what occurs in nature. Frequently, knowledge of the global balance is desired. For this, first-order kinetics are often acceptable. In first-order kinetics, the whole atmosphere is treated as one box and the rate of transfer is proportional to the amount in the box. Such a box model hides important geographical variations of concentration. One of the most important variations, the one considered in this report, occurs when either the land or the sea surface is the source of the substance yet both areas receive wet and dry deposition. Another variation occurs when the surface is a source at certain times of year, and a sink at other times (in the case of CO₂, for example).

Many countries monitor air concentrations or ground deposition rates of chemicals of particular interest such as heavy metals (e.g. lead, mercury, etc.) or sulphur compounds. Most often the purpose of the measurements is to estimate whether certain sources require regulation. Many of the chemicals monitored for this or other reasons possess both man-made and natural sources. Very extensive and expensive monitoring programmes can often provide the evidence to distinguish between these sources. Alternatively, it may be possible to use the curves given in the figures of this report to estimate air concentrations or deposition rate due to natural sources.

One can also compute the air concentration or deposition rate due to a man-made source. One often then asks "How much above the natural background, the irreducible lower limit, is the man-made contribution?". With natural source strengths, the present report will provide an estimate of the natural background.

To use the figures in this report, the natural source strength must be known. This is the average emission rate per unit area of land or sea surface for the region surrounding, and far upwind of, the point at which the calculation of air concentration or deposition rate is desired. If this emission rate per unit area is variable over the region or variable with time of day, weather pattern, or season, an average value should be chosen. Unfortunately, emission rates from the land or the sea are still poorly known or entirely unknown. The estimated values of the air

concentrations or deposition rates must be long-term monthly, seasonal or annual averages in order to average out non-typical airflow or precipitation patterns.

2.0 The assumptions

In general, the prevailing airflow in temperate latitudes is from west to east and in tropical latitudes from east to west. Individual daily trajectories meander considerably; that is, the wind is normally variable both in time and space. One consequence of this variability is that the vector mean wind is less than might be expected from an inspection of daily weather charts. For example, the vector mean wind at 850 millibars (about 1500 m, or 5000 ft, above mean sea level) over the United States does not exceed 15 knots (about 8 m s⁻¹). Yet it is at this altitude that one might consider the winds as transporting substances east of the Rocky Mountains.

Atmospheric diffusion from a point source proceeds outward in three dimensions. However, for a large uniform area source, horizontal diffusion may be neglected; only vertical diffusion need be considered.

A set of calculations treating vertical diffusion by classical eddy diffusion theory has been performed on a high-speed computer for average conditions of turbulence (Draxler and Elliot, 1977). The results form the basis for estimating the dilution due to vertical dispersion.

Removal processes may occur under either dry or precipitating (rainy or snowy) weather. Dry deposition is modelled in the usual way by assuming a numerical value for the dry deposition velocity, V_d . The adjustment to the loss from the bottom box in the model occurs through ordinary vertical diffusion. For those substances which deposit on the earth's surface, the usual range of deposition velocities is about 0.1 to 1.0 cm s⁻¹.

Wet deposition is very poorly known. The present calculations assume a value for E, the ratio of substance concentration in precipitated water to that in air, from past experience. The air concentration is taken as the average value between ground (or sea surface) and 4000 m, the assumed top of the rain-bearing clouds. Often another expression for precipitation scavenging is given, a wet deposition, Λ cm⁻¹ (where Λ is the fraction of the pollutant removed by 1 cm of rain). Johnson, Wolf and Mancuso (1975) suggest values of Λ of 1.0 cm⁻¹ in winter, 6.5 cm⁻¹ in spring and autumn, and 10.0 cm⁻¹ in summer. These values may be compared with the equivalent value used in the present calculations where $\Lambda = 1.02$ cm⁻¹.

3.0 Results

The largest contribution to both ground-level air concentrations and deposition rates derives from the source most immediately upwind of the point of interest or sampling point. But some contributions derive from all upwind sources.

3.1 Approach to uniformity of concentration and deposition rates. For purposes of discussion a land source is assumed.

Figure 1 shows the build-up of ground level air concentrations over a land surface with travel time starting from the windward edge of the land mass. With no dry or wet deposition losses (the uppermost curve), there is little or no tendency towards an asymptotic, constant concentration within five days. Geographically uniform concentrations are not present.

Each of the other three curves assumes deposition losses, the lowermost curve having both the largest deposition velocity and the greatest rate of precipitation. Travel times may be converted to transit distances given mean transport winds. The conversions shown on the chart include a factor of 1.5 for trajectory meandering, i.e. a typical trajectory on the scale of hundreds of kilometres takes 1.5 times the straight-line distance between two points even along the mean wind direction.

Concentrations, the ordinate in Figure 1, assume a source strength of one unit of substance per square centimetre of ground area per second. The unit of substance may be 1 gram, 1 curie, * 1 particle, etc. To find the concentration from other than unit source strength, multiply the concentration, the ordinate, by the actual source strength. To convert concentration expressed as a mixing ratio (e.g. amount of substance per gram of air) to volume concentration (e.g. amount of substance per cubic metre), multiply by the ground level air density, 1.223 x 10³ grams of air per cubic metre if at sea level.

The various ordinate values are directly comparable in Figure 1. Thus, after 72 hours of travel, the concentration with marked dry and wet deposition is about one-fourth that with no deposition.

The approach to a geographically uniform concentration takes place within 24 hours of travel for the fast removal (bottom curve) but takes over five days for slow removal. In terms of transcontinental air transport with, say, a 5 m s⁻¹ wind, fast removal approaches uniformity after

* The replacement of the curie (Ci) by the becquerel (Bq), the SI unit of activity of a radioactive source, occurs in this report. $1 \text{ Ci} = 3.7 \times 10^{10} \text{ Bg}$.

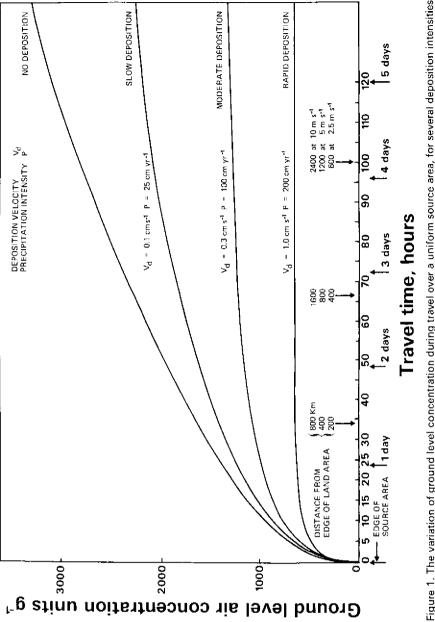


Figure 1. The variation of ground level concentration during travel over a uniform source area, for several deposition intensities. The concentrations have been normalized to a unit source strength of 1 unit cm⁻² s⁻¹. The concentration per unit volume (m⁻³) can be found by multiplying the ordinate by air density (1.223 x 10³g m⁻³).

about 150 km of travel but over 1500 km are needed if the deposition is slow.

It should be emphasized again that the concentrations are long-term climatological averages. The trajectories and wet deposition rates will vary greatly from day to day. But averaged over many trajectories, observed concentration should tend to approach calculated values.

If the deposition is rapid, it is argued that any place with more than a few hundred kilometres of overland travel would have a similar longterm average concentration at ground level.

3.2 Transition of concentration from water

Figures 2 and 2a present a case of 24 hours of land travel followed by about four days of overwater travel; only the land surface is the assumed source. The ground level air concentration decreases over water with or without deposition since the substance continues to diffuse upward from the ground level source. But the decrease is much more rapid for the case of removal from the air. For very rapid removal, it became necessary to use a logarithmic scale for air concentration to display the very rapid drop-off over water. After long overwater travel, the differences in air concentration among the several removal rates, including no removal, become more marked than the overland travel for the same travel times measured from the upwind land edge. The very abrupt decrease downwind of the land-water interface is especially noticeable.

3.3 Deposition rates over land

Figure 3 shows the change of deposition rate with travel time. As with the previous discussion on concentration, travel time and travel distance may be interchanged given a transport speed. The pattern is similar to that in Figure 1 with one obvious exception: the higher deposition rates derive from larger deposition velocities and greater rainfall rates. The curves for the higher deposition velocities and the greater rainfall rates again approach geographical uniformity more quickly than for the smaller values.

During the early stages of the build-up of substances in the air, the concentrations are less disparate than the deposition rates among the several removal rates; after four or five days of travel, however, the reverse is true (compare Figures 1 and 3).

The deposition rates also derive from a source of one unit of a substance (1 gram, 1 curie, etc.) per square centimetre per second. Multiplication of the ordinate values of deposition rates by the actual source

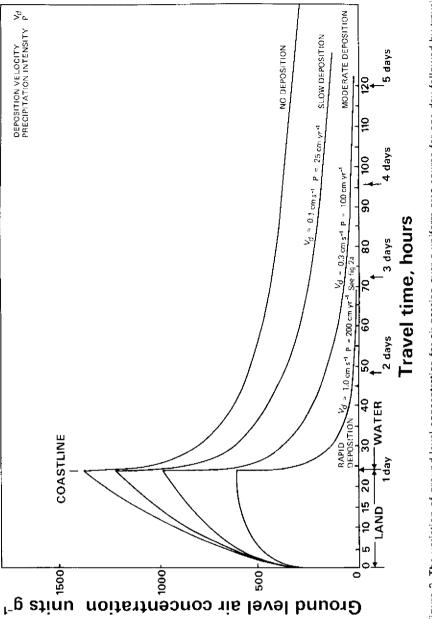
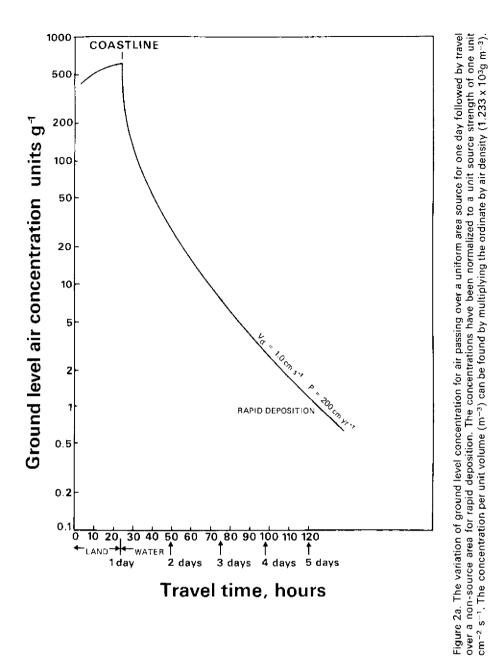


Figure 2. The variation of ground level concentration for air passing over a uniform area source for one day followed by travel over a non-source area, for several deposition intensities. The concentrations have been normalized to a unit source strength of one unit $cm^{-2} s^{-1}$. The concentration per unit volume (m^{-3}) can be found by multiplying the ordinate by air density (1.233 x 10³g m⁻³). (Note change of scale of the ordinate between Figures 1 and 2).



strength per square centimetre per second converts the chart numbers to those appropriate for the true source strength. The numerator of the deposition rate contains the same unit as the source input.

3.4 Transition of deposition rates from land to water

Figure 4 presents the deposition rates for the case of 24 hours of travel over land followed by about four days over water. The build-up over land is identical with that in Figure 3. The decrease of the deposition rates on the water side of the land-water interface is very rapid, especially for the high removal case. Farther from land, the curve for the heavy deposition actually lies below the other two because of the large earlier removal. The deposition over water, like that for air concentration, is far smaller than over land once removed from the boundary of the land source, especially if the initial removal is heavy.

4.0 Some verifications

4.1 Lead

Elias, Hirao and Patterson (1975) estimate the natural wind-blown (or airborne) lead for a canyon in California, U.S.A. as being about 0.2 kg yr⁻¹ per 12 km². This may be converted to about 5 x 10^{-17} g cm⁻²s⁻¹. The long-term air concentration in the high Sierra Mountains of California is 25 x 10^{-9} g m⁻³. But the authors estimate that only 0.002 to 0.01 of the concentration is of natural origin. Thus the wind-blown (non-man-made) lead concentrations are between 5-25 x 10^{-11} g m⁻³.

From Figure 1 it is estimated that the lead concentration for a unit source might lie between $5 \cdot 15 \times 10^2 \text{g} \text{ g}^{-1}$ or between $6 \cdot 18 \times 10^5 \text{g} \text{ m}^{-3}$ depending on travel time (about one day is appropriate) and removal rates. However, correcting for the above source strength, $5 \times 10^{-17} \text{g} \text{ cm}^{-2} \text{s}^{-1}$, yields concentrations of $3 \cdot 9 \times 10^{-11} \text{g} \text{ m}^{-3}$. This is to be compared with $5 \cdot 25 \times 10^{-11} \text{g} \text{ m}^{-3}$.

4.2 Radon

Israel (1951) summarizes land radon emission rates as lying between about 0.74 and 2.59 μ Bq cm⁻²s⁻¹ with an average of 1.48 μ Bq cm⁻²s⁻¹. His estimates of radon air concentrations are about 2.22-14.8 μ Bq cm⁻³ with the average a bit closer to the lower end.

Radon -222 is a non-depositing, noble, radioactive gas with a halflife of 3.8 days. This half-life may be simulated by no dry deposition

but with wet deposition corresponding to a precipitation rate of about 25 cm yr⁻¹. Interpolating for this value in Figure 1 (at about five days) suggests a concentration, for a unit source strength of radon, of about 92.5 T Bq g⁻¹ or about 0.111 T Bq cm⁻³. Multiplication by 40 x 10⁻¹⁸ yields 4.44 μ Bq cm⁻³. This is to be compared with 2.22-14.8 μ Bq cm⁻³.

4.3 Summary

In both cases, for lead and radon, validation is good, considering the very wide range of variation in source strengths and air concentrations of the natural substances and the simplifying assumptions of the calculations.

5.0 Applications

5.1 Water Sources

The calculations and the charts apply equally to a uniform continuous area source over water and no source over land.

5.2 Estimation of source strength from concentrations and deposition rates

The charts may equally well be used to estimate average continuous source strengths, given ground level average concentrations or depositions. However, if the substance also has anthropogenic sources, the natural source strength will be over-estimated from the charts.

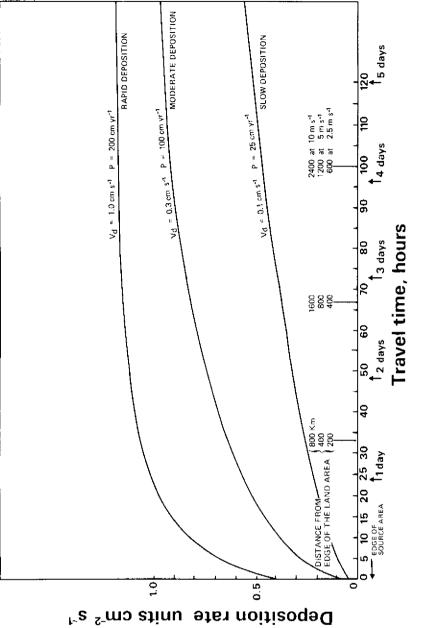


Figure 3. The variation of deposition rate during travel over a uniform area source for several deposition intensities. The deposition rates have been normalized to a unit source strength of one unit $cm^{-2} s^{-1}$.

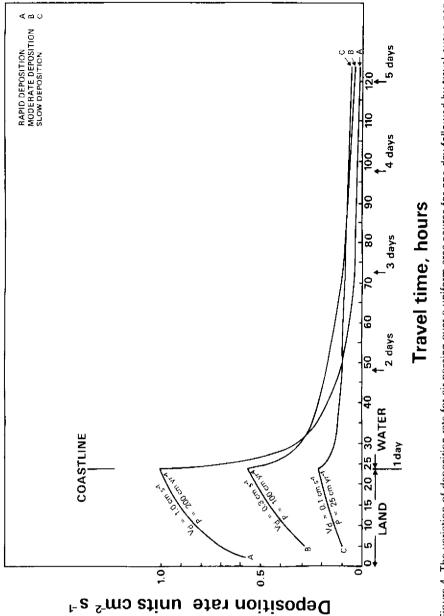


Figure 4. The variation of deposition rate for air passing over a uniform area source for one day followed by travel over a non-source area, for several deposition intensities. The deposition rates have been normalized to a unit source strength of one unit cm⁻² s⁻¹.

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