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GLOBAL VERTICAL MASS TRANSPORT
BY CLOUDS — A TWO-DIMENSIONAL
MODEL STUDY

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Global vertical mass transport by clouds – a two-dimensional model study

by Mats Olofsson

Abstract

A two-dimensional global dispersion model, where vertical transport in the troposphere carried out by convective as well as by frontal cloud systems is explicitly treated, is developed from an existing diffusion model. A parameterization scheme for the cloud transport, based on global cloud statistics, is presented.

The model has been tested by using Kr-85, Rn-222 and SO₂ as tracers. Comparisons have been made with observed distributions of these tracers, but also with model results without the cloud transport, using eddy diffusion as the primary means of vertical transport. The model results indicate that for trace species with a turnover time of days to weeks, the introduction of cloud-transport gives much more realistic simulations of their vertical distribution. Layers of increased mixing ratio with height, which can be found in real atmosphere, are reproduced in our cloud-transport model profiles, but can never be simulated with a pure eddy diffusion model. The horizontal transport in the model, by advection and eddy diffusion, gives a realistic distribution between the hemispheres of the more long-lived tracers (Kr-85).

A combination of vertical transport by convective and frontal cloud systems is shown to improve the model simulations, compared to limiting it to convective transport only. The importance of including cumulus clouds in the convective transport scheme, in addition to the efficient transport by cumulonimbus clouds, is discussed. The model results are shown to be more sensitive to the vertical detrainment distribution profile than to the absolute magnitude of the vertical mass transport.

The scavenging processes for SO₂ are parameterized without the introduction of detailed chemistry. An enhanced removal, due to the increased contact with droplets in the in-cloud lifting process, is introduced in the model.

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1. Introduction

The thin layer around the globe representing the troposphere contains almost all of the clouds and other meteorological features normally referred to as "weather". Air is transported within the troposphere with the general circulation pattern as well as with turbulent small-scale motions. These transport mechanisms also affect aerosols and gases present in the tropospheric air. The increasing release of man-made pollutants into the atmosphere has raised the need to investigate their influence on the environment. Numerical modelling of the atmosphere represents an important tool to estimate the distribution of various pollutants.

Because of the large variations in both space and time exhibited by reactive trace gases and aerosol particles, an accurate description of their global distribution requires a complex three-dimensional (3D) transport model. However, such models call for very large computer resources. Only a few global 3D chemical tracer models have been formulated so far (Jacob et al., 1987).

In most cases it is thus necessary to simplify the analysis by using 1D (normally the vertical) and 2D (height/latitude) models of the atmosphere. In 1D models all horizontal motions have to be neglected. They are therefore only useful in horizontally homogeneous situations, when the dominant fluxes are directed in the vertical direction. An example of this is in studies of the distribution of trace gases in the boundary layer of the remote marine atmosphere.

In studies of the global distribution of trace constituents the most common type of dispersion model has been the 2D category. Since the general circulation pattern will reduce horizontal differences in the zonal direction much quicker than in the meridional, a height - latitude model is normally used. Several such models have been introduced during the last 10-15 years (Machta, 1974; Crutzen, 1977; Isaksen and Rodhe, 1978; Rodhe and Isaksen, 1980; Pearman and Hyson, 1986). Chatfield and Crutzen (1984) developed a hybrid of two and three dimensions, working on a limited area.

In most of these models, an eddy diffusion parameterization has been used to describe the vertical transport, in addition to a rather slow exchange of air by the mean vertical wind field. Emissions, winds and diffusion parameters have to be specified, to calculate the horizontal and vertical distribution of the species studied.

A major problem in numerical modelling of tracer substances is the verification. The magnitude of sources as well as removal mechanisms are still uncertain for most pollutants. The number of tracers with a welldefined source and a predictable loss is very limited. We have chosen Krypton-85 and Radon-222 as initial model tracers to test the validity of the transport schemes, due to their uncomplicated removal processes (Machta, 1974), in spite of the limited amount of observational data.

Another characteristic that defines a useful tracer is its turnover time in the atmosphere. The time-scale of the transport mechanisms must be similar to the turnover time of the tracer, to make comparison with observed data meaningful. The tropospheric exchange time-scale we are

interested in ranges from hours–days for the vertical in-cloud transport up to years for the exchange between the hemispheres.

An important factor in the vertical transport is the massive lifting of air in cloud systems, convective clouds as well as frontal cloud systems, and the compensating subsidence in surrounding areas. This process (hereafter referred to as the "cloud-transport" mechanism) accounts for an important redistribution of air in the troposphere, but can not be represented by a pure eddy diffusion parameterization (see below). This problem has been described previously by Gidel (1983), and especially by Chatfield and Crutzen (1984), who introduced the concept of "Staubsauger" (German for vacuum-cleaner) to describe the rapid transfer of boundary layer air to the upper troposphere, carried out by cumulonimbus clouds. They used a 2D model (horizontal radial distance – height) applied to a limited area, into which five regions of different cloud activity were introduced. Each region resulted in a specific vertical redistribution of mass.

Our approach has been to parameterize the vertical mixing process using cloud statistics, extending the model region to the global scale, using latitude and height as model coordinates. The lifting process has been divided into two separate mechanisms – convection primarily in Cumulonimbus clouds (Cb) in unstable air masses, and the slower, more organized lifting in midlatitude cyclones and frontal systems. This vertical redistribution of air – and thereby of trace species mixed with the air – is a mechanism that can transport boundary level air directly to the levels of outflow in the middle and upper troposphere, in a short timescale and without intermediate mixing and dilution on the way (Raymond and Wilkening, 1985). As the result of such a transport, a tracer emitted from the ground may exhibit layers with increasing concentration with height. This was considered already by Jacobi and André (1963): "It must be concluded that the high Rn-222 content.....is mainly due to upward-directed convection, which may occur especially above continental areas, rather than to turbulent diffusion". A discussion of the importance of storm venting is also presented by Dickerson et al. (1986).

In this study, the vertical mass transport directly related to convective and frontal cloud systems has been explicitly treated. In the two-dimensional model by Isaksen and Rodhe (1978), which has been the base for our model, this vertical transport is only implicitly described, by the use of vertical turbulent diffusion parameters, K_{zz} .

Parameterization schemes using the eddy diffusion theory are based on descriptions of vertical mixing as a slow "seeping" process by small-scale unorganized motions. The assumed K_{zz} -values used in most such schemes (in the range 1-20 m^2s^{-1}), give a time-scale which is long compared to the time-scales of removal of many chemical species. As a result, the calculated concentrations of reactive trace species often show a rapid decrease with height (Rodhe and Isaksen, 1980). However, in the atmosphere, most vertical mixing occurs in relatively isolated events of great rapidity and depth (Chatfield and Crutzen, 1984). The time-scale of vertical air motions in synoptic disturbances is 2-3 days, while the transport from ground level to the tropopause in Cb clouds can be accomplished within an hour. Thus, when

modelling trace species like SO₂ and Radon-222, with turnover times of 2-5 days, the eddy diffusion theory is inappropriate. Also, since diffusion processes can only transport a tracer down the concentration gradient, the profile of the tracer would always decay with height from a source at the surface. No matter how much the vertical diffusion is enhanced, an increase in the tracer mixing ratio with height can never be produced (Gidel, 1983).

2. Objectives

The aim of this study has been

- to modify an existing two-dimensional model (Isaksen and Rodhe, 1978) to include an improved description of the vertical transport mechanisms
- to develop a parameterization scheme of convective as well as frontal clouds, as transporters of boundary layer air to higher levels in the troposphere
- to estimate the transport parameters of such a scheme, by making systematic comparisons between model simulations and observed distributions of Krypton-85, Radon-222 and sulfur dioxide

3. The model

3.1 The continuity equation

The present model has been developed from the 2D global model presented by Isaksen and Rodhe in 1978. It uses a height/latitude plane, extending horizontally from pole to pole and vertically between the ground level and 16 km. The transport is given by advection and by eddy flux. In our version the model is expanded to include vertical cloud transport, in convective clouds as well as in frontal cloud systems.

The transport by advection and by eddy flux is given by the continuity equation

$$\begin{aligned} \frac{\partial c_i}{\partial t} = & \frac{-1}{a \cos \phi} \frac{\partial (c_i v \cos \phi)}{\partial \phi} - \frac{\partial (c_i w)}{\partial z} + \frac{1}{a \cos \phi} \frac{\partial (M K_{yy} \cos \phi \frac{1}{a} \frac{\partial (x_i)}{\partial \phi})}{\partial \phi} + \\ & + \frac{\partial (M K_{zz} \frac{\partial (x_i)}{\partial z})}{\partial z} - L_i c_i + P_i \end{aligned} \quad (1)$$

where c_i is the tracer concentration in the grid box, M the number density of air, x_i the volume mixing ratio, ϕ the latitude and a the radius of the earth. v is the mean meridional and w the mean vertical wind components respectively, while K_{yy} and K_{zz} are the corresponding eddy diffusion coefficients. $L_i c_i$ and P_i represent removal and production processes. All parameters are zonally averaged. We have in this study excluded the anisotropic eddy diffusion coefficients K_{yz} and K_{zy} , which are used in the Isaksen and Rodhe model, since they are of little importance in the troposphere.

In addition to these terms we have introduced the vertical redistribution by clouds. The mass transport parameter, F , is given fixed values for each latitude and season (further described in chapter 4). A secondary vertical velocity field is calculated, representing the subsidence between the levels within a latitude band corresponding to the magnitude of the in-cloud lifting process. The exchange of air between the different model levels due to these processes is assumed to be confined within each latitude band. In this way continuity is retained while not affecting the large-scale w - or v -fields, representing the general circulation cells. Naturally, since the cloud-transport process is changing the concentration profile of a trace element within the grid column, it will also affect adjacent columns indirectly. The cloud-transport terms added to the continuity equation then take the following form:

$$\frac{\partial c_i}{\partial t} = F_i x_{i(m=1)} - \frac{\partial (c_i w_{sub})}{\partial z} \quad (2)$$

where F_i is the prescribed airflow into the gridbox due to the cloud-transport, $x_{i(m=1)}$ the volume mixing ratio at the bottom level and w_{sub} the compensating subsidence velocity.

3.2 Numerical solution

The horizontal model resolution covers 19 gridpoints. The end points ($l=1$ and $l=19$) represent the two pole-caps between 85° and 90° , while the remaining 17 gridpoints are the central points of latitudinal bands with a width of 10° (about 1100 km).

In the vertical direction the model consists of 20 levels. They represent the mid-level in layers of 500 m from the ground up to 3.5 km. Above the highest of these levels, the grid distance is 1000 m. Thus, the lowest model level ($m=1$) is 250 m, while the highest is 16250 m.

The numerical solution of the continuity equation is accomplished in the following way: The time derivatives are approximated by forward differences ($t, t+\Delta t$). The advection terms and the eddy diffusion terms are approximated by centered differences in space ($l-\Delta l, l+\Delta l$ and $m-\Delta m, m+\Delta m$). In the advection scheme a correction is introduced in the form of an "anti-diffusion velocity" (Smolarkiewicz, 1983). This is done to compensate for the implicit numerical diffusion.

The time integration has been carried out using a time-step of two hours for the turbulent diffusion and the cloud transport processes, while the advective process time-step was set to 24 hours. These time-steps are sufficiently short to avoid the risk of numerical instability (Smolarkiewicz, 1983).

The use of tracer concentration as the dependent variable in equations (1) and (2) results in a problem of mass-continuity at the shift of seasons. Since the air density is calculated from seasonal averages of climatologically determined temperature fields (thus given in four discrete sets of values, one for each season), a sudden change in air density will occur at the seasonal shifts. The tracer mass will thereby not be conserved. This is of little importance for tracers with a short turn-over time (day-week), since they will regenerate a correct mass-field several times within a season, but it will be of significance for species with a life-time of years. To avoid this continuity problem, we integrated the model using the number of tracer atoms within each grid box as the dependent variable. Since this quantity is given by the concentration multiplied by the size of the respective grid box, we now had to introduce some finite differences into the equation. We replaced equations (1) and (2) with the new continuity equation

$$\begin{aligned} \frac{\partial n_i}{\partial t} = & - \frac{1}{a \Delta \phi} \frac{\partial \left(n_i v \frac{\cos(\phi-0.5\Delta\phi)}{\cos\phi} \right)}{\partial \phi} - \frac{1}{\Delta z} \frac{\partial (n_i w)}{\partial z} + 2\pi a \Delta z \frac{\partial \left(M K_{yy} \cos\phi \frac{\partial x_i}{\partial \phi} \right)}{\partial \phi} + \\ & + 2\pi a^2 \Delta \phi \cos\phi \frac{\partial \left(M K_{zz} \frac{\partial x_i}{\partial z} \right)}{\partial z} + F_i x_i (m=1) - \frac{1}{\Delta z} \frac{\partial (n_i w_{sub})}{\partial z} - L_i n_i + P_i \end{aligned} \quad (3)$$

where n_i is the number of tracer atoms (or molecules) in the grid box, $\Delta\phi$ the meridional and Δz the vertical grid distances.

A schematic representation of the transport mechanisms included in equation (3) is given in figure 1.

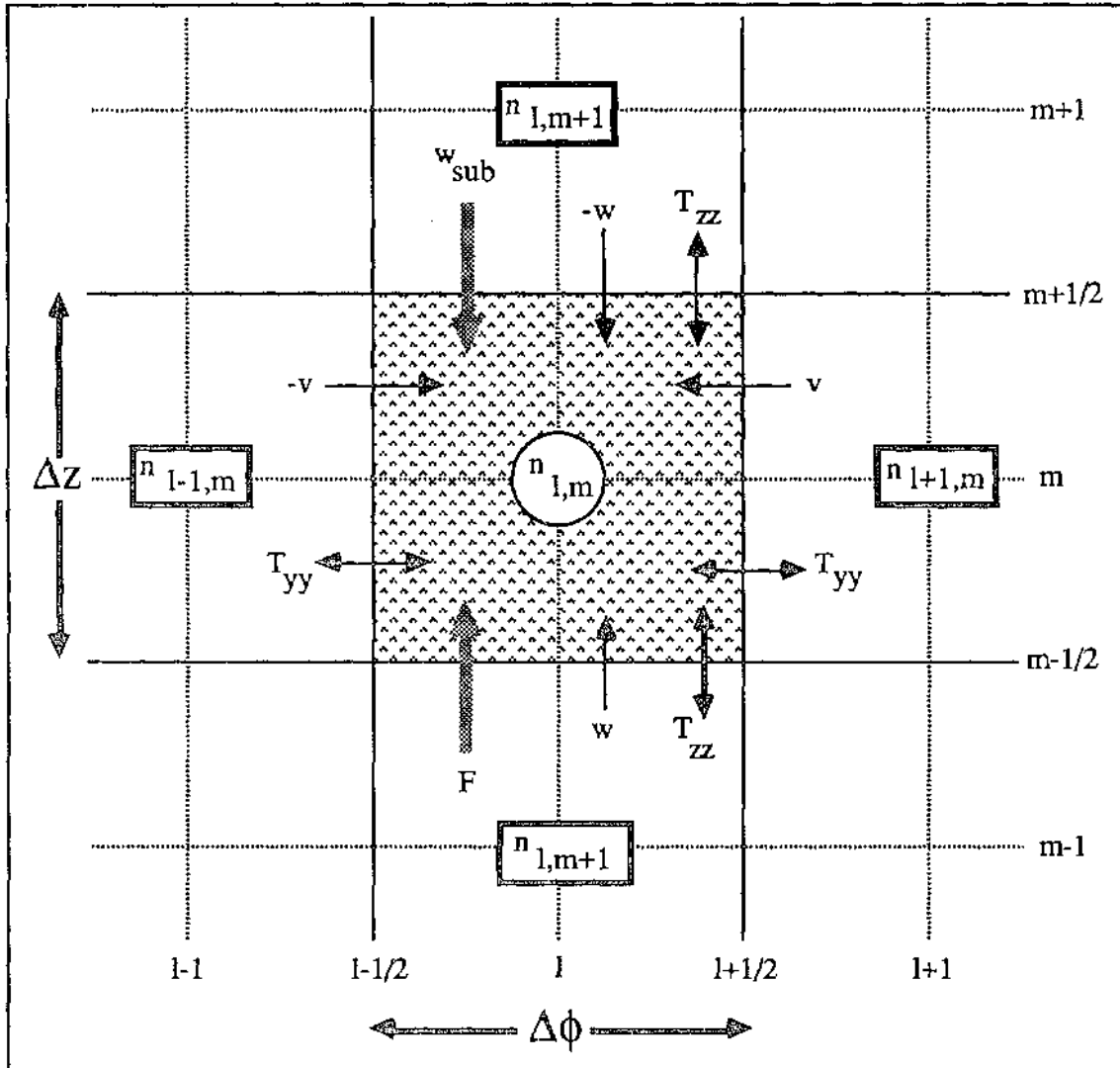


Figure 1. Schematic picture of the transport parameters in the model. There are three different mechanisms affecting the tracer content within a grid-box:

- A: Advection from the adjacent boxes – horizontally by the meridional wind, v or $-v$
– vertically by the vertical wind, w or $-w$
- B: Turbulent exchange – horizontal exchange by T_{yy} ($T_{yy} = -K_{yy} \partial n / (\text{acos}\phi \partial \phi)$)
– vertical exchange by T_{zz} ($T_{zz} = -K_{zz} \partial n / \partial z$)
- C: Cloud transport – by the inflow parameter F (air from the ground level)
– by the downward compensating velocity w_{sub}

3.3 Model version separating continents and oceans

When using a zonally averaged 2D model with a tracer having a short turnover time (on the scale of days to weeks), a certain aspect of the verification problem has to be considered. A comparison of model results with observed data becomes extra sensitive to where the measurements were taken, unless the tracer emission is completely uniform in the zonal direction.

Radon-222, with a half-life time of about four days, is a good example of this problem. Because of the westerly mean zonal winds, a vertical concentration profile of Rn-222 measured over the eastern part of a continent should show higher values than one over the west coast or over the ocean. This is due to the almost negligible emission of Rn-222 over the ocean, and its turnover time being less or equal to the time-scale for advection of an air-mass over a continent or an ocean. Thus, a zonally averaged concentration profile, as estimated in a 2D model, could be a poor approximation of the observed state at a specific longitude. To overcome this problem, we constructed a modified model to use with Rn-222. Still a two-dimensional model, it allows the Radon-emission to switch between a continent rate and an ocean rate. The "switch" is triggered by a time-function, governed by the climatological mean zonal wind component for the season. We have regarded the wind at 500 mb to be representative for the advection velocity of an air-mass. Moving eastwards around the globe, the model atmosphere is affected by two continents and two oceans, their geographical relation and the advection velocity taken from the observed values at latitudes 30-50N (Newell et al., 1972). The exposure times over the different surfaces, for each season, are presented in table 1.

Season	[u] m/s	Time spent over different surfaces (days)				Total time (days)
		Cont	Ocean	Cont	Ocean	
1	15	9	7	3	4	23
2	14	10	7	3	5	25
3	9	15	11	5	7	38
4	12	11	9	4	5	29

Table 1. Time during which the model atmosphere is exposed to emission from continents and oceans, in each cycle around the globe. Also shown is the total time for a full cycle, for each season. [u] is the mean zonal wind at 500 mb (about 5500 m) for each season respectively.

This continent/ocean model could be used for any tracer with a well-defined differentiated emission pattern in the zonal direction. For a species with a life-time considerably longer than the time of exposure (see table 1) this separation is unnecessary.

Another advantage of this modified model is that it gives an option to differentiate the cloud transport parameters between continent and ocean. For example, we know from climatology that the summer-season convection is more pronounced over continents than over oceans, and that the convection is suppressed over the eastern parts of the subtropical oceans. However, we have not yet carried out any such attempts to further refine the 2D model.

3.4 Boundary conditions

In each time-step, the mass concentration above the poles is set equal to that of the same level in the adjacent grid box, i.e. $c(1,m) = c(2,m)$ and $c(19,m) = c(20,m)$. In this way no abrupt jump in the concentration values will occur close to the poles, although the transport into the polar regions is kept zero ($v=0$ and $K_{yy}=0$).

As a result of this procedure, the total mass inventory will not be in exact balance with the total emission. However, the polar regions (latitudes 85-90°) account for less than 2% of the total mass of the atmosphere, and the concentrations in these regions is low for most species. We have calculated the balance in equilibrium global inventory, due to this model description, to be less than 1% for the tracers treated in this report.

The vertical advective transport between the two highest grid levels, $w(1,20)$ is set equal to zero. Still, the K_{zz} -field allows exchange between the highest grid level and the underlying, as does the cloud transport mechanism.

No flux is allowed through the upper boundary of the model. The small fluxes that might occur in reality are considered insignificant in this work. Our purpose is here mainly to study species emitted from the ground.

The treatment of the lower boundary is described in some detail in Isaksen and Rodhe (1978) and their method has been adopted into our model. A main problem is to relate the dry deposition velocity parameter at the surface, and thereby the vertical flux, to that which applies to the lowest model level.

A point of interest, when comparing our model results with observed data on concentrations at the ground, could be mentioned here. For a tracer emitted from the ground and with a short turnover time, the concentration difference between "true ground" and our lowest model level should be considered. As an example we can regard the radioactive element Radon-222 (half-life 3.8 days) with a boundary layer K_{zz} of $5 \text{ m}^2\text{s}^{-1}$. Using the formula of concentration changes with height in a diffusive environment (Machta, 1974)

$$c(z) = c_0 \exp\left(-z \sqrt{\frac{\lambda}{K_{zz}}}\right) \quad (4)$$

where c_0 is the concentration at the ground, $c(z)$ the concentration at height z and λ the radioactive decay constant, we get $c(250) = 0.85 c_0$. The results presented in this report have not been recalculated to represent "true ground".

3.5 Parameter values

As has been described in the previous sections, the model is based on many parameters that have to be prescribed at all the grid-points, in order to calculate the fields of the dependent variables. An ambition to continuously calculate the transport parameters by the use of the dynamical equations of motion, would require a computing power far beyond our resources, and such an effort would not be in consequence with the zonal averages approximation of the output.

The *advective transport* in the model is carried out by zonal average values of v and w , that need to be stated at the boundary surfaces between the grid boxes. As Isaksen and Rodhe (1978), we have used values taken from Newell et al. (1972), where the w -values are calculated from the v -values and the continuity requirement. The continuity is obtained by the use of a stream function.

The *mean temperature* field has also been taken from Newell et al. (1972). It determines the *air density* field used in the model calculations.

Turbulent diffusion coefficients are crucial parameters in all diffusion models. They have commonly been estimated by calibrations of the models, using known distributions of some tracers. In this study, most of the vertical eddy diffusion has been replaced by an explicit cloud transport. The K_{zz} -values have been reduced accordingly and their exact magnitude becomes less critical. The K_{yy} -values have, with some modifications, been adopted from Hidalgo and Crutzen (1977). The main modification is a three-fold increase in the tropics, to intensify the interhemispherical exchange which otherwise seemed to be too slow. The K_{zz} -values used in our "diffusion only" model simulations (no cloud-transport) were taken from the same source, with only slight modifications.

The vertical mixing through the boundary layer is parameterized as in Isaksen and Rodhe (1978). Above this lowest model level, we have introduced a uniform field of $K_{zz} = 5$ (middle and high latitudes) and 10 (tropics) $\text{m}^2 \text{s}^{-1}$ up to the cloud base level of about 2 km, and $K_{zz} = 0.5 \text{ m}^2 \text{ s}^{-1}$ for the remaining model atmosphere. These values are subjectively simplified averages from Hidalgo and Crutzen (1977) for the lower levels, while at the levels above the model cloud base most of the turbulent exchange is replaced by cloud-transport, as described in the following section.

The *scavenging mechanism* is divided into three separate processes. These are:

1. Reversible incloud scavenging - a removal taking place when the species is dissolved reversibly into cloud droplets which fall to the ground as precipitation.

2. Irreversible incloud scavenging - a removal by transformation in the cloud droplets without the reappearance of the species if the droplet evaporates.

These two processes are based on an estimate of the probability for a tracer to be exposed to cloud water in the atmosphere, and the solubility of the species concerned. The parameter values have been adopted from Isaksen and Rodhe (1978), where the processes are described in more detail. The efficiency factor for the removal of SO₂ was set to 0.3 for the reversible scavenging and to 0.17 for the irreversible, based on an assumed turnover time of SO₂ of about 4 days with respect to irreversible scavenging. For Krypton-85 and Radon-222 we have regarded the solubility in cloud water, and thereby the cloud scavenging, as negligible.

3. An enhanced scavenging in Cb and frontal clouds is introduced in our model, in addition to the processes mentioned above. The reason for this is the considerably increased probability for a tracer to be exposed to cloud water in the cloud-transport processes. This increased likelihood for removal has been discussed by Hales and Dana (1979), Rodhe (1983) and in a NAPAP-publication (Albritton et al., 1987). We have used a relation between the inflow and removal of SO₂ in a convective cloud, as estimated in the latter publication. However, the magnitude of this scavenging efficiency must be considered as uncertain.

In addition to the above mentioned scavenging processes, a boundary layer removal by dry deposition was calculated in the model. A deposition velocity of 0.6 cm s⁻¹ was used for SO₂. This process was neglected for Kr-85 and Rn-222.

4. Cloud parameterization

The vertical transport by clouds in the model has been divided into two parts – one carried out by convective cloud systems and one by frontal cloud systems. The former is present in the model at all latitudes except in the polar regions, while the latter has been restricted to the belt between latitudes 35 and 75 degrees in each hemisphere. To compensate for the vertical exchange explicitly described by the cloud processes, we had to reduce the K_{zz}-values substantially compared to those used in a diffusion model like that of Isaksen and Rodhe (1978). The K_{zz}-field now accounts only for the turbulent exchanges not parameterized as cloud processes (see section 3.5).

In the results presented below, we have tested the model sensitivity to the magnitude of vertical mass transport, but the relative distribution between the latitude bands has not been altered. The latter is based on cloud observation statistics, while the magnitude of the flux is calculated also from assumptions on the average updraught velocity. The values of the cloud-transport has been tuned by comparisons with observed tracer distributions. In the commonly

used eddy diffusion approach parameter values are determined in a similar way. The cloud-transport mechanism is another, in our opinion more realistic, way to describe the vertical mixing in the atmosphere.

4.1 Convective clouds

4.1.1 Data

The parameterization of convective clouds is based on global statistics of synoptic observations, presented by Hahn et al. (1982) and Warren et al. (1986). They have analyzed eleven years of observations from WMO synoptic stations, 1971-1981. From these data we have used the information concerning frequency of Cb clouds in each latitudinal band and amount of Cb when present (the latter only available for land observations). The amount of Cb clouds obtained in this way is however far too high, if used without considering the construction of the synoptic code. When Cb is present it is always reported as *the* low cloud type (C_L in the code), while the amount stated tells the total amount of low clouds (N_h) including Cumulus (Cu) and Stratus (St) clouds etc. As soon as a single Cb cloud appears on a sky with for example 4 octas of Cu and some St, a synoptic observation will report this as 4 or 5 octas of Cb, since Cb has got the highest priority. In this way, the Cb-amount will be highly overrepresented, if just multiplying the frequency of Cb observations with the amount-figures.

Thus, the final Cb-amount was obtained by multiplying the above described Cb cloud coverage by a factor of 0.2. This factor is a somewhat subjective figure, obtained through a non-systematic check of observations from some major stations in Europe and Africa, that give detailed cloud information in addition to the ordinary code.

Since the aim of the Cb-study has been to get a value of the vertical mass transport, we have to consider only the part of an average Cb cloud being active as updraught "towers". Riehl and Malkus (1958) estimate that these updraught towers, as an annual average, occupy about 0.1% of the total surface area in the tropics. We have assumed that the updraught towers cover 5-10% of the cloud base area as an average in space and over the cloud life cycle. This will give an active area slightly smaller than the above mentioned 0.1% of the total surface area. Our assumption is guided by a summary presented by Atkinson (1981), where total air flux calculations into various storms are stated. With updraught velocities of $5-10 \text{ ms}^{-1}$, as used in our model, these flux values represent "towers" covering 5-20% of an average storm.

After calculation of "active areas" in each latitude band, we now introduce three different classes of Cb clouds (table 2). The base and top will govern the altitude range of the outflow, while the updraught velocity multiplied by the active area will give the vertical mass

transport. Each latitude, in each season, is then designated one Cb-class as the typical. The indicated updraught velocities (Newton, 1966; Atkinson, 1981) refer to the cloud base level.

In addition to the mass flux in Cb clouds, we have also considered a flux due to Cu clouds (see section 4.1.2). The chosen parameter values are influenced by the Chatfield and Crutzen (1984) study, and partly related to the Cb-flux at each latitude.

The resulting vertical mass transport due to convective clouds, representing different latitudes, is shown in table 3.

Cb-class	Average base (m)	Maximum top (m)	Updraught velocity (m/s)
1	700	11000	5
2	1200	14000	7
3	1200	16000	10

Table 2. Values of cloud base, cloud top and updraught velocity (at the cloud base) in the active core, for each Cb-class.

4.1.2 Vertical redistribution by Cb clouds

With the field of vertical mass transport described in section 4.1.1, a redistribution profile was estimated and introduced in the model. A first attempt with only three outflow levels for each cloud-class was soon found to be too crude, giving an unrealistic concentration distribution for the tracers tested. Based on the model by Chatfield and Crutzen (1984), and the theoretical calculations by Gidel (1983), we decided to use the redistribution profiles shown in figure 2.

The outflow from the updraught column should be regarded as successive "detrainment" from the cloud, whereby the air in the cloud is "leaking" out. The profile can also be regarded as the result of using a model cloud being a composite of many real clouds, with varying top heights and thereby varying outflow levels. The major contribution to the profile is given by outflow in the anvil part of fully grown Cb clouds, with cloud tops in the upper troposphere (Newton, 1966; Chatfield and Crutzen, 1984).

These profiles express a bimodal redistribution structure, where the peak at low levels is associated mainly with cumulus clouds. In a convective cloud cluster, the mass transport due to Cu clouds can be regarded as less significant than that due to Cb clouds. However, in a zonal average the great number of low fair-weather cumuli give a considerable redistribution of air, and thereby of tracer mass, within the first 2-3 kilometres of the troposphere (Nitta, 1975; Gidel, 1983; Chatfield and Crutzen, 1984). The explicit value of the average vertical velocity in

Cu, and thereby of the mass outflow, is uncertain and less thoroughly studied than for Cb. The necessity of including this low-level outflow is discussed in the following chapters.

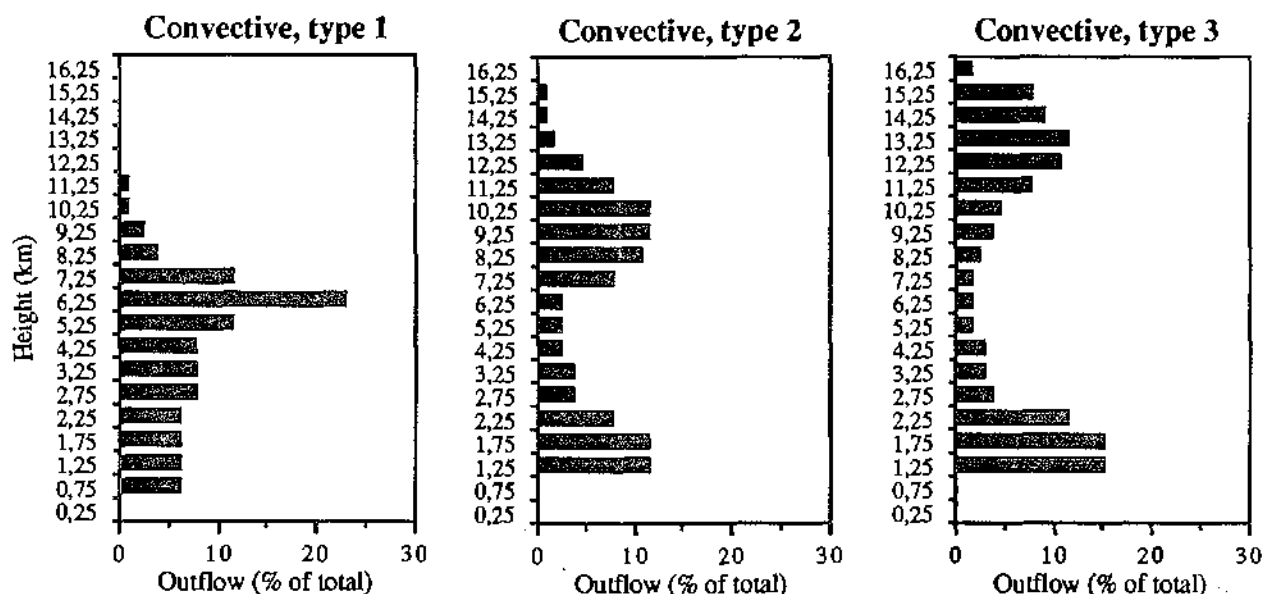


Figure 2. Detrainment distributions showing the outflow at each level, in % of the total vertical mass transport at cloud base, for convective cloud types 1, 2 and 3 respectively.

4.2 Frontal cloud systems

4.2.1 Data

While the vertical transport due to Cb clouds has been discussed in a few other studies (Chatfield and Crutzen, 1984; Dickerson et al., 1986), we have not found any published study concerning the influence by frontal cloud systems on the vertical distribution of a tracer. The vertical lifting velocity is two orders of magnitude smaller than that in convective systems, but the surface area affected by frontal systems is considerably larger. The cloud distribution atlases by Hahn et al. (1982) and Warren et al. (1986) used for convective clouds, also contain statistics about nimbostratus clouds (Ns). However, we regard the Ns-observations as a less good indicator of vertical mass transport than the corresponding Cb-data. In many cases Ns is incorrectly reported in connection with drizzle and light snow-fall, in warm sector situations.

We calculated the vertical mass transport in frontal cloud systems from estimates of the average total area covered by such systems, for each latitudinal band. An average of three different calculations was used, all based on data from Palmén and Newton (1969).

In the first estimate, we considered the simultaneous number of synoptic disturbances on the Northern Hemisphere polar front, getting a yearly average of 16. Each frontal wave

structure was expressed as a 50-100 km wide and an about 2500 km long zone of upgliding, in addition to a low pressure centre with a radius of 200 km. The vertical "upglide" velocity was set to 5 cm/s.

The second estimate was calculated from the mean meridional distribution of cyclones for the Northern Hemisphere, originally from Petterssen (1950). The frequency pattern corresponds to the total area covered by cyclonic disturbances in each latitude band. Since the area thus obtained is larger than the active upgliding area, we now used a mean vertical velocity of 2.5 cm/s in the calculation of the vertical mass transport. In the model runs we then neglected the transport due to frontal clouds outside the region between 35 and 75 degrees of latitude.

The third estimate was based on assumptions of the upgliding velocities in different sections of a frontal system, presented by Palmén and Newton (1969). They indicate wider frontal upgliding zones than we did in our first estimate, giving a higher value of the total vertical mass transport.

In a paper by van Loon (1965) the cyclon frequency on the Southern Hemisphere is stated to be about 15% higher than on the Northern Hemisphere. Our figures have been adjusted according to this.

The resulting vertical mass transport due to both convective and frontal cloud systems is shown in table 3. Season 1 is defined as the months Dec-Feb (DJF), season 2 as Mar-May (MAM) etc.

Latitude	Season 1		Season 2		Season 3		Season 4	
	C	F	C	F	C	F	C	F
NP 90	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
80	1.2	0.0	2.3	0.0	2.4	0.0	3.0	0.0
70	3.9	1.7	4.4	1.7	3.9	0.8	5.0	1.7
60	3.9	5.7	4.7	4.5	5.7	4.1	5.6	4.5
50	2.7	2.2	3.6	3.5	6.1	4.4	3.7	3.5
40	3.5	1.1	4.2	0.7	4.9	0.7	3.9	0.7
30	3.9	0.0	4.5	0.0	10.0	0.0	4.6	0.0
20	3.0	0.0	3.9	0.0	11.4	0.0	9.1	0.0
10	3.9	0.0	10.4	0.0	15.2	0.0	13.0	0.0
0	11.7	0.0	13.3	0.0	12.5	0.0	13.4	0.0
-10	13.9	0.0	14.2	0.0	5.9	0.0	10.7	0.0
-20	11.3	0.0	5.8	0.0	4.4	0.0	4.7	0.0
-30	4.8	0.0	5.5	0.0	3.8	0.0	5.0	0.0
-40	3.9	0.8	5.7	0.8	5.2	0.8	3.8	0.8
-50	5.7	5.8	6.8	5.8	7.8	5.8	5.8	5.8
-60	2.8	2.5	3.0	2.5	4.3	2.5	3.0	2.5
-70	0.4	1.8	0.0	1.8	0.0	1.8	0.0	1.8
-80	0.4	0.0	0.0	0.0	0.0	0.0	0.0	0.0
SP -90	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0

Table 3: Vertical mass transport from the bottom model level, in $\text{g m}^{-2} \text{s}^{-1}$, due to convective cloud systems (C) and frontal cloud systems (F).

One uncertainty in these calculations is the magnitude of the average vertical velocity in the frontal cloud systems (Atkinson, 1981). We discussed to attempt to calculate the vertical velocity from measurements of precipitation production. However, global values are not available and we would expect difficulties in separating convective and frontal precipitation in the mid-latitudes in this way.

As with the convective cloud transport, we consider the latitudinal distribution of the flux due to frontal systems to be fairly well represented by our calculations. The absolute magnitude of the flux is more uncertain. Some tests will be presented later where this magnitude has been changed without changing the latitudinal distribution.

4.2.2 Vertical redistribution by frontal cloud systems

The vertical "detrainment" profile for frontal cloud systems has been defined in a similar way as for convective clouds. In this case the outflow is more concentrated around one major level for each latitude and season, and there is no bimodal profile. The profiles have been subjectively chosen, to cover the variance in tropopause height and includes a less pronounced detrainment in the lower troposphere than in the convective case. The profile for season 2 in latitude 50N is shown in figure 3, as an example.

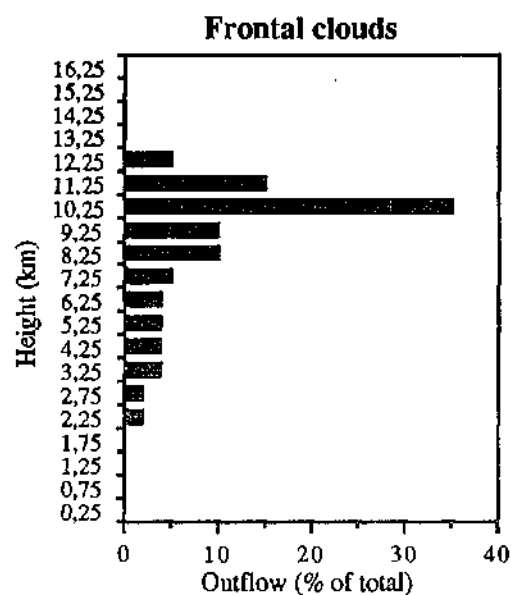


Figure 3. The detrainment distribution profile for the frontal cloud systems in the model, valid for latitude 50°N in season 2 (MAM).

6. Model simulations – results and discussion

6.1 Kr-85

The radioactive isotope Kr-85 of the noble gas krypton has a half-life of 10.8 years and is a waste product of nuclear reprocessing plants. It is continuously being released to the atmosphere since about three decades, the emission being almost totally concentrated to the Northern Hemisphere at latitudes 30-60N. The emission rates from the Soviet Union are not well documented, but can be estimated from the increase of the global inventory. The emission rates and latitudinal distribution of the sources used in our study were taken from Jacob et al. (1987).

Since Kr-85 is an inert gas, with a very low solubility in water, its only sink in the atmosphere can be assumed to be radioactive decay. This means that Kr-85 has the potential of being an ideal tracer of transport processes. Unfortunately, the number of measurements of its vertical distribution is still very limited. The only vertical profile extending through the troposphere, which is known to us, was measured in 1973 by Telegadas and Ferber (1975). It was measured during a period of high annual increase in the release of Kr-85, which might explain a somewhat greater vertical gradient than what could be expected today. As seen in figure 4, the observed profile shows a more pronounced decrease with height than even the model simulation with only diffusive mixing. The results shown in figure 4 indicate a smaller vertical mixing in the observed data set than in our simulation with a pure diffusion exchange. In consequence with our discussion in section 1, we doubt that the measured profile can be regarded as representative for an average atmosphere.

We calculated the average K_{zz} -value that the observed vertical distribution would correspond to, in the layer between 3 and 15 km. From equation (4) we have

$$K_{zz} = \frac{\lambda}{\left(-\frac{1}{z} \ln \frac{c_z}{c_0}\right)^2} \quad (5)$$

By comparison with the Telegadas and Ferber data we get an average K_{zz} of about $1.5 \text{ m}^2\text{s}^{-1}$. This is only about one third (even less if we include the bottom 3 km) of the values used in our model in its diffusion-only version, where K_{zz} is about $5 \text{ m}^2\text{s}^{-1}$, taken as an average for the mid-latitudes (Hidalgo and Crutzen, 1977). This means that the observed profile corresponds to a vertical diffusion much smaller than is normally expected.

In any case, the time-scale of Kr-85 in the troposphere is so long, that it is not a very useful tracer for a tuning of the parameters describing the vertical mixing of the troposphere. However, on the time-scale of transport between the hemispheres, it should give valuable information.

To reduce the execution time of the model, we started the simulations from a homogeneous Kr-85 field, corresponding to an average tropospheric concentration. This value was taken from indicated values in Jacob et al. (1987). One simulation was run for five years starting from 1983, with an initial field of 18 pCi/SCM (pCi/m^3 at standard pressure). The resulting profiles for 1988 are shown in figure 4. Even without the cloud-transport, with the slow process of turbulent diffusion only, the vertical gradient was about half as steep as the profile from Telegadas and Ferber. The profile obtained including the cloud-transport mechanism shows an even smaller vertical gradient.

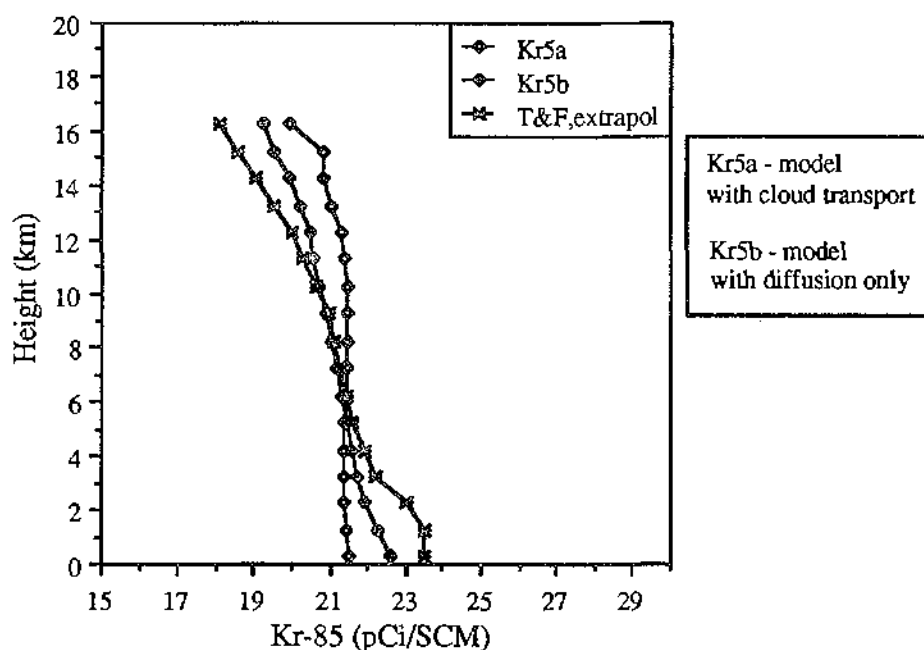


Figure 4. Profiles of Kr-85 concentration from model simulations, with and without the cloud-transport, initiated with a uniform field of 18 pCi/SCM in 1983. The profile from Telegadas and Ferber (1975) has been extrapolated to 1988, to be comparable with the model results.

For comparison with data from Weiss et al. (1983), another simulation was run for three years, starting with a concentration field of 17 pCi/SCM in 1980. It can be mentioned that the vertical profile calculated by Jacob et al. (1987) for October 1983 (our verification time), using a more sophisticated 3D-model, is similar to our model profile.

We have used the Kr-85 simulations to compare the latitudinal profile at ground level with measurements from Weiss et al. (1983). In this way we got a tool to verify the fields of the meridional wind, v , and the horizontal eddy diffusion coefficient, K_{yy} . The values of these parameters in the latitudinal bands surrounding the equator actually define the interhemispheric exchange. Kr-85, with its source confined to one hemisphere only, should be an excellent tracer in this regard. The model profile showed to be in good agreement with the data from Weiss et

al. (figure 5a). Simulations with K_{yy} reduced and increased by a factor of 2 respectively, showed less good similarity with the observed profile (figure 5b). We therefore conclude that the K_{yy} -fields adopted in the model represent a reasonable parameterization of the actual exchange mechanism.

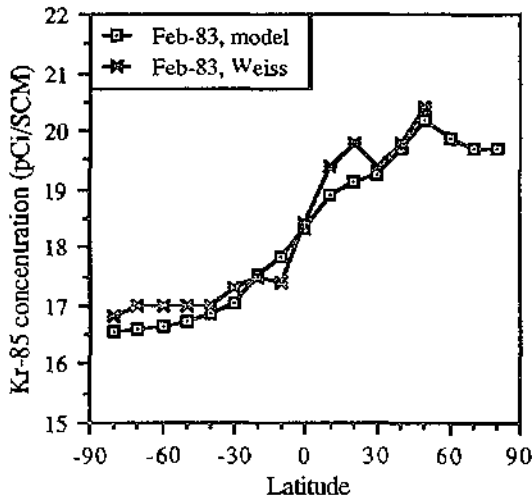


Figure 5a. Latitudinal profiles of Kr-85 surface concentrations; model simulation compared to measurements by Weiss et al. (1983).

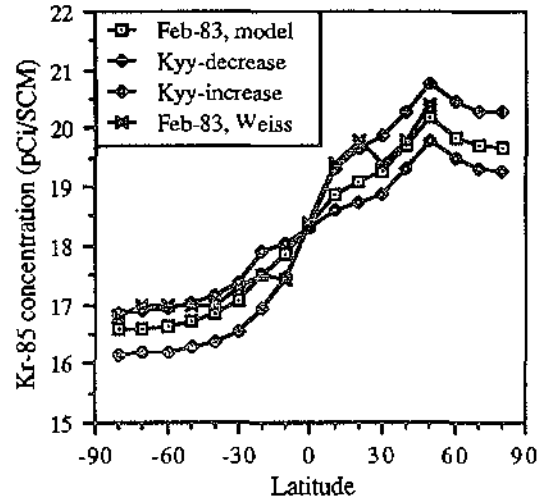


Figure 5b. Model profiles, showing the effect on the distribution due to a change of K_{yy} . The K_{yy} -values were increased and decreased at all levels, between lat 20N and 20S, by a factor 2.

5.2 Rn-222

The radon isotope 222 is a decay product of U-238. Rn-222 is a noble gas which disappears from the atmosphere mainly by radioactive decay with a half-life of 3.8 days. The atmospheric Rn-222 emanates essentially from the soils of the continents. The magnitude of the emission rate has been given different values in the literature. We have run our model using two different emission rates. A "high emission" of $1.2 \text{ atoms cm}^{-2} \text{ s}^{-1}$ (Moore et al., 1973; Turekian et al., 1977), and a "low emission" of $0.72 \text{ atoms cm}^{-2} \text{ s}^{-1}$ (Lambert et al., 1982). For the oceans, we used a degassing rate of the sea surface being 1% of the continental emission (Polian et al., 1986). We have neglected the variation in emission over continents due to different surface material, getting a total emission in each latitude band as a function of ocean/continent distribution only. The resulting vertical concentration profiles for latitude 50N, using a "high emission" input, are shown in figure 6a, together with a verification profile with data from Moore et al. (1973). Unfortunately, most other experimental data on Rn-222 show concentrations in low levels only. To avoid scaling problems in the verification, we used the

"high emission" rate in all of the following model calculations. Thus, we got a higher yearly input and global burden of Rn-222 than stated in more recent publications (Lambert et al., 1982, Polian et al., 1986). As expected, the model simulations with a "low emission" input gave concentration values about 40% lower at all heights (corresponding to the 0.72/1.2 emission ratio).

From figure 6a it is obvious that the model underestimates the concentration in the lower troposphere. The figure also shows the significant shortcoming of a pure diffusion model, with its much too low concentration values in the upper troposphere.

Because of the distinct difference in emission rate between land and oceans, the measured concentration of Rn-222 is highly dependent on where the sampling is done. As described in section 3.3, we also used a modified version of the model, that took into account the different emission rates and exposure times. With a half-life of 3.8 days, the concentration in an air mass moving out over an ocean is gradually decreasing, until after 5-10 days the impact from continental emission is almost lost. In the same way, measurements taken about 3-5 days "inland" over a continent, should be regarded as a sample from an almost pure continental air mass, although a steady state is not yet obtained. This points at one of the limitations of a two-dimensional model, which we have tried to reduce with the current modification.

Since our verification profile from Moore et al. (1973) consists of data from the central USA, it is more appropriate to compare it with the result from the continental mode of our model. Figure 6b shows the profiles obtained from this model for season 1, over the central "USA"-continent and over the central "Pacific"-ocean respectively, compared to the original model with averaged emission and to the verification profile. The continental profile shows a good agreement at 5-10 km altitude, while the concentrations above (below) this part of the troposphere are somewhat high (low). The reason for this is discussed below.

The emission-separating model was also used to test the influence of changes in some of the model parameters. The following model runs were carried out:

1. Differentiated emission, full cloud-transport over both ocean and continent (figure 6b).
2. As in 1, but with a reduction of the vertical mass transport from the ground level. Reductions to 75% and 30% of the original values were tested (figure 6c).
3. As in 1, but using a K_{zz} -value of $1.0 \text{ m}^2 \text{ s}^{-1}$ at all levels (figure 6d).

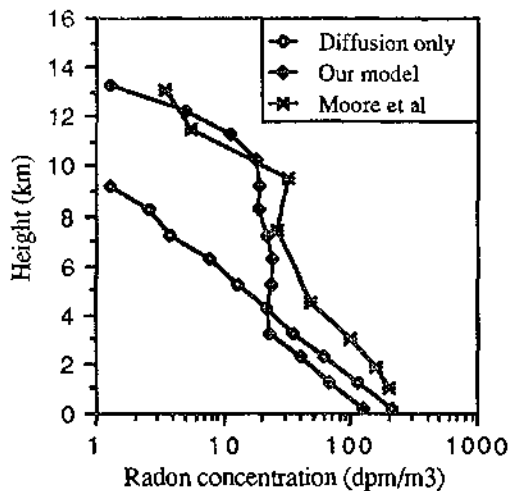


Figure 6a. Model profiles of Radon concentration, with the cloud transport parameter included and without it (diffusion only). The profile measured by Moore et al. (1973) is added for comparison.

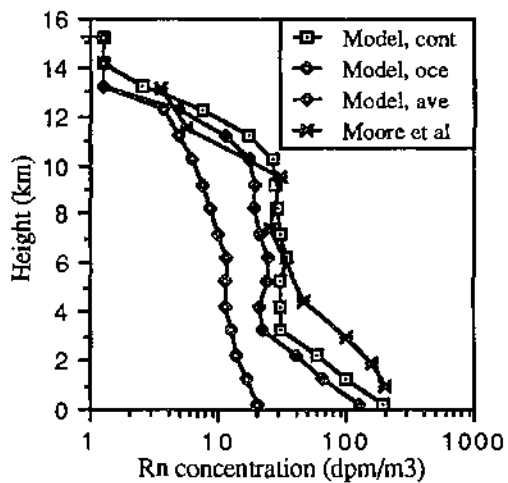


Figure 6b. Result of model run using different emission rates for continents and oceans. Also shown are the result from the zonally averaged model and the verification profile.

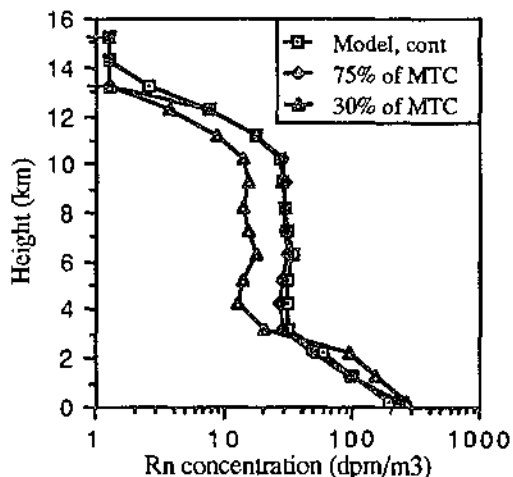


Figure 6c. Model profiles showing the effect of a reduction in the total mass transport by clouds (MTC) from the bottom level. Results are shown for a reduction to 75% and 30% of the "normal" MTC values.

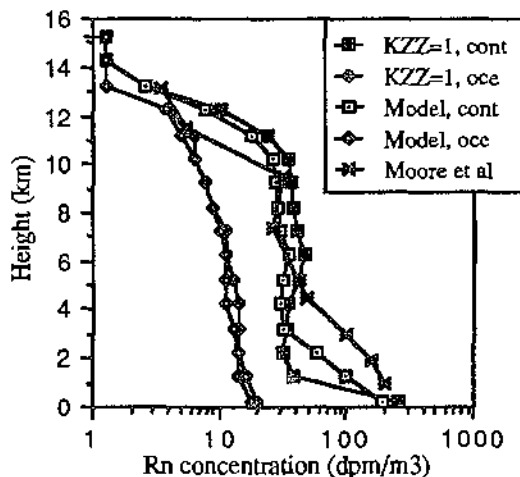


Figure 6d. Model profiles showing the effect of a reduced K_{zz} -field below the lowest detrainment level, for the continental and ocean modes of the model, respectively.

It was a bit surprising to find that the concentration profile showed very little sensitivity to a reduction to 75% of the mass transport from the ground level. Even with a reduction to 30%, the profile was much closer to the measured data than the diffusion-only profile. This shows that while the cloud-transport function as such is extremely important for a fair vertical distribution, the estimate of the exact magnitude of the mass transport does not seem to be a very crucial parameter for Rn-222 (in an averaged twodimensional model like this). With Kr-85 (half-life 10.8 years) the effect of a reduction to 30% was hardly noticeable. On the other hand, a simulation of tracers with a turnover time close to the time-scale of the cloud-transport process, is likely to be much more sensitive to the magnitude of the flux. No attempts to run the model with such tracers have been made, since we regard the output from a zonally averaged model totally unrealistic for tracers with time-scales less than a day.

A simulation with a uniform K_{zz} -field of $1.0 \text{ m}^2 \text{ s}^{-1}$, replacing our normal low-level K_{zz} of $5\text{-}10 \text{ m}^2 \text{ s}^{-1}$, was carried out to see the sensitivity to a reduced K_{zz} -value in the sub-cloud levels, while retaining the full cloud-transport function. As expected, the effect was a much steeper concentration gradient in these levels and an increase of the concentration at all levels above 3 km. A substantial *increase* of the K_{zz} -field at low levels, which would reduce the difference between our model profile and the verification profile, was not considered. The K_{zz} -values would then be much greater than normally expected (Jacobi and André, 1963).

The cloud transport, as described in this model, does not just represent a very efficient eddy diffusion. It may also produce profiles with increasing concentration with height, similar to what is measured in the atmosphere. However, from figures 6a-6d it can be seen that the model results show somewhat high concentrations at high levels, compared with the verification profile, while at low levels the situation is opposite (similar results were obtained for SO_2 , see section 5.3). We have found, as was discussed above, that the concentration gradient is not very sensitive to the absolute magnitude of the cloud-transport flow. Our conclusion is that the most crucial factor for the vertical distribution of the tracer is the relative vertical distribution of the outflow. While initially having regarded the cumulonimbus updraughts as the most important factor, we now assume that the transport due to ordinary cumulus clouds plays an essential part in the total mass redistribution. The much less powerful "vacuum-cleaner" effect of each Cu could be compensated for by the large number of clouds. The average Cu cloud, including the numerous trade-wind cumuli, would give an effective redistribution acting in the 1-4 km height range (Nitta, 1975). This may still be underestimated in our outflow profiles in figure 2.

One problem in the verification is the limitation in the two-dimensional model concept itself. While the model uses the averaged values of the transport parameters to give a certain profile for a certain latitude band, the measured data used as verification are naturally momentarily taken (in space and time) in specific synoptic situations. A fair comparison between the real atmosphere and its model image is thereby hard to achieve, especially for tracers with a short turnover time in the atmosphere.

A part in our cloud parameterization scheme, which could show to be important in the relative distribution between the lower and higher troposphere, is the vertical extension of the layer from which the cloud-transported air emanates. In the simulations presented in this report, the layer was confined to the bottom model grid box, i.e. the bottom 500 m of the model atmosphere. We assume that an extended boundary layer, which would imply a lower average concentration in the air volume that represents the cloud-transport flux and thereby in the air that is detained at higher levels, would increase the vertical concentration gradient. The importance of the boundary layer extension in this respect will be investigated in a further study.

5.3 SO₂

In contrast to radon and krypton, sulfur dioxide provides an opportunity for global scale verification. Sulfur dioxide is however a more complex tracer than radon and krypton – both sources and sinks being more uncertain (Georgii, 1978). The man-made emission of SO₂ was taken as 80 Tg S yr⁻¹ (Rodhe and Isaksen, 1980), while the emission from natural sources was taken as another 80 Tg S yr⁻¹ (Andreae, 1985). The latitudinal distribution of these emissions is showed in table 4 and in figure 7. All natural sulfur emissions were assumed to occur in the form of SO₂.

The emission from biomass burning has been distributed according to the continent fraction at each latitude. The emission from volcanos is irregular, but has been taken as the average value in time (12 Tg yr⁻¹) evenly distributed between latitudes 60N and 50S.

Scavenging efficiencies for reversible as well as irreversible scavenging were taken from Isaksen and Rodhe (1978). A removal of 15% of the man-made emission was made, to account for the immediate dry deposition in the source area (Rodhe and Isaksen, 1980). As was discussed in section 3.5, a further dry deposition in the boundary layer was calculated in the model, as well as an enhanced scavenging in connection with the cloud-transport processes.

The gas phase conversion of SO₂ to sulfate through the reaction with OH radicals (Rodhe et al., 1981) is modelled using a very simple approach. We have regarded the average turnover time for SO₂ of 7 days above 6 km (Rodhe and Isaksen, 1980) as a result of reaction with OH radicals. The corresponding removal rate is introduced everywhere in the model domain.

Latitude		Man-made	Natural			Total
		SO ₂	DMS	Biomass	Volcanic	
NH	90	0.0	0.0	0.0	0.0	0.0
	80	0.0	0.1	0.0	0.0	0.1
	70	0.0	0.3	0.0	0.0	0.3
	60	9.1	0.4	0.9	1.0	11.4
	50	22.0	0.7	1.8	1.0	25.5
	40	22.8	2.5	2.4	1.0	28.7
	30	13.9	2.9	4.8	1.0	22.6
	20	5.0	3.6	4.2	1.0	13.8
	10	1.8	4.1	3.3	1.0	10.2
	0	0.9	4.2	3.3	1.0	9.4
	-10	0.9	4.2	3.0	1.0	9.1
	-20	1.7	4.0	3.0	1.0	9.7
	-30	1.5	3.9	2.1	1.0	8.5
	-40	0.7	4.0	0.6	1.0	6.3
	-50	0.3	1.5	0.3	1.0	3.1
	-60	0.0	1.2	0.0	0.0	1.2
	-70	0.0	0.5	0.0	0.0	0.5
	-80	0.0	0.0	0.0	0.0	0.0
SH	-90	0.0	0.0	0.0	0.0	0.0

Table 4: Sulfur emission figures used as input in the model calculations, given in Tg S yr⁻¹, for each latitudinal band respectively.

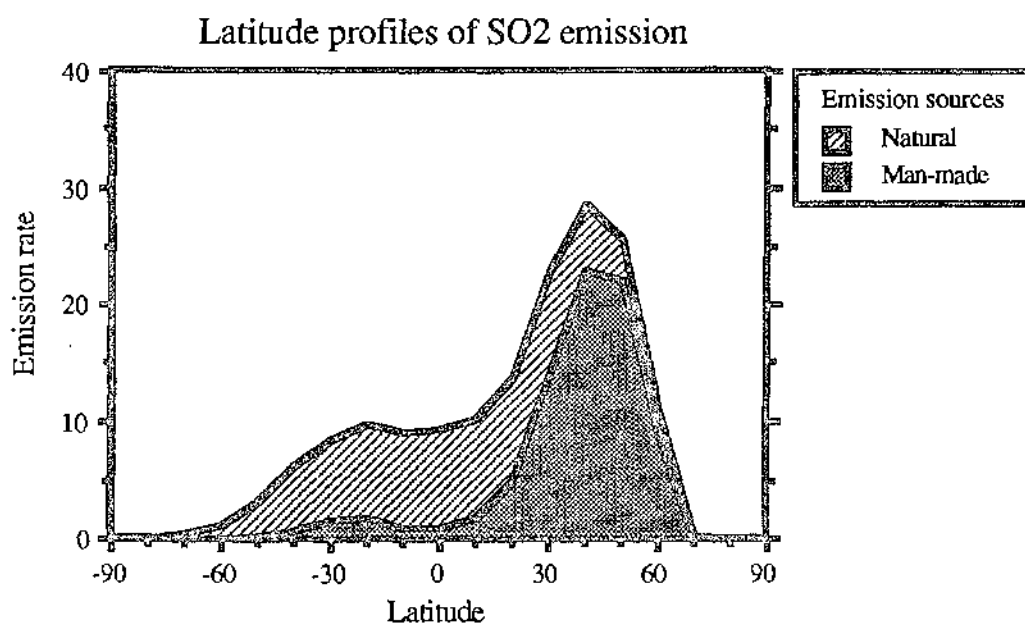


Figure 7: Latitudinal profile of man-made and natural sulfur emission. Units: Tg S yr⁻¹ per 10° latitude band.

The model results for SO₂ are presented in figures 7a-7i. Simulations were carried out for all four seasons, using the total emission and both convective and frontal cloud-transport as model input (figures 7a-7d). We also ran the model separating the cloud-transport process into convective and frontal cloud transport (7e-7f), as well as separating man-made and natural SO₂ emissions (7g-7h). The result obtained without any cloud-transport process is shown for comparison (7i).

The model results for the end of season 2, i.e. the shift May/June, are compared with a comprehensive set of aircraft measurements presented by Ockelmann (1988). A data collection campaign was carried out with aircraft measurements between lat 70N and lat 60S, in June 1987. The resulting concentration distribution is shown in figure 8. Measurements from western Europe, presented by Meixner (1984), show similar profiles, with layers of increasing mixing ratio with height in the upper troposphere.

With the following two aspects in mind, we find the model results to be encouraging:

1. The emission of SO₂ from natural sources is still uncertain. Some previous estimates of this emission are substantially smaller than that used in our simulations (Andreae, 1985).

2. The concentration of sulfur dioxide, having an average turnover time of 2-4 days in the troposphere (Rodhe and Isaksen, 1980), is expected to vary with the synoptic weather situation and the emission situation along the aircraft's sampling track. Thus, the observed values need not be quite representative of zonal averages.

The model with only a vertical eddy diffusion can not simulate the observed increased mixing ratio with height (cf. figure 7i). The introduction of explicit cloud transport in the model makes the simulations much more realistic. The importance not only of convective clouds, but also of frontal cloud systems, is indicated by a comparison between figures 7b, 7e and 8. If the transport by frontal clouds is excluded (figure 7e), the agreement with observations (figure 8) is less good than when it is included (figure 7b).

SO₂ MIXING RATIO IN PPT(V); SEASON 1

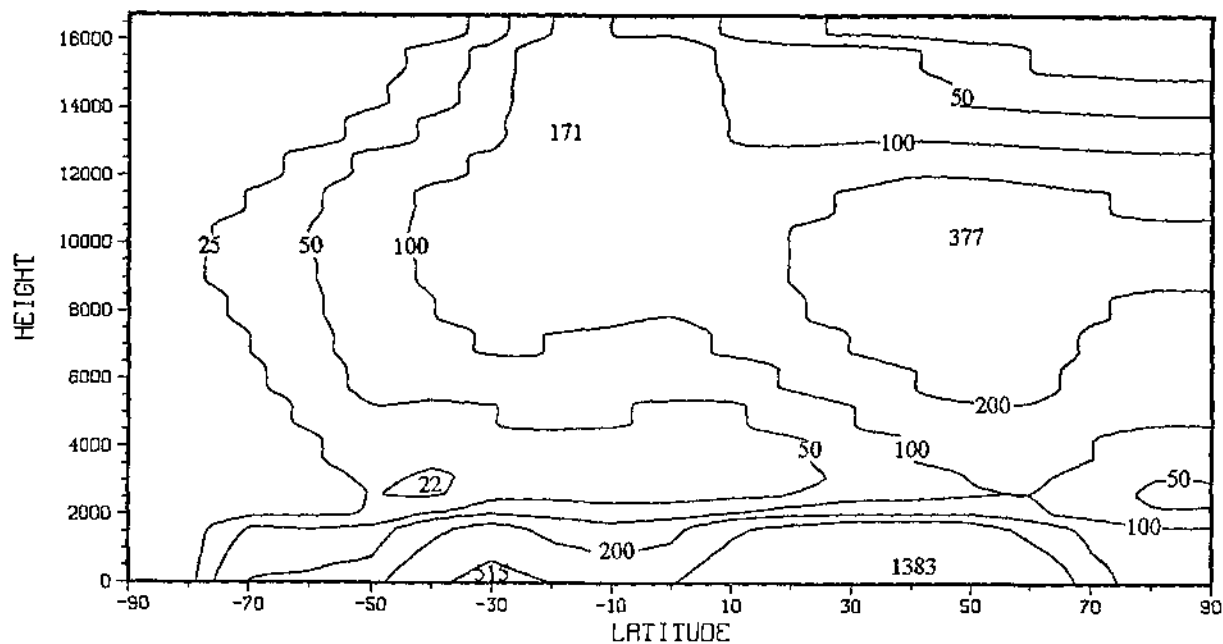


Figure 7a. Simulated global distribution of SO₂ (zonally averaged), in ppt(v), for season 1 (DJF). Negative values of latitude refer to the Southern Hemisphere.

SO₂ MIXING RATIO IN PPT(V); SEASON 2

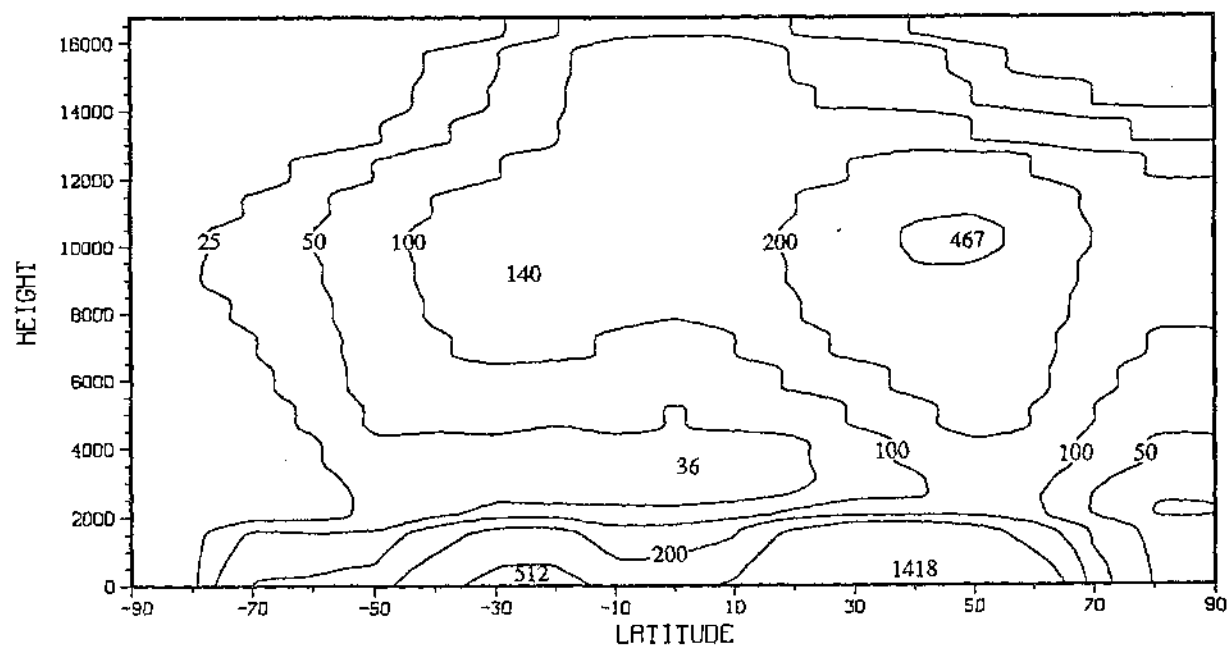


Figure 7b. As in figure 7a, but for season 2 (MAM).

SO2 MIXING RATIO IN PPT(V); SEASON 3

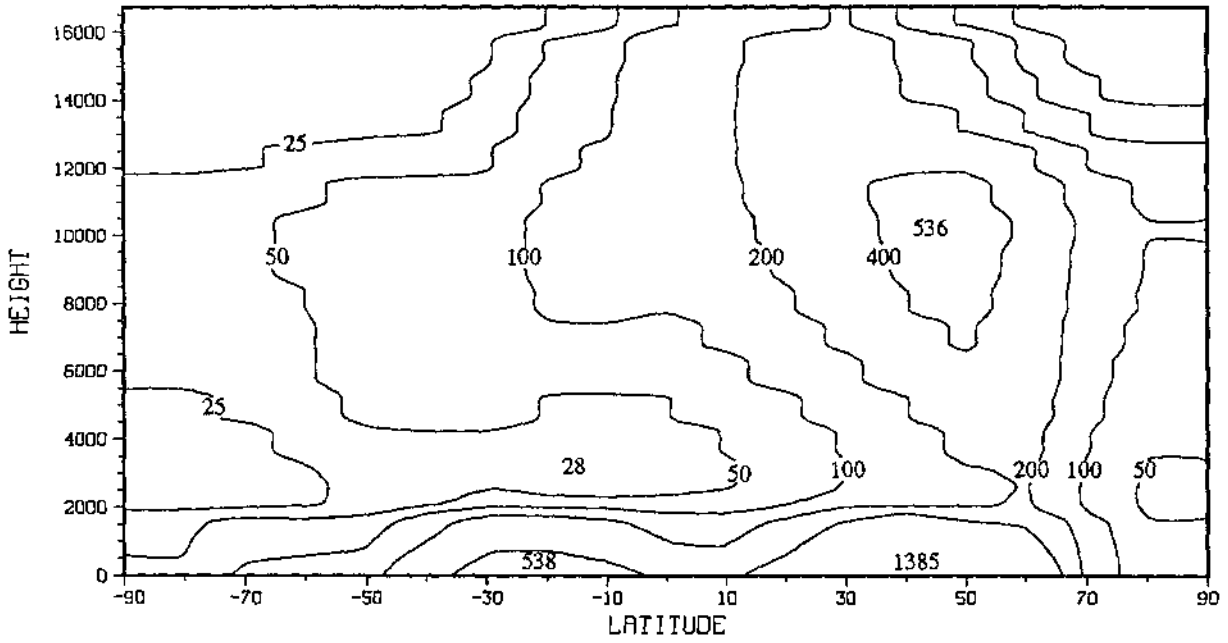


Figure 7c. As in figure 7a, but for season 3 (JJA).

SO2 MIXING RATIO IN PPT(V); SEASON 4

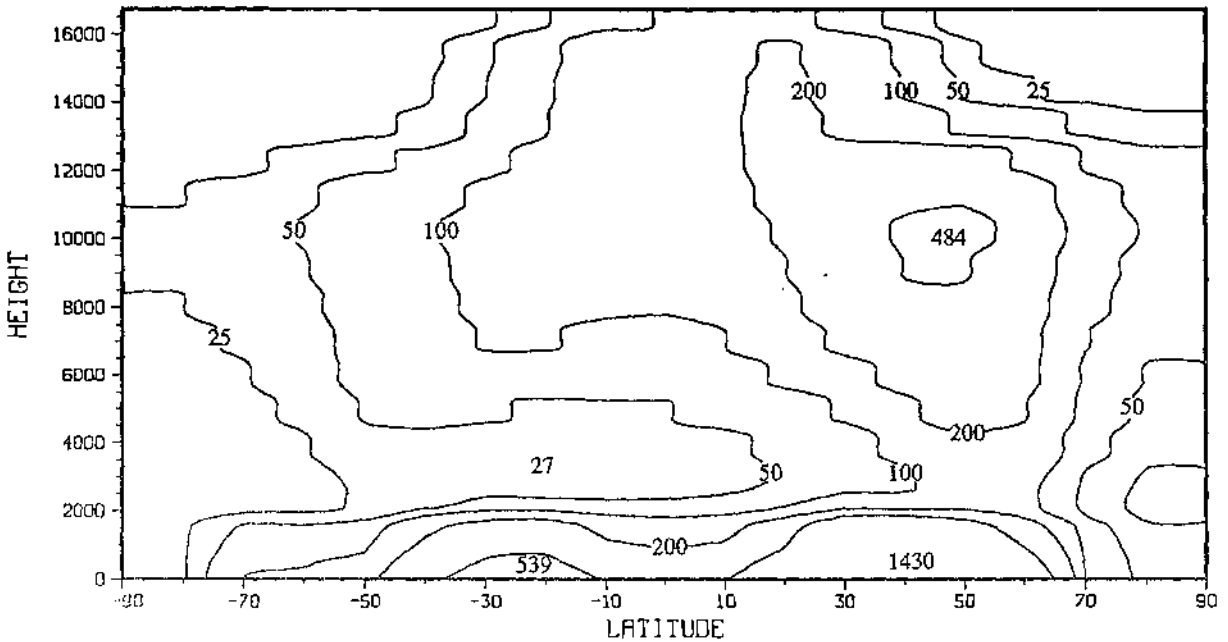


Figure 7d. As in figure 7a, but for season 4 (SON).

SO₂ MIXING RATIO IN PPT(V); SEASON 2
NO CYCLONIC CLOUD TRANSPORT

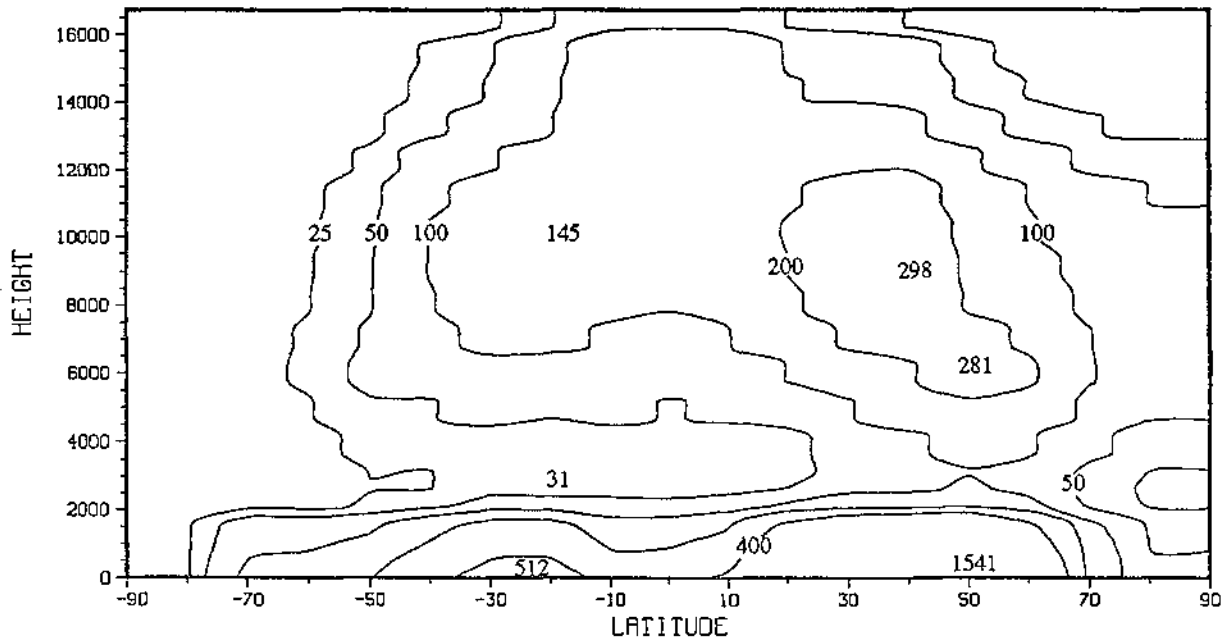


Figure 7e. As in figure 7a, but for season 2 and excluding the transport by frontal, or cyclonic, cloud systems.

SO₂ MIXING RATIO IN PPT(V); SEASON 2
NO CONVECTIVE CLOUD TRANSPORT

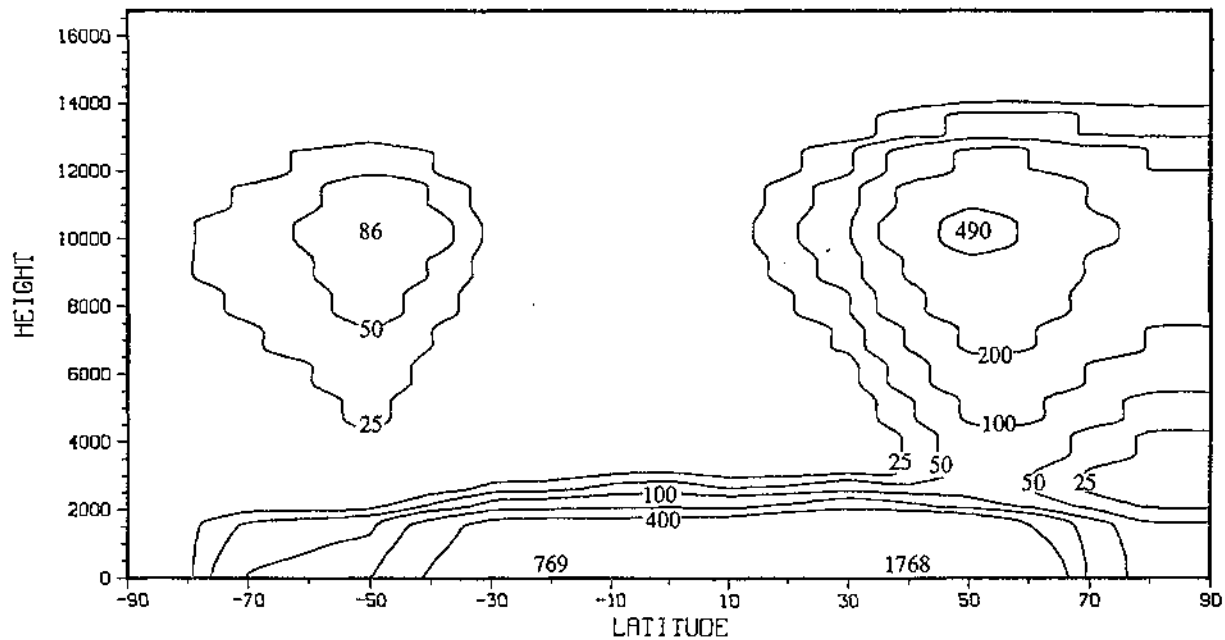


Figure 7f. As in figure 7a, but for season 2 and excluding the transport by convective cloud systems.

S02 MIXING RATIO IN PPT(V); SEASON 2
MAN-MADE SOURCES ONLY

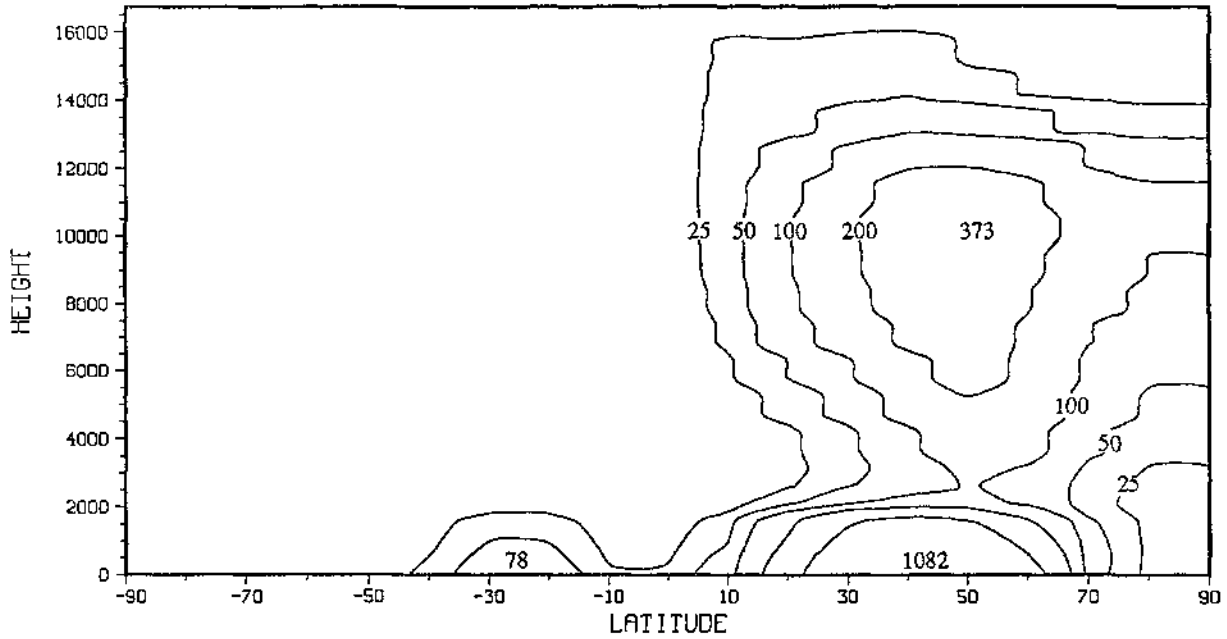


Figure 7g. As in figure 7a, but for season 2 and excluding the emission from natural sources.

S02 MIXING RATIO IN PPT(V); SEASON 2
NATURAL SOURCES ONLY

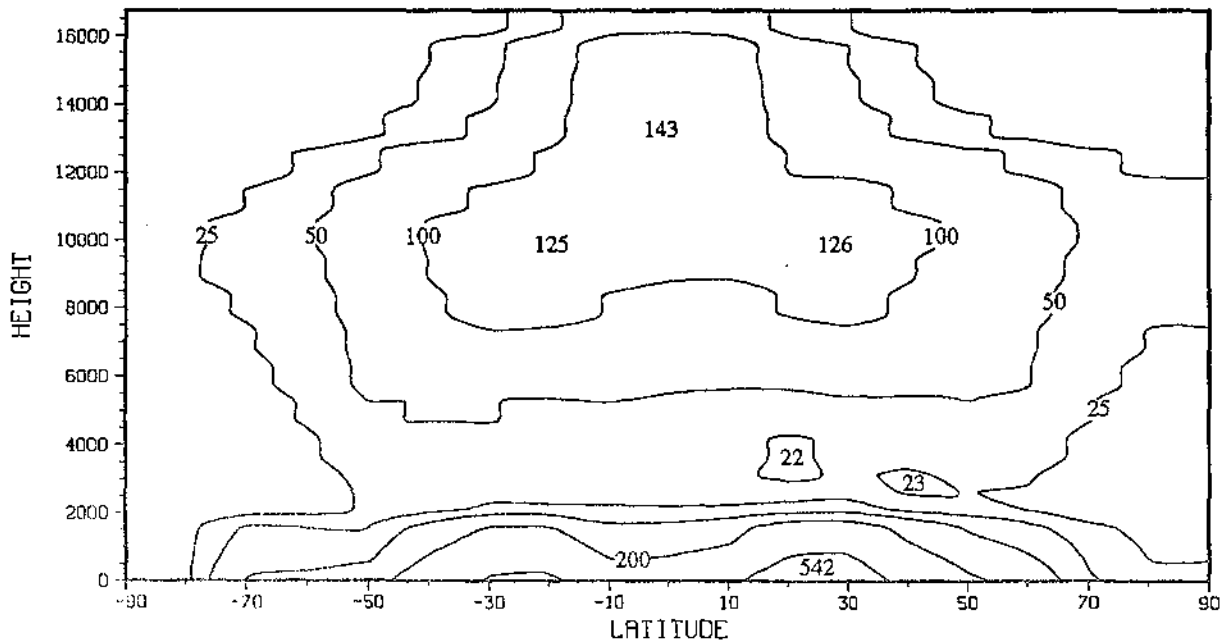


Figure 7h. As in figure 7a, but for season 2 and excluding the emission from man-made sources.

SO₂ MIXING RATIO IN PPT(V); SEASON 2
NO CLOUD TRANSPORT, DIFFUSION ONLY

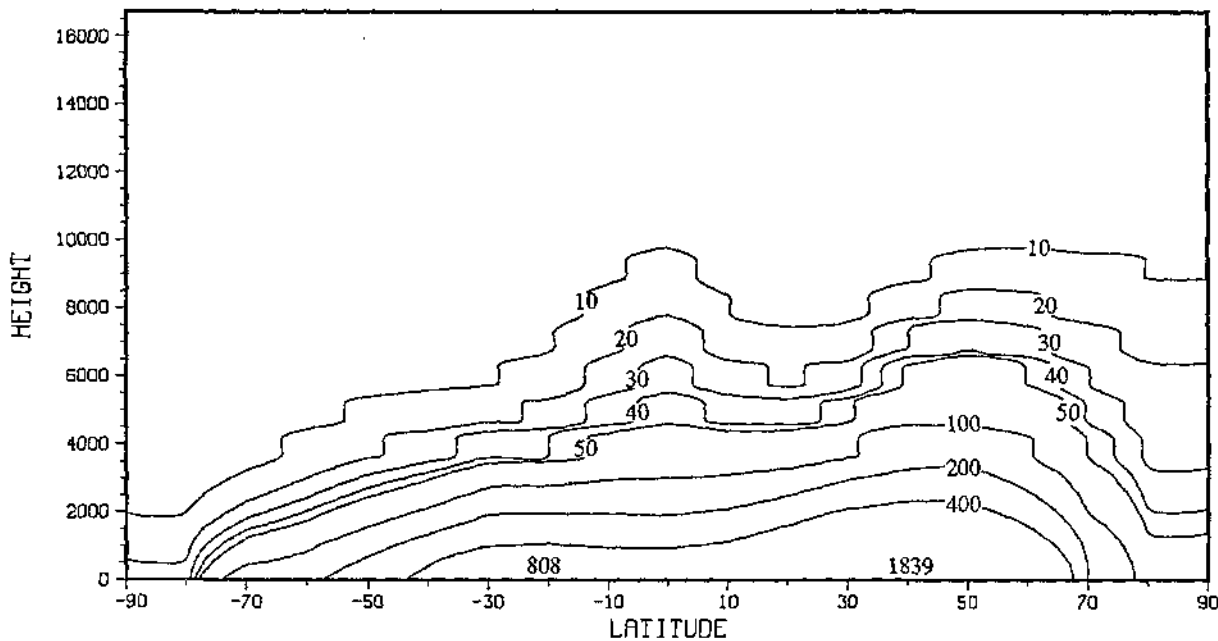


Figure 7i. Simulated global distribution of SO₂ for season 2, with the transport parameterized with diffusion only.

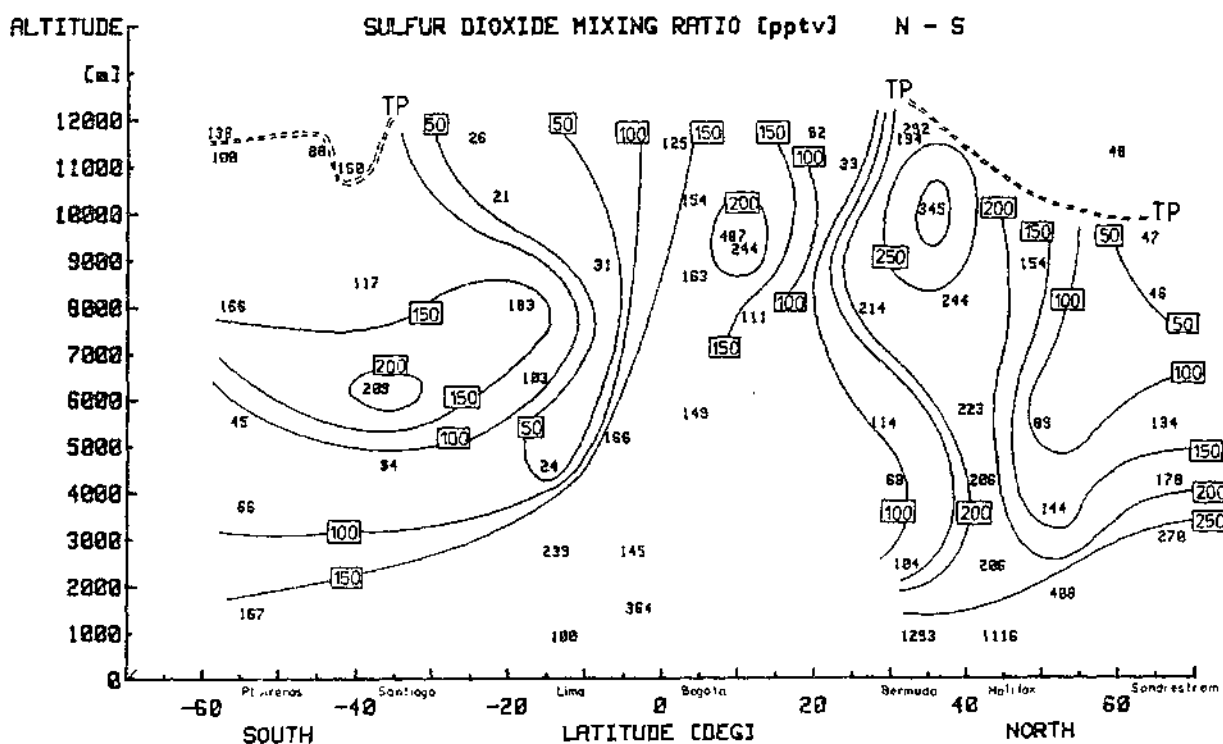


Figure 8. Global distribution of SO₂, from aircraft measurements in June, 1987. Figure from Ockelmann (1988).

8. Conclusions

Model simulations using lifting of low level air in cloud systems, and compensating subsidence in the surrounding air, as the main process of vertical redistribution, are found to give a realistic result of vertical as well as latitudinal tracer profiles. The following may be concluded:

1. Simulations using only eddy diffusion parameterization of vertical transport do not produce realistic results for trace species with a turnover time of the order of days.

2. A combination of vertical transport by convective and frontal cloud systems gives an improved result, compared to using convective transport only.

3. The cloud-transport processes result in an increased contact between the tracer and the cloud water. A further study of the resulting scavenging efficiency is needed.

4. When studying a trace species with a continent/ocean difference in emission rates and a short turnover time, the use of a modified two-dimensional model as outlined in this report could be an advantage. Such a model will enable more realistic comparisons between model results and observations.

5. In the present model, the entrainment of air into the cloud systems occurs entirely in the lowest 500 m. A possible improvement of the model would be to extend the entrainment layer up to 1-2 km, particularly in the tropics.

9. Acknowledgements

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10	11 Contract number	12 Starting year	13 Finishing year	14 MI Report number	
15 Sponsoring organization Swedish Air Force					
16 Title and subtitle of project or report Global vertical mass transport by clouds - a two-dimensional model study					
17 Project leader/Author(s) The author					
18 Abstract (goal, method, technique, result etc.) A two-dimensional global dispersion model, where vertical transport in the troposphere carried out by convective as well as by frontal cloud systems is explicitly treated, is developed from an existing diffusion model. A parameterization scheme for the cloud transport, based on global cloud statistics, is presented. The model has been tested by using Kr-85, Rn-222 and SO ₂ as tracers. Comparisons have been made with observed distributions of these tracers, but also with model results without the cloud transport, using eddy diffusion as the primary means of vertical transport. The model results indicate that for trace species with a turnover time of days to weeks, the introduction of cloud transport gives much more realistic simulations of their vertical distribution. Layers of increased mixing ratio with height, which can be found in real atmosphere, are reproduced in our cloud-transport model profiles, but can never be simulated with a pure eddy diffusion model. The horizontal transport in the model, by advection and eddy diffusion gives a realistic distribution between the hemispheres of the more longlived tracer (Kr-85). A combination of vertical transport by convective and frontal cloud systems is shown to improve the model simulations, compared to limiting it to convective transport only. The importance of including cumulus clouds in the convective transport scheme, in addition to the efficient transport by cumulonimbus clouds, is discussed. The model results are shown to be more sensitive to					
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the vertical detrainment distribution profile than to the absolute magnitude of the vertical mass transport.

The scavenging processes for SO_2 are parameterized without the introduction of detailed chemistry. An enhanced removal, due to the increased contact with droplets in the in-cloud lifting process, is introduced in the model.