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A GLOBAL NUMERICAL STUDY OF RADON^{222} AND LEAD^{210} IN THE ATMOSPHERE USING THE AES AND YORK UNIVERSITY CDT GENERAL CIRCULATION MODEL, (AYCG).

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Abstract.

The Canadian Climate Centre (CCC) GCM has been modified to allow its use for studies in atmospheric chemistry. The initial experiments reported here have been run to test and allow sensitivity studies of the new transport module. The impact of different types of parameterization for the convective mixing have been studied based on the large scale evolution of Rn^{222} and Pb^{210}. Preliminary results have shown that the use of a scheme, which mixes unstable columns over a very short time scale, produces a global distribution of lead that agrees in some aspects with observations. The local impact of different mixing schemes on a short lived tracer like the radon is very important.

1. Introduction.

Recently the modeling of chemistry has moved to the global scale, permitting the assessment of the multitude of chemical reactions within a global atmospheric model. The model described herein, the AES and York University Chemistry and General Circulation model, is a version of the Canadian Climate Centre General Circulation Model, (GCM). The model used is the operational version of the CCC spectral GCM (McFarlane et al, 1992). Initial experiments were aimed at examining the use of the spectral method for tracer transport and assessing the transport module. This module includes, multi-tracer advection, convective and boundary layer mixing, wet deposition, vertical and horizontal diffusion. This work describes some experiments on radon and lead tracers chosen to assess the part played by convective mixing in the transport of species within the GCM.

The current results are for T32 resolution and the model time step is 20 minutes. At this resolution the model resolves synoptic and planetary scale motions, however subgrid meteorology which impacts on these scales are absent.

We have chosen Rn^{222} as being representative of species with short chemical lifetimes such as isoprene and propane, and Pb^{210} as being representative of a longer lived species, *e.g.* CO and O_3. Very simplistically we are equating Rn^{222} with a precursor chemical like NO_x or hydrocarbons which have short lifetimes, while the lead is equated to the longer lived chemical products, whose presence in the upper troposphere can only be achieved if its precursors are transported rapidly from their surface source regions. In addition, radon tests the impact of a simple surface source while the Pb^{210} production is distributed throughout the troposphere.

2. The Radon Experiment.

As a first test of the model and transport module, a number of paired Rn^{222} and Pb^{210} like tracers were used. This allowed several convective mixing schemes to be considered and a general assessment to be made of the overall use of the spectral GCM as an online tracer advection model. A test of the mixing, rainout and spectral advection plus a first check on mass conservation and other numerical problems associated with sharp tracer gradients modeled in a spectral model was thus possible.

Initially, radon was distributed uniformly in the horizontal, with a fixed vertical profile, decreasing exponentially with height. The ground level value of 100 pCi/m^3 STP was chosen based upon typical observed surface radon measurements. Lead was set as a low background which would be dominated rapidly by the radon decay source. Due to its short lifetime, the radon rapidly reached a global balance between the model source and decay within the first month and was largely unaffected by the initial state thereafter. For the lead, a global mass equilibrium state was not expected within the first several months due to its longer tropospheric lifetime. Since the radon surface flux is confined to the continent, the sharp boundaries and the hemispheric gradient, due to the source distribution, provide test problems for the model transport and analysis. The radon source was modeled by assuming a constant surface flux of 1 atom/sec/cm^2 from all non-ice covered land on the globe. The radon sink due to radioactive decay was obtained using a tendency based on the tracers 5.5 day e-folding time. Thus excluding the transport, the budget for Rn^{222} is described in equation 1,

$$\frac{\delta \chi_r}{\delta t} = -\frac{\chi_r}{\tau} + F_g \qquad (1)$$

where χ_r= Rn^{222} tracer mass mixing ratio, τ= radon e-folding time, and F_g= surface flux of radon. The Pb^{210} was assumed as the only decay product of the radon so that the sink of radon in molecules was the same as the source of Pb^{210} in molecules. Its sink was modeled using a simple rainout scheme, proportional to the local column rainfall amount and the local tracer mixing ratio (Mahlman and Moxim, 1978). Thus the budget for Pb^{210} is;

$$\frac{\delta \chi_p}{\delta t} = \frac{\chi_r M_p}{\tau M_r} - \frac{\chi_p}{\tau_{ro}} \qquad (2)$$

where χ_p= Pb^{210} tracer mass mixing ratio, M_p = molecular mass of Pb^{210}, M_r = molecular mass of Rn^{222} and τ_{ro} = rainout e-folding lifetime as defined below as;

$$\tau_{ro} = \frac{\tau_w(k). < P >}{P} \qquad (3)$$

where τ_w= wet deposition lifetime (as a function of height, k,), P = local average precipitation amount (mms^{-1}), and $< P >$ = the global average precipitation amount (mms^{-1}).

Convective mixing of tracer from the boundary layer into the free troposphere has been identified as an important component in a complete transport description of the troposphere. Tracer mixing schemes that have been used for radon include those of Jacob and Prather (1990), Feichter and Crutzen (1990) and Brost and Chatfield (1989). These schemes vary in complexity with a balance being struck between detailed model description and expense. However due to the large scale of the GCM's horizontal grid, parameterizations are, at best, highly approximate. Such a position poses a serious difficulty in global modeling and requires careful assessment and observational basis before interpretation can be considered valid.

It was decided to test two schemes in parallel using fundamentally different transport assumptions. The two basic schemes used are; a) a direct relocation scheme and b) a diffusive scheme. The direct relocation scheme is based on the scheme described in Prather et al.(1987), which assumes that within a model grid box a fraction of tracer mass is directly advected from the convection base to the top of the convectively unstable column. Continuity then requires an instantaneous subsidence to move tracer from the top down to the next lower box, throughout the column. Thus total mass is conserved in the column while redistributing tracer. The diffusive scheme was based upon the convective adjustment used in the GCM for convective parameterization of heat and moisture and in this sense, it is internally consistent with the dynamics. The diffusive scheme is different from that of the direct relocation, in that it tests adjacent points in a column and mixes between them if an instability exists. This process is done from the ground up so it is possible for tracer to slowly 'diffuse' up through the column. The two schemes are fundamentally different and will have significantly different impacts upon the transport of tracers with different lifetimes.

3. Results.

Due to its relatively short life time, the radon is on average mostly concentrated over the land area. Figure 1 shows the mass mixing ratio of radon at the surface, averaged over the second month, on which can be seen many features associated with the large scale circulation. In the tropics, zonal transport produced by the trade winds is evident as well as the impact of the tropical circulation due to the presence of the high pressure belt in the southern hemisphere. The persistence of such an anticyclonic circulation over the tropical ocean produces surface winds following the west coast of Africa and south America causing a strong gradient on the edge of both continents. Average surface concentrations of radon obtained over most of the northern continents range from 100-300 pCi/m^3 which fall in the range of what is currently observed (Liu et al.,1984). However, spatial and temporal variability of radon is such that comparison between observation and model results over specific regions would necessitate further analysis (Feichter, 1988; Jacob, 1990).

At mid-and high-latitude in the winter season, the impact of convective mixing is weak no matter which scheme is used. Tropical regions are however strongly affected by the choice of parameterization and show highly localized spatial and temporal variations. To illustrate the differences between the schemes, Figures 2b and c show time snap-shots of the radon distributions for the two different parameterizations, for 71°West at model day 33, 00:00 GMT. Figure 2a shows, for comparison, the result achieved when no convective mixing is applied. The 'diffusive' and non-convective mixing variants are very similar with only a slight spread of tracer due to diffusion from the tracer plumes generated by synoptic scale ascent. This indicates that the 'diffusive' technique spreads tracer slowly to high altitudes but has little power to inject tracer higher due to the relatively short lifetime of radon. The 'relocation' scheme however is strikingly different producing a high level tracer maxima in the tropics right up to the tropopause and lower stratosphere. In addition, without convective mixing the model tracer (due to the source at the ground) builds up strong gradients in the lowest levels (Figure 2a), producing numerical problems at land-sea boundaries and above continents.

The mean residence time of lead in the stratosphere is considerably higher than in the troposphere, where the lead is washed out by precipitation. Previous studies have shown that the quantity of lead in the stratosphere is a significant fraction of the total (Moore, 1973) and could correspond to 33% of the global mass of lead in the atmosphere (Lambert, 1982). Our results show that such a reservoir can be approached by using the relocation technique

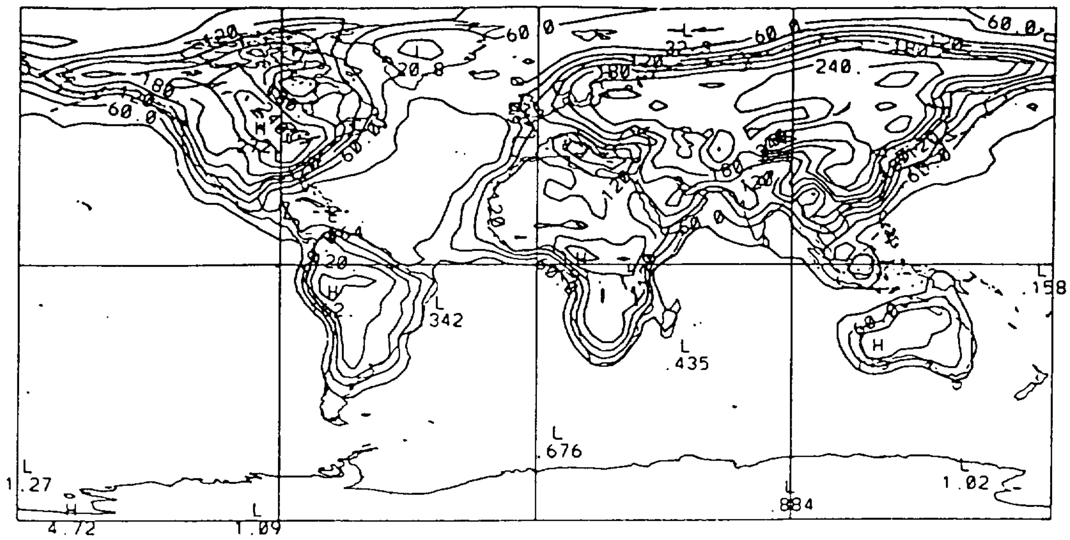

Figure 1 : Radon^{222} surface mass mixing ratios, (pci/m^3 at STP), for Radon variant ii) 'Diffusive' convective mixing, Monthly averaged data for February.

which gives a stratospheric mass fraction of 25% at quasi-steady state. The fraction given by the diffusive mixing scheme is of the order of 5-10% which is significantly lower.

In spite of the slowness of the diffusive scheme, it still contributes to the transport of species into the higher troposphere. Budget analyses show in fact that close to 30% of the transport of radon to the tropopause could be attributed to this process. To allow the tracer to go further into the stratosphere, an additional type of parameterization could be needed in that case to represent the exchange of tracer that occurs across the tropopause.

Part of the aim of these experiments was to determine the capabilities and limitations of the spectral technique. Initial model experiments reveal the model conserves mass well, giving a maximum global mass variation of 0.1 percent in 2 months. Running at T32 resolution the radon and lead tracers together with the convective mixing schemes give rise to strong gradients locally, regionally and globally. Despite the noisy nature of the convection and the permanent strong land-sea gradients which arise the total global magnitude of the numerical errors remain small, less than 0.25 percent of the global mass being used to remove negative values in the tracer field. For Radon the combined impact of the ground source and the convective mixing is a relatively severe test on the use of the spectral advective scheme. Consequently sharp gradients and low background values will be produced and numerical problems be more likely. A surprising bonus of the convective mixing has been its ameliorating effect upon the need for hole-filling in the model. As the result of active convection, the gradients of Rn^{222} are no longer so sharp and trapping in the lower model levels is reduced.

A number of tracers in future studies will be like Rn^{222} in having a source restricted to over land. This creates a tracer distribution with steep horizontal gradients at the continental boundaries. Such a distribution must be modeled by the advection scheme, maintaining the gradients, to be accurate. Figure 1 illustrates the spectral model's description of Rn^{222} near the surface, with strong gradients in the horizontal, with the tracer mass predominantly 'clinging' to the land surfaces. Future tracers will also have detailed geographical source distributions, e.g. dependent on terrain characteristics. The impact of similar sources varying on very small model scales (sometimes below the models $3 - 4\Delta x$ limit) will be partially tested as a result of the 'noisy' nature of the convective mixing which to the upper troposphere is acting like a highly variable source function in both space and time. With these types of additional requirement for advecting chemical tracers, the spectral technique is one among many alternative schemes proposed. GCMs using the spectral technique at low resolution cannot resolve strong gradients. Other alternatives exist including semi-Lagrangian and second-order moment techniques which can provide more accurate advection but normally at the expense of more computational effort and sometimes poorer mass conservation properties.

The radon distributions exhibit strong transport of tracers into the upper troposphere particularly in the tropics both via synoptic and subgrid motions. The consequences of this transport are important in any study of tracers whose source is the troposphere but whose sink lies in the stratosphere. With this type of transport scenario in mind the radon and lead tracers have been examined to view how the model moves tracers to the upper troposphere and across the tropopause into the lower stratosphere. The impact of the 'relocation' convective mixing and large synoptic ascent upon the redistribution of radon from the boundary layer to the upper troposphere is considerable. It could have also an important effect, in the long term, upon the cross equatorial transport of long lived tracer.

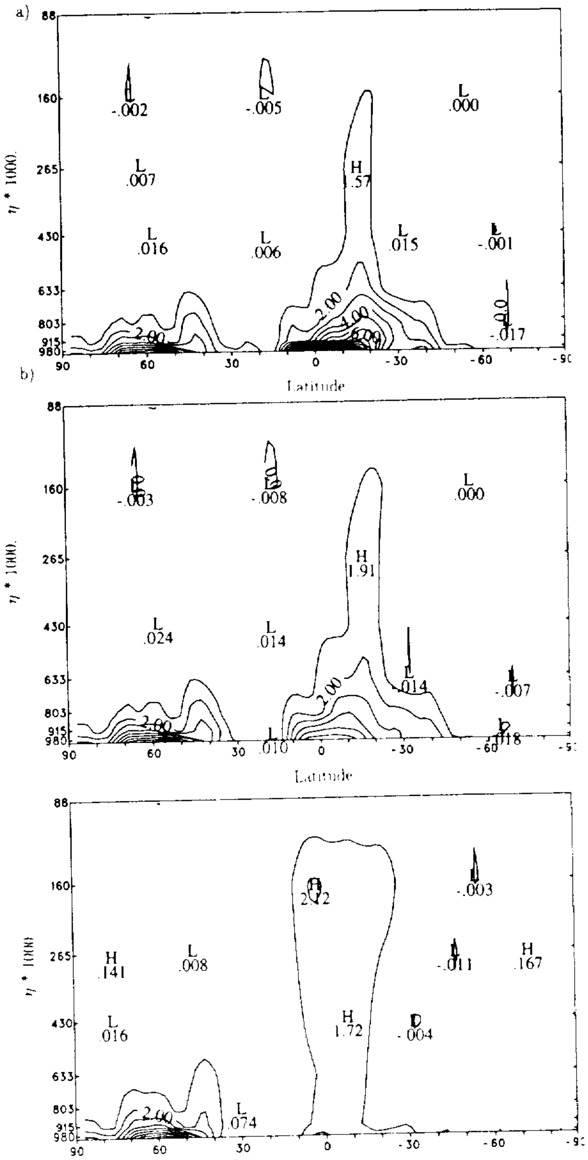

Figure 2 : $Radon^{222}$ mass mixing ratio constant longitude cross-sections, longitude 71°West, for variants: a) No convective tracer mixing, b) 'Diffusive' convective tracer mixing, and c) 'direct-Relocation' convective tracer mixing, for Day 33, 00:00 GMT. (Scaled by $1.0e^{19}$) Vertical axis shows data on model hybrid levels, where $\eta = f(p, \sigma)$.

4. Conclusions

A level 0 online tracer-model is now complete and testing indicates that mass conservation and negatives are not significant problems but that a better advection scheme may be needed if sharp gradients produced dynamically and chemically are to be maintained accurately. However the spectral technique with sufficient resolution is still a powerful and viable alternative. Sensitivity studies reveal the importance of the parameterization of convective mixing on tropospheric tracer modeling. The model's convective mixing has maximum impact over the equatorial continents, with very small effect in the winter hemisphere higher latitudes. In terms of global impact, the parameterization of convective mixing based on the relocation technique appears to be more efficient in representing adequately the transport of long lived tracers into the lower stratosphere. However, the local impact of both schemes over specific regions still needs to be assessed with more analysis. It has been also noted that convective mixing acts to reduce spectral problems due to its diffusive effect on tracer gradients. Strong synoptic vertical motions produce important transport of radon into the tropical upper troposphere, greatly increasing the potential pool of tracer available for transport into the lower stratosphere. The convective mixing parameterizations used are relatively simple and questions on the degree of entrainment throughout the column, fraction of mass 'relocated' and the height of the ensemble average convection within a GCM grid box remain outstanding. Future modeling is intended using alternative and more complex schemes in order to assess and improve our ability to parameterize and interpret subgrid mixing within global models.

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