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Modelling the Global Sources and Sinks of Radiatively Active Gases [and Discussion]

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Modelling the global sources and sinks of radiatively active gases

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The use of numerical models in understanding the budgets of atmospheric trace gases is discussed. The budget of methane is calculated in a two-dimensional model. The contrasting behaviour of the Northern and Southern Hemisphere reflects changes in the relative importance of emissions, transport and chemistry. Models can also be used to test hypotheses. An example of such a study is presented in which it is shown that changes in stratospheric ozone could have played a significant role in the dramatic change in methane trend observed in the early 1990s. Finally, use of a three-dimensional model to study tropospheric trace gases is introduced.

1. Introduction

The budgets of many atmospheric gases involve a number of different physical and chemical processes. For example, many gases important in the climate system are emitted into the atmosphere at the surface. The emissions may occur naturally or arise from human activities. Once in the atmosphere the gas will be transported by atmospheric winds. At the same time chemical and physical transformations can possibly occur. Finally, the gas may be removed from the atmosphere chemically or at the surface by various deposition processes.

One way to quantify the budget of these gases is by using numerical models which include descriptions of the relevant processes. Thus, by including the best estimates of emissions, atmospheric chemistry and transport, the modelled behaviour represents our best understanding of the behaviour of the atmosphere. Such a calculation, for atmospheric methane, is described in § 3. The modelled methane concentration is compared with observations. When the comparison is favourable, the model calculations of the methane budget can be treated with some confidence.

An alternative use of numerical models is in the testing of hypotheses. In § 4 we ask whether the reduction in stratospheric ozone since the late 1970s could have played a role in changing the growth rate of methane and other trace gases. Use of a model to ask this question enables us to quantify the possible effect.

Much of our interest in atmospheric trace gases arises from their possible role in climate change. To study this question calls for a model with the most detailed descriptions of radiative transfer, chemistry and meteorology – a weather forecast model adapted for the use of climate/chemistry studies. In § 5 preliminary results

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from such a model will be presented. Four trace gases have been included in the model. The modelled behaviour will be compared with observations, allowing some speculation about the role of chemistry, transport and emissions.

2. The models

Two numerical models will be used here, members of the hierarchy of models available to the UK Universities Global Atmospheric Modelling Programme (UGAMP). The models are: a two-dimensional model, which has been used extensively for a variety of stratospheric and tropospheric studies; and a new three-dimensional model, recently developed for studies of tropospheric chemistry.

(*a*) *The Cambridge two-dimensional model*

The model extends from pole to pole with a horizontal resolution of about 9.5° and vertically up to 60 km with a resolution of half a pressure scale height (*ca.* 3.5 km). Averages are taken over longitude at a fixed latitude, altitude and time. Two-dimensional forms of the thermodynamic, momentum and tracer continuity equation are solved for each 4-hour time step to give updated fields of temperature, velocity and chemical species. Thus, in contrast to many such models, the circulation is calculated consistently within the model, permitting interactive chemistry/climate studies. A detailed description of the basic model formulation is given by Harwood & Pyle (1975, 1977). For tropospheric chemistry studies several improvements to the model chemical scheme, to emissions, to removal processes and to the treatment of water vapour have been described in detail by Law & Pyle (1993*a*).

(*b*) *The UGAMP General Circulation Model (UGCM)*

A version of the model developed for forecast studies by the European Centre for Medium-range Weather Forecasts (ECMWF) has been modified for atmospheric research by scientists from UGAMP. The model used below is a spectral model run at a triangular truncation of T42 (*ca.* 3° latitude × 3° longitude). There are 19 vertical levels from the surface to 10 mb, with the majority of the levels in the troposphere. The model includes detailed treatments of physical processes (see ECMWF research manual 2, 1987, for details).

Two particular developments were necessary for the work described below. First, advection schemes for trace gases were introduced. The spectral method, used for other model variables, is employed in the horizontal. In the vertical a second order finite difference method, or a higher order total variation diminishing scheme (Thuburn 1993), can be used. The latter scheme is more accurate and better able to resolve steep gradients in species concentrations. This scheme is used in the UGCM tropospheric simulation described later. Second, schemes to treat the emission of gases from the surface and the subsequent atmospheric chemistry were included. We have developed detailed schemes for stratospheric chemistry (see Lary *et al.* (1993) for a description of the first stratospheric studies with the model). Detailed tropospheric chemistry schemes are currently being implemented. Here a rather simple chemistry scheme will be used.

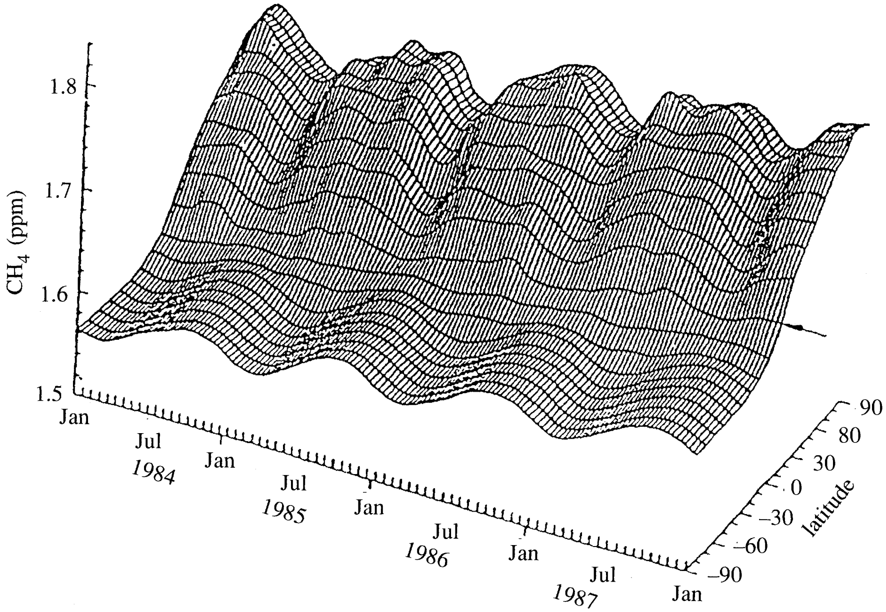

Figure 1. Three-dimensional latitude–height–time plot of the global distribution of the background concentration of CH$_4$ in parts per million by volume (ppmv) measured in the marine boundary layer at the NOAA/CMDL cooperative network for 1984–87. The arrow denotes the equator. The raw data have been smoothed to provide a uniform grid spacing of 10° in latitude and 10 days in time. From Fung *et al.* (1991).

3. The budget of methane calculated in the Cambridge two-dimensional model

The two-dimensional model was introduced in the previous section. For this study detailed emission inventories for CH$_4$, CO and NO$_x$ were developed (see Law & Pyle 1993*b*). Sources include ruminants, wetlands, rice paddies and anthropogenic emissions including landfill sites. Methane emissions totalling 575 Tg a^{-1} are included in the model. Latitudinal and seasonal variations are also taken into account wherever possible. For example, the biomass burning emissions of CH$_4$ and CO peak in the tropics with a slightly larger source assumed in the Southern Hemisphere and larger emissions in the winter season. The northern wetland source has its peak at 60° N in the early autumn. Rice paddy emissions peak in the summer at about 25° N.

Figure 1 shows surface methane observations as a function of latitude and time (Fung *et al.* 1991). The main features include a double peak in the winter northern high latitudes with the main peak in January–February. There is a steep latitudinal gradient with higher concentrations in the north (*ca.* 1.8 ppmv) than the south (*ca.* 1.56 ppmv). There is little seasonal variation in the tropics. The seasonal variation in the south is small and rather smooth with a peak in September–October.

Figure 2 shows a similar plot from one year of an integration with the model (for full details see Law & Pyle 1993*a*). The broad features of the observations have been well reproduced by the model. For example, both the latitudinal and seasonal variations are captured reasonably well in the model simulation. There

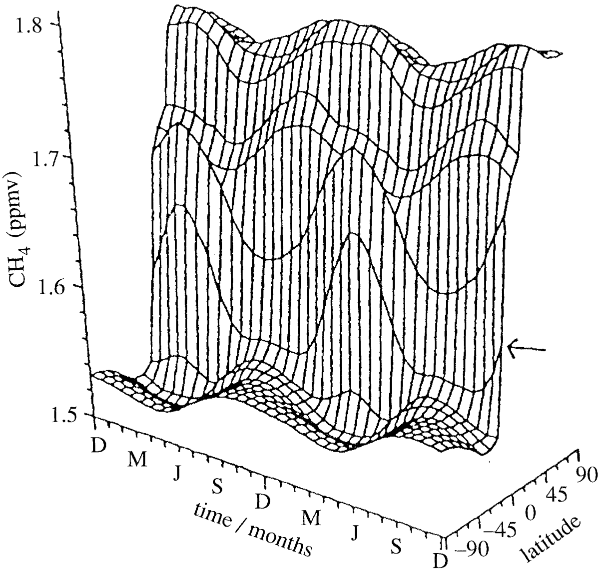

Figure 2. Three-dimensional latitude–height–time plot of modelled CH_4 mixing ratio in ppmv at *ca.* 2 km for a two-year period. Note that the data have been interpolated. The arrow denotes the Equator. From Law & Pyle (1993*b*).

are some differences. For example, the peak model values in the Northern Hemisphere are a little higher than the observations, although pollution events have been removed in compiling the latter. The model shows more seasonal variation in the tropics than seen in figure 1, although again at least one station (Mauna Loa, see Steele *et al.* 1987) does show somewhat similar seasonality.

The great advantage of models is not that they can reproduce observations with a degree of realism but that the model behaviour can subsequently be diagnosed to reveal the relative importance of different processes. Thus, if the model does reproduce the observations it is then perhaps a reasonable assumption that diagnosis of the model processes does cast some light on how those same processes operate in the atmosphere. Accordingly, the model methane budget for this calculation has been investigated. Three separate regions have been considered, bounded by the surface below and the tropopause above. The regions are the Northern Hemisphere (90° N–30° N), the tropics (30° N–30° S) and the Southern Hemisphere (30° S–90° S). Notice that, because the tropopause occurs at a higher altitude in the tropics, the volume of the tropical region is larger than the other two regions combined. For each region four terms were calculated: the surface emission into the region; the chemical destruction within the region; the flux across the tropopause, giving the exchange with the stratosphere; the flux into the other regions (for example, the exchange between the northern and tropical region).

Figure 3 shows the calculated budgets as a function of time for the three regions. From figure 3*a*, for the Northern Hemisphere region, it can be seen that methane emissions, mainly from wetlands, peak in August–September. The chemical destruction of CH_4, mainly due to reaction with OH, is reducing during this period of high emissions and reaches a minimum in the winter. Thus, it appears that the autumn and winter time peaks in the observations (see figure 1) arise from

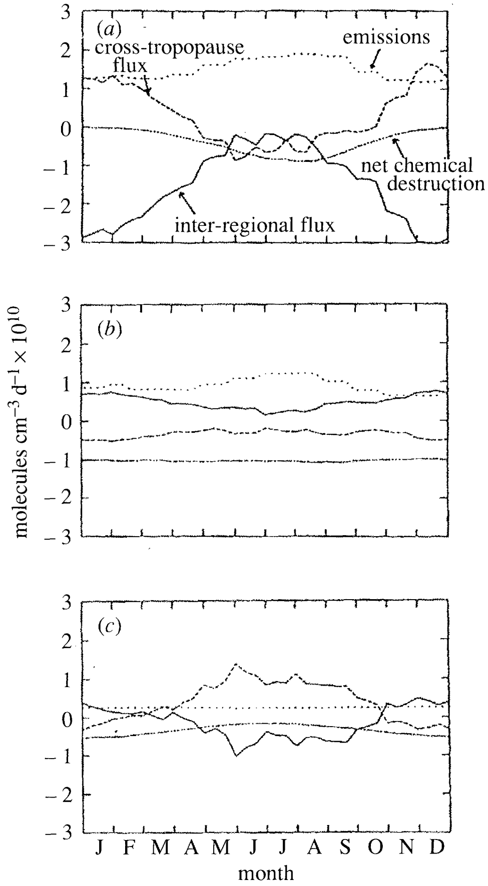

Figure 3. Seasonal variation of CH_4 budget in (*a*) the Northern Hemisphere (*ca.* 30–90° N), (*b*) the tropics (*ca.* 30° N–30° S), and (*c*) the Southern Hemisphere (*ca.* 30–90° S). Fluxes shown are net chemical destruction (effectively $-k_{30}[CH_4][OH]$); inter-regional flux, the flux in and out of a region (positive into region); cross-tropopause flux, the flux across the model tropopause (positive into troposphere); and emissions. Units are molecules cm^{-3}d^{-1}. From Law & Pyle (1993*b*).

the build up from emissions in the late autumn and the drop off in destruction by OH during the same period. Similarly the minimum in CH_4 in the summer can be explained largely by the build up in chemical destruction.

The transport fluxes make a significant contribution to the budget. Notice that the cross-tropopause flux and the inter-regional flux (in this case the flux into the tropics) are largely counteracting. Nevertheless, more than 140 Tg are transferred out of the Northern Hemisphere region, contributing to the decrease in CH_4 following the winter maximum. The various budget terms are presented quantitatively in table 1.

In the tropics, figure 3*b*, the transport fluxes cancel to a large degree. The budget is essentially a balance between emissions and chemical destruction. Subdividing the region, it appears that CH_4 in the northern tropics, where the model shows more seasonality than the observations, is driven by the seasonal variation in OH, which itself is largely a product of the seasonally varying biomass burning emissions of CO. The behaviour in the whole of the region is a complex

Table 1. *The annual budget of* CH_4

(Units, Tg (CH_4) a^{-1}. SH, Southern Hemisphere; NH, Northern Hemisphere. Stratospheric (cross-tropopause) flux, positive flux into troposphere. Horizontal transport (inter-regional flux), positive into a region.)

	global	SH 90–30° S	tropics 30° S–30° N	NH 30–90° N
emissions	575	20	368	187
net chemical destruction	−488	−30	−398	−60
stratospheric flux	−61	45	−142	36
horizontal transport	0	−17	194	−177
net	26	18	22	−14

interaction between emissions and chemistry; the observed lack of seasonal variation (figure 1) is difficult to reconcile with the seasonally varying emissions used here (see Law & Pyle (1993*a*) and the primary references therein (e.g. Aselmann & Crutzen 1989; Cicerone & Oremland 1988; Matthews & Fung 1987)). Notice though that figure 1 is based on only one observation site for every 10° latitude so that any regional variations have inevitably been ignored.

Finally, the seasonal variations in the Southern Hemisphere are driven primarily by variations in chemical destruction. However, the fact that the maximum in CH_4 (September–October) occurs some two to three months after the minimum in the chemical destruction by OH suggests that transport also plays an important role.

The model CH_4 budget is summarized in table 1. The greatest chemical destruction occurs in the tropics. Only 12% is destroyed in the Northern Hemisphere region although 33% of emissions occur there. There is a net loss of about 140 Tg a^{-1} from the Northern Hemisphere region. In the Southern Hemisphere, there is a net annual gain from transport of 28 Tg.

4. Stratospheric ozone and the trend in methane

A particularly useful role of numerical models lies in hypothesis testing. For example, the recent dramatic change in the trend in the surface concentration of methane, and other gases, could be due to a variety of causes. The rate of increase in methane was reduced during the 1980s and then, in the early 1990s, dropped close to zero (figure 4, from Dlugokencky *et al.* 1994). Surface levels of CO also decreased unexpectedly during the same period (Novelli *et al.* 1994). One explanation is that anthropogenic emissions, which amounted to more than one fifth of the total emissions of methane included in the calculation in § 3, could have changed. This could have arisen possibly because of tightening up of leakages in natural gas pipelines, or because of a downturn in the economies of the former Eastern bloc countries, or to a combination of both factors. Alternatively, changes in agricultural practice could have led to a reduction of emissions. These various hypotheses could be tested in the model by changing the appropriate source term. In this section, however, we consider the alternative hypothesis that reductions in stratospheric ozone could have produced consequent changes in the

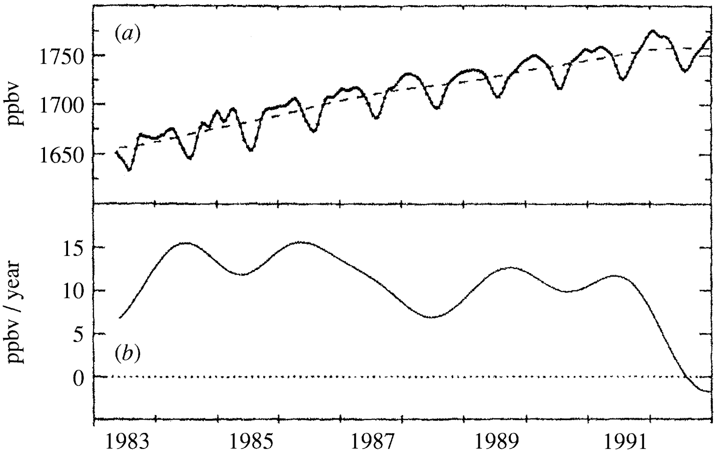

Figure 4. (*a*) Biweekly CH_4 mixing ratios for the Northern Hemisphere. The dashed line is the deseasonalized trend. (*b*) Growth rate curves determined as the derivative of the trend lines in (*a*). From Dlugokencky *et al.* (1994).

troposphere which may then have led to a reduction in the growth of CH_4 and CO.

The chemistry of the troposphere was reviewed by Derwent (this volume). OH is the major tropospheric oxidant and reaction with OH is the primary destruction pathway for both CH_4 and CO:

$$\mathrm{OH} + \mathrm{CH_4} \rightarrow \mathrm{CH_3} + \mathrm{H_2O}, \tag{4.1}$$

$$\mathrm{OH} + \mathrm{CO} \rightarrow \mathrm{CO_2} + \mathrm{H}. \tag{4.2}$$

The OH is produced by the reaction of water vapour with excited oxygen atoms, which arise from the photolysis of ozone by UV radiation of wavelength below 310 nm:

$$\mathrm{O_3} + h\nu \rightarrow \mathrm{O_2} + \mathrm{O(^1D)}, \quad \lambda < 310\ \mathrm{nm}, \tag{4.3}$$

$$\mathrm{O(^1D)} + \mathrm{H_2O} \rightarrow 2\mathrm{OH}. \tag{4.4}$$

Most radiation below 310 nm is absorbed by O_3 in the stratosphere and it is only in the tail of the absorption band close to 310 nm that penetration of radiation into the troposphere occurs. If the level of ozone in the stratosphere were to change then this would alter the amount of ultraviolet radiation reaching the troposphere and hence modify the OH concentration. For example, a reduction in stratospheric ozone should allow more radiation to reach the troposphere, thereby increasing the production of $O(^1D)$, via reaction (4.3), and then OH, following reaction (4.4). Stratospheric ozone levels have indeed changed. Throughout the 1980s, the TOMS satellite instrument has recorded a decline in ozone globally. The decrease was most dramatic in the Antarctic springtime, but in Northern Hemisphere mid-latitudes the springtime decline amounts to approaching 10% (Stolarski *et al.* 1991). The decline was even more dramatic in the early 1990s. For example, ozone values between 30° N and 60° N for the first three months of 1993 were between 10 and 15% below the mean for 1979 to 1990. Thus, it is a reasonable hypothesis that, all other things being equal, there should have been

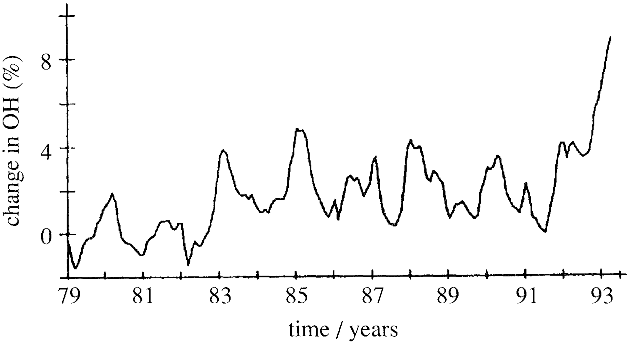

Figure 5. Time series of the percentage difference in Northern Hemispheric average OH at the surface level (*ca.* 800 mb) between the control and perturbation runs. From Bekki *et al.* (1994).

an increase in tropospheric OH since the late 1970s. The increase in OH should, in turn, have reduced CH_4 and CO by reactions (4.1) and (4.2).

This hypothesis has been tested in the Cambridge two-dimensional model by Bekki *et al.* (1994). The model was run for two cases from January 1970 to April 1993. The control run used a perpetual annual ozone column calculated by averaging the TOMS data over the period from November 1978 to October 1981, before ozone losses were large. The perturbation run used the same values before November 1978 followed by the actual TOMS ozone columns up to April 1993.

Figure 5 shows the percentage difference in Northern Hemisphere average OH in the model surface level (*ca.* 800 mb) as a function of time. There is a modelled trend in the globally averaged OH of approximately 0.2% per year between 1979 and 1990. The interannual variations in OH mirror the changes in TOMS ozone which probably arise from a number of natural causes. From mid 1991 to spring 1993 there is a large modelled increase, exceeding 8% in the Northern Hemisphere.

The difference in the modelled methane growth rates between the control and perturbation run is shown in figure 6. Results are plotted showing the average for the Northern Hemisphere, the Southern Hemisphere, and also the global average. The variations follow the variations in ozone and OH. In the early 1980s the model indicates that this mechanism alone should have lead to decreases in CH_4. Thus stratospheric ozone changes may have contributed to the slow decline in the CH_4 growth rate before 1985. The most striking feature in figure 6 is the marked decrease of 5–6 ppbv per year in the Northern Hemisphere from the spring of 1991 to autumn 1992. This is not so large as the observed decrease of about 12 ppbv but it does suggest that the record low ozone column values in 1992 may have accounted for almost half of the unusually large reduction in CH_4 growth rate recorded that year.

We should not expect the reduction in stratospheric ozone to explain all the trend in methane. Gases which are emitted at the surface and which have no substantial tropospheric chemistry have also shown reductions in the atmospheric growth rates in the early 1990s. Furthermore, changes in the emissions of CH_4 mentioned above could also have contributed to the change in the trend. Other factors, for example, increased emission of photochemically active species, may also have conspired to produce an upward trend in OH. What the model calculation does show is that changes in stratospheric ozone may have made a substantial contribution to the change in the methane trend.

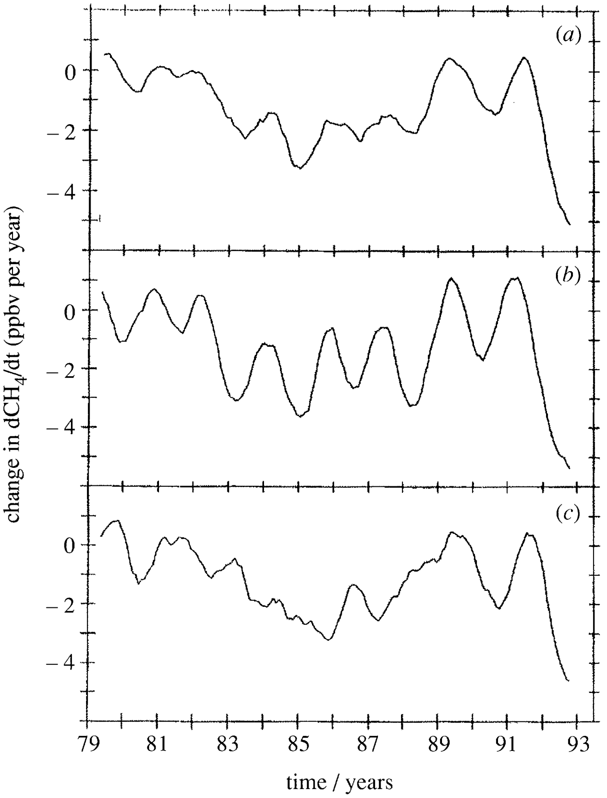

Figure 6. Time series of the difference between the runs in the growth rates of CH_4 mixing ratios at the surface level (*ca.* 800 mb). Growth rates are calculated from the deseasonalized monthly means using the method of 'linear moving slopes' described in Khalil & Rasmussen (1990) with a time span of one year. From Bekki *et al.* (1994).

The hypothesis is also supported by the model calculations of CO. These too show a decrease in line with observations. The model also predicts that the growth rate of a number of other species removed in the troposphere by OH should have changed. These include CH_3CCl_3 and CH_3Br (decreases of 1 pptv per year between spring 1991 and autumn 1992 for both species). Trend analyses of the observations of these species would be an interesting test of the hypothesis. Moreover, recent measurements indicate that there has been some recovery in stratospheric ozone from the record low values of 1992 and 1993. This should translate into a further change, now an increase, in the growth rates of CH_4 and CO.

5. Three-dimensional modelling of tropospheric trace gases

To predict the detailed impact of various trace gases on the climate system three-dimensional models of meteorology, atmospheric chemistry and radiative transfer are required. Such a model, the UGCM, is being developed by the UK Universities Global Atmospheric Modelling Programme (UGAMP) and preliminary calculations for a few trace gases have been made. First results are presented in this section.

Four gases have so far been included in the UGCM. The gases were chosen because they are being measured routinely at a number of sites globally, so that comparison between model and observations is possible. The gases chosen cover a range of atmospheric lifetimes so that comparison of their respective modelled behaviour should give an indication of the relative importance of atmospheric chemistry and transport. Furthermore, all of the gases chosen have simple chemistries; they are emitted at the surface and destroyed in the atmosphere. None has an atmospheric source. The gases are $CFCl_3$ (CFC11), which is destroyed photolytically in the stratosphere, CH_3CCl_3, CH_4 and HCFC123, a shortlived gas being developed as an alternative to the CFCs (like $CFCl_3$). The latter three gases are destroyed mainly by reaction with OH.

To simplify the calculation, no attempt was made to model the chemistry in detail. Instead, the loss rates (essentially the concentrations of OH, $O(^1D)$ and Cl and the appropriate photolysis rates) were specified to the model from previous calculations with the two-dimensional model. Zonally averaged values were supplied to the three-dimensional model at a frequency of ten days.

Since the chemistry in the model is particularly simple, relatively long integrations of several years were possible. For each model year the same annually varying sea surface temperature distribution was used. While there is interannual variability in the model this is not so large as might be expected had observed sea surface temperatures been used. In this calculation there is no treatment of tracer transport by convection. Enhanced diffusion allows gases, emitted at the surface, to enter the free troposphere. A scheme to describe the transport of these species due to convection is currently under development.

Emissions of three of the gases were based on their industrial production figures, from the Alternative Fluorocarbons Environmental Acceptability Study, and distributed globally according to electricity consumption figures and population density (Hartley & Prinn 1993, for CFC-11 and HCFC123; Midgley *et al.* 1992, for CH_3CCl_3). Thus the majority of emissions occur on the eastern seaboard of the United States, in Europe and in the Far East, with smaller pockets of emission elsewhere. For methane, several different emission scenarios were run covering the various emission types mentioned in § 3.

The distributions of all four gases have several features in common. At the surface, all the gases show the influence of the source regions, with highest concentrations there and downwind. In the mid troposphere the local influence of sources is no longer evident in the modelled distributions. However, a strong hemispheric gradient is seen in all cases with the intertropical convergence zone representing a barrier to interhemispheric transport throughout the low and mid troposphere. The zonal distributions calculated within the model are in remarkable agreement with those calculated in the two-dimensional model. In particular, transport from the Northern to the Southern Hemisphere clearly takes place in both models in the tropical upper troposphere above the ascending branch of the Hadley circulation.

Figure 7 shows the time series of $CFCl_3$ calculated for the grid box containing Oregon, the site of one of the ALE/GAGE measurement stations. There is a difference between the measured and modelled concentrations of $CFCl_3$, due to a poor initialization at the start of the run. Of more interest is the fact that the amplitude of the modelled seasonal variation is larger than observed. This may be due, in part, to the model holding emissions within the boundary layer. Recall also that the model was run at a horizontal resolution of about 3° × 3° so that

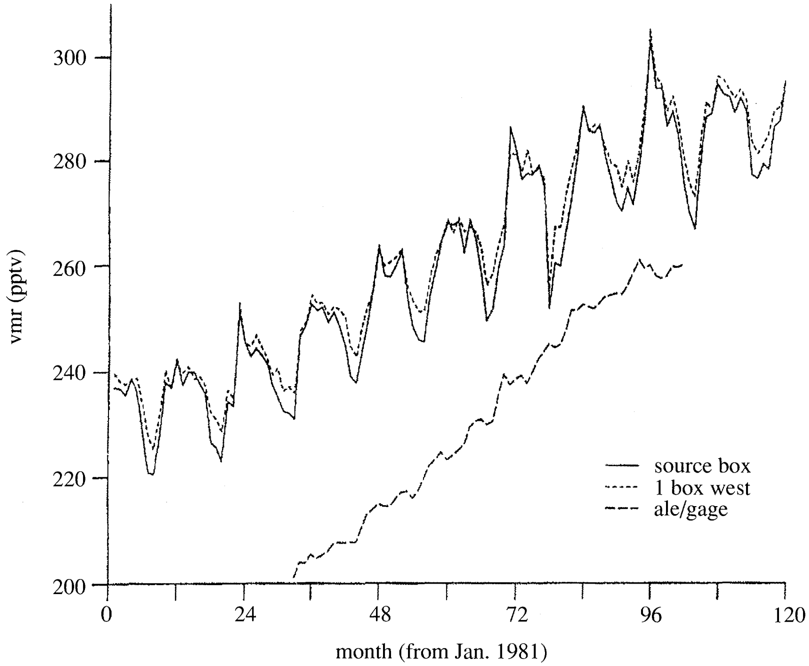

Figure 7. The calculated trend of $CFCl_3$ (pptv) in the model at Oregon. The ALE/GAGE data are also plotted.

emissions occur directly into the Oregon grid box. Runs with a deep convection scheme may help to resolve this discrepancy (also seen to a lesser extent in the calculation of Spivakovsky *et al.* (1990)).

The timing of the seasonal variations are in reasonable agreement with the observations, with minimum values found in late summer in both model and observations. The model behaviour can be explained by reference to the model surface pressure fields. In winter, pressure over the United States is high and air will be carried from the source regions on the eastern seaboard, around the southern edge of the anticyclone, bringing elevated concentrations of emitted species to Oregon. In contrast, in the summer months low pressure persists over north America with high pressure off the west coast so that much cleaner air is carried southwards to Oregon. It is interesting that Spivakovsky *et al.* (1990) explained their model behaviour as arising from venting of the boundary layer by increased convective activity during the summer months. It is clear that in this model simulation the explanation lies in the changing transport to Oregon.

Moving southward, and away from the main emission regions, the model agrees rather better with observations. For example, at Barbados (not shown) the model in August–September shows low values of the trace gases, characteristic of the Southern Hemisphere, a feature seen often in the measurements at that time of the year. This appears to be due to movement of the intertropical convergence zone, probably exaggerated in the model. Figure 8 shows the model behaviour of CH_3CCl_3 in the Southern Hemisphere at Cape Grim, Tasmania. As with the observations, the model has a lower seasonal variation in the south, presumably arising from the reduced influence of local emission sources. The modelled sea-

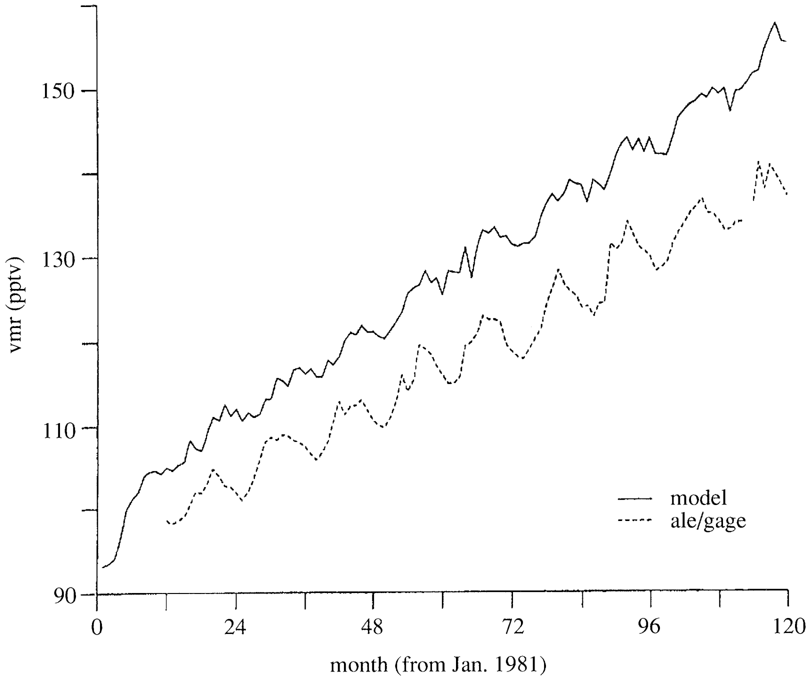

Figure 8. The calculated trend of CH_3CCl_3 (pptv) for Cape Grim, Tasmania. The ALE/GAGE data is also plotted.

sonal cycle for CH_3CCl_3 agrees well with the observations. Minimum values are found in mid to late summer, reflecting the importance of chemical destruction by reaction with OH, although this is also consistent with the seasonal cycle in transport.

The modelled methane also follows the above behaviour. In sites remote from emissions the model behaves quite well whereas closer to the emission regions comparison between model and observations is less satisfactory. Figure 9 shows the behaviour at the Guam Marianas Islands (13° S, 144° E), where the model reproduces the observed seasonal variation extremely well although the modelled concentrations are a few percent too high. In contrast at Alert, in the Canadian North West Territories (82° N, 62° W), the model fails completely to reproduce the observed seasonal behaviour (figure 10). Alert lies within the region of wetland emissions and the modelled behaviour follows the seasonal variation in emissions. It is unclear why the modelled and observed behaviours are so different. If the emissions, following Matthews & Fung (1987), are specified accurately then to reproduce the observed seasonal variation would require a rapid transport away from the region. Notice, though, that Choularton *et al.* (this volume) report CH_4 concentrations in excess of 2 ppmv over a wetland emission region in northern Scotland. The magnitude of the modelled late summer peak may therefore not be unreasonable for a wetland region. It could be that the spatial resolution in the model is insufficient to capture the real horizontal variability. Another possibility (Nisbet, personal communication) is that the wetland emissions are very temperature dependent and in consequence fall off with increasing latitude north of 50° N much more rapidly than in the model.

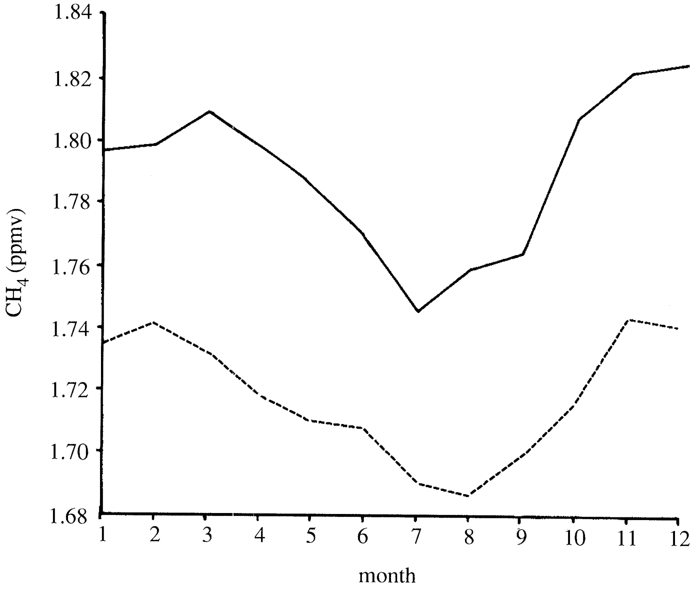

Figure 9. The modelled annual variation of methane (ppmv) at Guam, in the Marianas Islands. Also plotted are data from the same location collected by the NOAA/CMDL cooperative network.

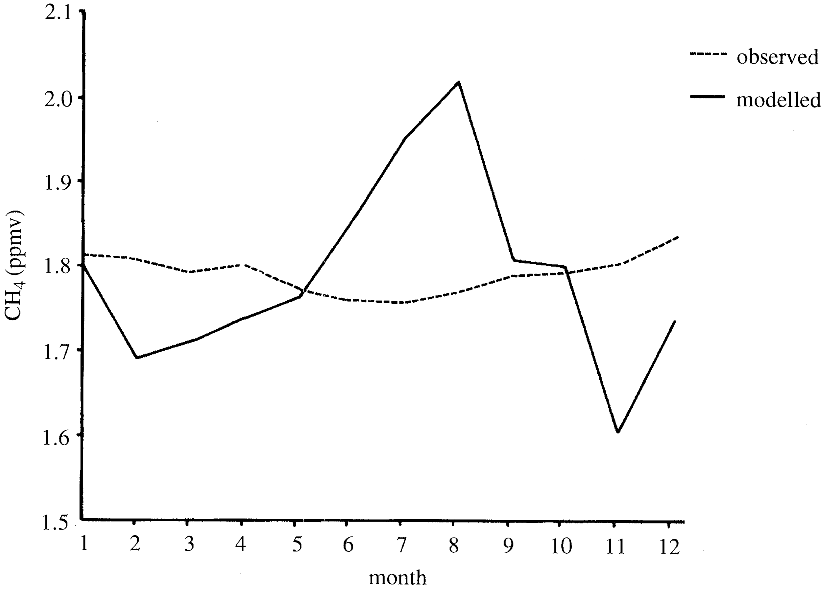

Figure 10. As figure 9, for Alert, North West Territories.

It is clear that three-dimensional models, of the type introduced here, have an extremely important role to play in understanding emissions of trace gases into the atmosphere. As computing power increases they will be further developed to become the workhorse for climate/chemistry studies.

6. Conclusions

Numerical models are powerful tools for studying the lifecycles of trace gases from their emission at the Earth's surface to their eventual destruction in the atmosphere or by removal at the surface again. Two-dimensional models are now well established and two studies were described, one an investigation of the tropospheric budget of methane and the other an investigation of recent changes in the methane trend. The budget study highlighted the role of different atmospheric regions. For example, methane is exported from northern latitudes whereas in the south the budget is dominated by chemical processes. The trend investigation showed that recent changes in stratospheric ozone could have had a significant impact on the growth rate of various tropospheric gases.

Three-dimensional models are now being developed at several centres. They are becoming extremely important for the very detailed study of the processes controlling trace gases and will be the primary tool for future studies of the coupling between atmospheric chemistry and climate.

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Discussion

C. JOHNSON (*Hadley Centre for Climate Prediction and Research, Meteorological Office, London, U.K.*). Is our knowledge of tropospheric chemistry sufficiently robust to allow comparison between model simulations and atmospheric measurements, and can these be used to identify errors in our current understanding of the distribution of methane sources?

J. A. PYLE. There are two separate issues here. Firstly, validation of our understanding of tropospheric chemistry demands, as an essential step, detailed comparison between models and measurements. Secondly, and more specifically, what do these comparisons tell us about methane soruces? Here the answer is more difficult. Inference of methane sources from observations, or combinations of observations and models, in indirect. Subtle differences between models and observations will therefore be unlikely to shed significant light on our knowledge of the sources. On the other hand, when large differences occur the inferences made could be more instructive. Thus, in my paper, the large difference found between the observed and modelled seasonal cycle of methane at Alert could weel indicate that the northern wetland source in the model is too strong, an inference apparently supported by other evidence becoming available.