A SOUTHERN VIEW OF THE GLOBAL ATMOSPHERE

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(with three text figures)

CSIRO has been monitoring trace gases in the antarctic atmosphere since 1977. An increasing number of trace constituents is being recorded in an effort to better understand human impact on the atmosphere and the consequent climatic and biological effects. This paper focusses on Southern Hemisphere measurements of carbon dioxide, with particular attention to the stable isotope ratios of CO_2.

Secular, interannual, seasonal, and latitudinal variations in concentration, and in the isotope ratios $\delta^{13}C$, $\delta^{18}O$, are examined and their significance to future modelling of the global carbon budget discussed.

The competing requirements for baseline atmospheric monitoring are access to a large, unpolluted, well-mixed air mass and provision of adequate logistical and scientific support. Macquarie Island, remote from continental influences with an established scientific base, combines these requirements to an unusual degree.
Key Words: atmospheric monitoring, carbon cycle, Macquarie Island.

INTRODUCTION

Recognition that the composition of an atmosphere provides a sensitive measure of a planet's life processes came into focus with the NASA quest for life on Mars; an excellent discussion of the compelling atmospheric evidence for life on earth, as reflected in the levels of oxygen, carbon dioxide, methane and many other trace gases was given by Lovelock (1979). More recently, concern about the global impact of industrial and agricultural practices, through changes in atmospheric composition, has gathered momentum. The anticipated changes in the radiative properties of the atmosphere are most publicised in terms of a fear of undesirable climatic change, but other direct and indirect effects on life-forms are also anticipated. A concise overview of these problems has been published by the World Meteorological Organisation (WMO) (1986).

The response to the perception of these changes as a sensitive measure of life processes, of global pollution and of dynamical processes in the atmosphere, has been the establishment of several global atmospheric study programmes. A prime example is the WMO co-ordinated Baseline Air Pollution Monitoring Network, to which the Australian station at Cape Grim (northwestern Tasmania), recently described as "arguably the most comprehensive single surface atmospheric monitoring facility in the world" (Baseline 83-84, 1986), is a major contributor. A second example, on a smaller scale, is a CSIRO programme collecting monthly clean-air samples from Alaska, Hawaii, Samoa, Tasmania, Macquarie Island, Mawson and the South Pole. Preliminary data from both programmes are presented below.

These and similar programmes are the launching pad for major new international initiatives, such as the US Global Tropospheric Chemistry Program and the International Geosphere-Biosphere Program (IGBP), to improve understanding of atmospheric transport, chemical and radiative properties.

THE CSIRO PROGRAMMES

CSIRO commenced monitoring of CO_2 concentration from aircraft over southeastern Australia in 1972 (Pearman *et al.* 1983). Surface observations commenced at Cape Grim (41°S) in 1976, Mawson (68°S) in 1977 and Macquarie Island (54°S) in 1979; these programmes are fully described by Beardsmore & Pearman (1987). *In situ* infrared gas analysers (IRGA) are employed at Cape Grim and Macquarie Island, whereas at Mawson (as well as for the aircraft and at Cape Grim) whole-air samples are collected in 0.5 litre glass flasks for subsequent analysis at CSIRO. Since 1980, the 0.5 litre samples have also been analysed by gas

chromatography for CO_2, CO, CH_4, N_2O, CCl_3F, CCl_2F_2, CH_3CCl_3 and CCl_4 (Fraser *et al.* 1986). (Note that CSIRO measurements of airborne particulate matter over similar periods (Gras & Adriaansen 1985) are not discussed here.)

The monthly collections of clean air from Alaska through to the South Pole (including Macquarie Island and Mawson) commenced in late 1983/early 1984. These involve 5 litre pressurised glass containers which are returned to CSIRO. After gas chromatograph analyses for the range of trace gases listed above, the CO_2 is cryogenically extracted and analysed on a mass spectrometer for $\delta^{13}C$ and $\delta^{18}O$, where, for example,

$$\delta^{13}C = \frac{(^{13}C/^{12}C \text{ sample} - ^{13}C/^{12}C \text{ reference})}{(^{13}C/^{12}C \text{ reference})}$$

and the δ are normally multiplied by 1000 and expressed in per mil. A full description of this programme is in preparation for publication.

At CSIRO the collection and interpretation of these data involves interaction with modelling studies. The long-term trends and equilibrium distributions of trace gases are modelled with one-dimensional diffusion models (Enting & Pearman 1987), while the modelling of interannual, seasonal and latitudinal variations employs two-dimensional transport and exchange (Pearman & Hyson 1986, Enting & Mansbridge 1987).

The data discussed below are preliminary and recent, and have not been specifically modelled. Emphasis is placed on describing new and unusual aspects of the results, which may involve considerable evolution of the models for a quantitative interpretation.

CO_2 CONCENTRATION RESULTS

The observations of CO_2 concentration in the higher latitudes of the Southern Hemisphere have contributed substantially to our understanding of the global fluxes of that gas across the air-sea interface and in the atmosphere as a whole. While transport of gases in the atmosphere is not always known with great precision, it can be sufficiently constrained to interpret concentration gradients in terms of surface exchanges. For example, the current input of about 5 Gt C yr^{-1} (1 Gt C = 10^{15} g of carbon) as CO_2 from the combustion of fossil fuels, primarily in the Northern Hemisphere, on average maintains about a 3 ppmv concentration gradient

between the hemispheres. Similar gradients are established in the Northern Hemisphere, associated with the periodic exchange of carbon with the terrestrial vegetation (Pearman & Hyson 1986).

The increase in atmospheric CO_2 (around 1.5 ppmv yr^{-1}), due to fossil fuel combustion, can be seen in figure 1. The annual mean concentrations at Mawson and Cape Grim (41°S) are very similar, suggesting that, on average, there is little flux of CO_2 between the mid and higher latitudes in the lower atmosphere, and no major surface sink for CO_2 exists at these latitudes. Shipboard observations taken from Australian antarctic supply ships, together with the observations at Cape Grim, Macquarie Island and Mawson (Beardsmore & Pearman 1987) do show, however, that there may be a small regional maximum of concentration around 56°S, at least in the summer months. The maximum concentration is about 1 ppmv higher than at Cape Grim. Komhyr *et al.* (1985) have also suggested that a regional source of CO_2 may exist. It is possible that this source is related to the Antarctic Convergence zone. Perhaps carbon-rich intermediate water comes to the surface in this zone, giving an ocean surface layer with CO_2 partial pressure (pCO_2) substantially highly than that in the atmosphere. Only ocean pCO_2 measurements will confirm this.

Both Macquarie Island and Mawson exhibit somewhat larger seasonal variations in concentration than occurs at Cape Grim. Early summer concentrations are similar at both southern stations but, during the following months, Mawson appears to show a more definite decrease.

This larger seasonal amplitude at Mawson has become particularly obvious in our 1985 and 1986 data. The reason for this effect is not known. It is possible that it is a relatively local event, involving the disappearance of the sea ice and the summer melting of ice in the coastal region (Budd 1980). The annual supply of very cold fresh water, low in carbon content but with high solubility for CO_2, may provide the suggested sink for atmospheric CO_2. If it is a coastal effect, for example extending over 10° of latitude and through a depth of atmosphere of 1 km, then it can be calculated that this decrease represents a surface sink of order 10^{-2} Gt C. Given that the major fluxes in the carbon cycle are of order 1 Gt C, this annual feature, or the variations in its magnitude from year to year, are unlikely to be of great importance in the global perspective, though there may be appreciable influences on the local marine biosphere.
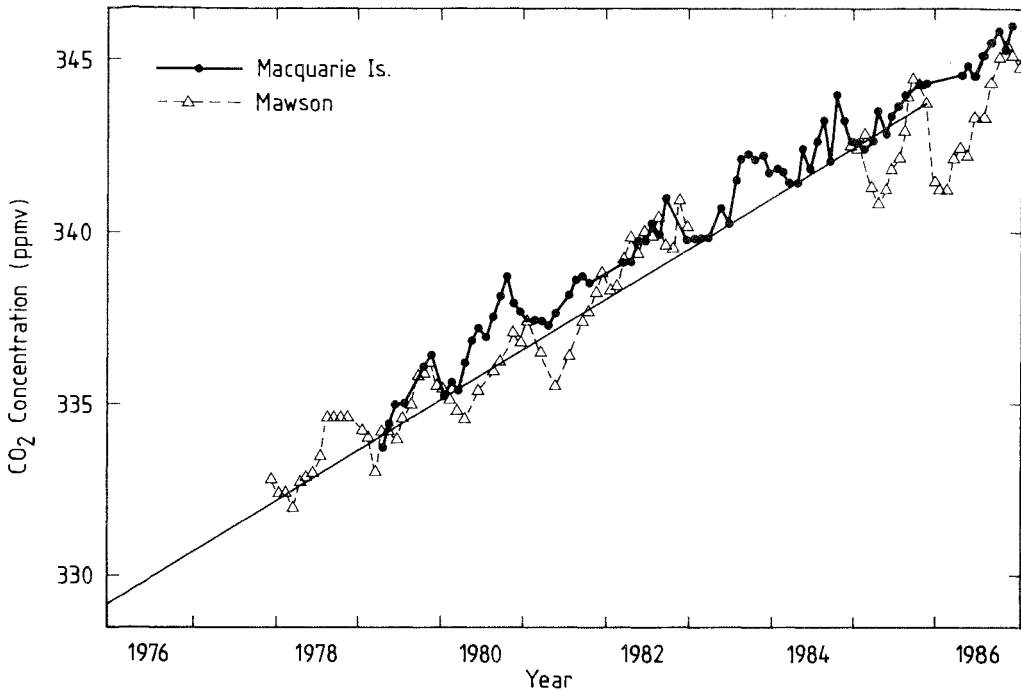
FIG. 1 — *Mean monthly carbon dioxide concentrations at Macquarie Island and Mawson. The straight line is a linear regression to the data from the Australian Baseline Air Pollution Station at Cape Grim, Tasmania.*

CARBON ISOTOPE RESULTS

An important generalisation for a qualitative interpretation of the stable carbon isotope ($^{13}C/^{12}C$) data is that atmospheric values are mainly determined by CO_2 exchange involving terrestrial biological material. The dominating mechanism is a discrimination against the heavier isotope, ^{13}C, or a preference for ^{12}C, shown by plants during photosynthesis (Farquhar *et al.* 1982, Francey & Farquhar 1982). Thus, a net uptake of CO_2 by plants results in an increase in atmospheric $\delta^{13}C$, whereas respiratory CO_2 or CO_2 from the combustion of terrestrial biological material (including fossil fuel) decreases $\delta^{13}C$.

In equilibrium exchange of CO_2 with surface water, in particular the oceans, the fractionation that occurs on entry is practically balanced by a reverse fractionation on exit, with no net result (for example, Siegenthaler & Munnich 1981). Note that plants growing in the ocean mixed layer also fractionate carbon; however, since the biological

carbon is largely extracted from, and returned to the ocean, and since dissolved CO_2 is a tiny fraction of the total available carbon, the influence of marine life on atmospheric isotope ratios is generally negligible.

The time constant for CO_2 equilibration with the ocean surface water is about ten times longer than most gases (for example O_2), and is around one year; the time constant for carbon isotope equilibrium is even longer, by a factor equal to the ratio of total inorganic carbon/dissolved CO_2; this ratio ranges from about 180 in warm water to about 150 in cold water (Broecker & Peng 1982). A factor to be kept in mind here is that for a given perturbation of CO_2 concentration and isotope ratio, the percentage change in $\delta^{13}C$ might considerably exceed the percentage change in CO_2, even though the equilibration of CO_2 is much more rapid. This merely reflects the fact that $\delta^{13}C$ is essentially a high precision measurement of a difference from a reference value which happens to be close to the equilibrium value.
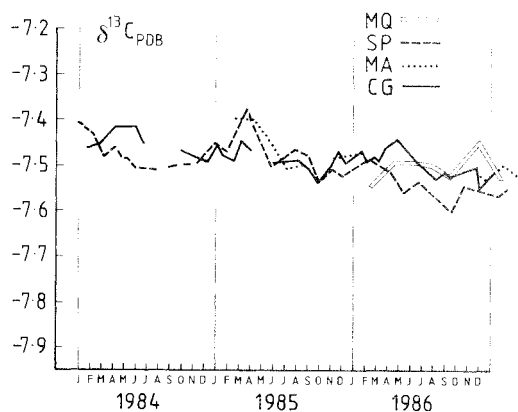
FIG. 2 — Preliminary (smoothed) stable carbon isotope ratios, $\delta^{13}C$, in CO_2 extracted from 5 litre flasks of air collected at Cape Grim, Macquarie Island, Mawson and South Pole. Values are in per mil, expressed in the CSIRO (1980) PDB reference scale. (Note: corrections have been applied for a N_2O contribution to mass 44.)

In the present context, that is, in examining data south of Cape Grim, the absence of appreciable industrial and terrestrial biospheric activity implies an absence of sources of isotopic variation south of Cape Grim. This is significant, given that we have already described sources of CO_2 concentration variation south of Cape Grim.

In figure 2 the preliminary stable carbon isotope values are presented for years 1984–86, for Cape Grim, Macquarie Island, Mawson and South Pole.

The most obvious feature is the steady trend of decreasing $\delta^{13}C$ evident at all stations with sufficiently long record. The change is about 0.03 per mil yr^{-1} which, for a concentration change of 1.45 ppmv yr^{-1}, implies a coefficient of around 0.02 per mil ppmv^{-1} (Francey 1985). These numbers are now relatively well established and understood in terms of the known input of ^{13}C-depleted CO_2 from fossil fuel combustion, mainly in the Northern Hemisphere, coupled with ocean uptake (Pearman & Hyson 1986). As discussed by these authors, and others, there is some controversy over the fact that the total release of carbon from forest clearing and agriculture is constrained by these measurements to values small compared to fossil fuel release. This is contrary to estimates from some ecological studies.

An important objective in setting up the study was to define more accurately the latitudinal gradient in $\delta^{13}C$ resulting from the Northern Hemisphere fossil fuel release (present estimates give a north–south high-latitude difference of around minus 0.2 per mil), which permits important refinements of the parameterisations of atmospheric transport and surface exchange. A close study of the data of figure 2 does, in fact, suggest systematic differences between Southern Hemisphere stations. For example, in 1984 and 1986, values of $\delta^{13}C$ at South Pole are distinctly lower than at Cape Grim (and Macquarie Island), while Mawson appears closer to South Pole, at least in 1985. Model studies (Heimann *et al.* 1985, Pearman & Hyson 1986) predict a lower value at high latitudes as a result of the high altitude at South Pole (2800 m), coupled with a predicted decrease in $\delta^{13}C$ with altitude, apparently reflecting southwards transport of ^{13}C-depleted Northern Hemisphere air in the upper arm of the Hadley cell. Figure 2 suggests inter-annual variations (with the gradient effectively absent in 1985), possibly related to variations in this transport.

Seasonality is evident in the Cape Grim data, with a $\delta^{13}C$ maximum in early March, practically coincident with a concentration minimum. This is characteristic of exchange with terrestrial biosphere, and the coefficient of 0.05 per mil ppmv^{-1} is consistent with this (Francey 1985). The 2D transport and exchange models predict a decoupling of concentration and isotopic seasonal variations going south from Cape Grim, mainly due to an increasing oceanic contribution to the concentration changes. There is some evidence for this decoupling in figure 2; however, several more years data are required for a quantitative description of this effect. For example, there is a suggestion of enhanced $\delta^{13}C$ at Mawson and South Pole in the late summer of 1985, coincident with the concentration dip observed at Mawson — if real, we have no simple explanation for such an isotope variation, and there is no evidence for similar variations in the other years.

The only other known programme (with comparable precision to the CSIRO programme) producing clean-air $\delta^{13}C$ from a global network is described by Mook *et al.* (1983). Their recent data exhibit a large interannual variation occurring around the globe throughout 1983–84 (Heimann *et al.* 1986). The variation is characterised in their South Pole record by a decline in early 1983 $\delta^{13}C$ to a value at least 0.1 per mil below the long-term linear trend (established between 1977 and 1982), followed by a recovery to the long-term trend line by the end of 1984. Such a variation implies appreciable net fluxes of biospheric CO_2 which, with current models of carbon transport and

exchange, should be accompanied by a concentration perturbation of some 2 ppmv; no such CO_2 concentration perturbation is observed. An isotope variation of 0.1 per mil throughout 1983-84 is definitely excluded by the data of figure 2, suggesting a systematic error in the Heimann *et al.* data.

OXYGEN ISOTOPE RESULTS

Data on the oxygen isotope ratio ($^{18}O/^{16}O$) in atmospheric CO_2 were originally collected solely for the purpose of correcting the carbon isotope data, to distinguish $^{13}C^{16}O^{16}O$ from $^{12}C^{16}O^{17}O$ at mass 45 (Craig 1953). Inspection of several years of Cape Grim data revealed marked systematic behaviour, and led to a biogeophysical hypothesis to explain the observed variations (Tans *et al.* 1986).

A distinguishing feature of the Cape Grim *in situ* isotope programme is the pre-drying of air prior to CO_2 extraction and analysis (Francey & Goodman 1986). This feature is also employed in the 5 litre flask sampling. It is now apparent that the drying prevents CO_2 hydration followed by equilibration of oxygen isotopes (particularly during extended sample storage), and thus information on the original CO_2 oxygen isotope is maintained. The latitudinal gradients emerging from the global 5 litre flask programme strongly support the original hypothesis (Francey & Tans 1987).

Basically, the dissolution of CO_2 in water, and its subsequent hydration, are considered necessary for the modification of atmospheric CO_2 $\delta^{18}O$; direct exchange with other sources of atmospheric oxygen (for example O_2, water vapour) appears to be negligible. Similarly, the injection of fossil fuel CO_2 has little effect on the $\delta^{18}O$ of atmospheric CO_2 (resulting in a latitudinal difference of only around 0.2 per mil). Hydration in the more obvious sources of water, for example, the oceans or cloud water, is governed by the CO_2 hydration rate; this turns out to be far too slow to explain the observed seasonal and interannual variations. Leaf water provides an answer because of the presence of the enzyme carbonic anhydrase. This enzyme is ubiquitous in plants, and the catalysis of CO_2 hydration by carbonic anhydrase is one of the fastest known enzymatic reactions — at room temperature the hydration process is speeded around a million times. Thus we have an appreciable source of water in which there is rapid and complete equilibration of CO_2 and its oxygen isotopes.
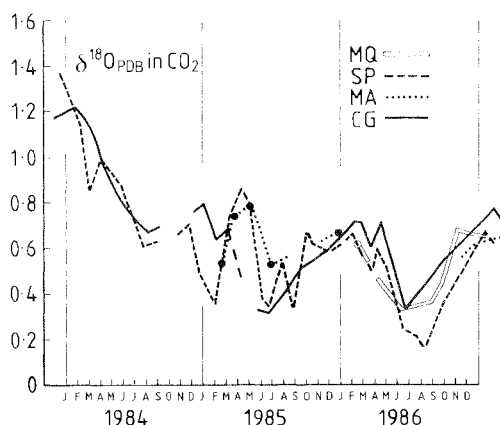

FIG. 3 — *Preliminary (smoothed) oxygen isotope ratios, $\delta^{18}O$, in CO_2 extracted from 5 litre flasks of air collected at Cape Grim, Macquarie Island, Mawson and South Pole. Values are in per mil, expressed in the CSIRO (1980) PDB reference scale. (Note: corrections have been applied for a N_2O contribution to mass 44.)*

With this model, factors which influence $\delta^{18}O$ in atmospheric CO_2 are then

(1) the ground water $\delta^{18}O$ — to a first approximation this is determined by the temperature of cloud condensation, with lower $\delta^{18}O$ at lower temperatures; the dominant feature is a latitudinal gradient from equator to pole, with lower $\delta^{18}O$ at the poles; a secondary effect is related to preferential precipitation of the heavier isotope during rain — thus $\delta^{18}O$ decrease going from the coast towards the interior of continents;

(2) the fractionation which occurs due to evaporation from leaves, resulting in leaf water appreciably heavier (with higher $\delta^{18}O$) than ground water; the amount of fractionation of leaf water compared to ground water depends inversely on the humidity;

(3) the total amount of CO_2 cycled through leaf water — to a first approximation this depends on the distribution of trees; second-order effects may relate to large-scale changes in gross biospheric turnover.

An important perspective on this process is the huge amounts of CO_2 exchange required to maintain the observed gradients against rapid atmospheric mixing. Francey & Tans (1987) estimate that exchange of some 200 Gt C yr^{-1} is required to explain the observed Northern Hemisphere gradient, compared to a total net

primary production of all plants on earth of 40–70 Gt C yr^{-1}, or a global CO_2 exchange across the air-sea interface of around 90 Gt C yr^{-1}.

With this background, we can examine the first detailed data on $\delta^{18}O$ in CO_2 values for the Southern Hemisphere in figure 3. There is quite good agreement in mean level between all four stations, as might be expected if the major source of the variations is associated with terrestrial biosphere, most of which is well to the north of Cape Grim. The strong coherent seasonality is also evidence that no major sources or sinks for $\delta^{18}O$ variations occur in the southern region. No explanation so far exists for the strong interannual variability, particularly evident in 1984, though with the dependence on rain distribution, humidity and biological activity, we can speculate that the climatic perturbations associated with the unusually severe El Nino event of 1982–83 may be a factor.

Perhaps the most interesting suggestion from the preliminary data is that, as with $\delta^{13}C$, Mawson is more closely associated with South Pole and Cape Grim with Macquarie Island. Since both species are linked to sources mainly north of Cape Grim, but involving somewhat different source mechanisms, this is reinforcement of the suggestion that transport in the upper levels of the atmosphere is a factor.

CONCLUSIONS

In summary, the CO_2 concentration and carbon isotope measurements, coupled with modelling studies, have yielded the following tentative description of the carbon cycle over the Southern Ocean. On the northern limits of the Southern Ocean (around 35° S) there is significant oceanic uptake, with a small seasonal exchange driven primarily by the effects of temperature on pCO_2; the carbon isotope data indicate that, at Cape Grim, the main seasonality in concentration is derived from exchange with terrestrial biosphere. Going south to the Antarctic Convergence, some uptake of CO_2 at the surface still occurs, although the seasonality now appears to be controlled by the activity of the marine biosphere. At the Convergence, there appears to be a release of CO_2 to the atmosphere. At higher latitudes and on the coast of Antarctica, there is no evidence of oceanic uptake or marine biological effects, with the exception of a regional (unmodelled) scale late summer uptake on the coast. In terms of the global budget in this region, the pCO_2 of surface water is probably maintained close to atmospheric levels by deep vertical mixing.

In addition, the carbon isotope results suggest refinements to the parameterisation of Southern Hemisphere transport at upper levels of the atmosphere. This is supported by the oxygen isotope data. Given apparent difficulties with past methods, the isotope data presented here give a first detailed picture of the variations in these species in the Southern Hemisphere.

The challenge of the current measurement programmes is to provide conclusive evidence for the existence, or otherwise, of these features of the carbon cycle, and in particular to understand more about the relative roles of surface exchanges and interannual atmospheric transport changes. A sound understanding of the carbon cycle in this region is important if accurate predictions of future levels of CO_2 are to be made, especially if the dynamics of the ocean are to evolve with the changing global climate.

It is against this background that the importance of Macquarie Island as an atmospheric monitoring site becomes apparent. Few other latitude bands in the world are so poorly represented in terms of human activities, biologically active land surfaces and continental shelfs, and orographic disturbance to wind fields. This remoteness is one essential ingredient for global monitoring, the other is a substantial level of logistical and scientific support. With the excellent support and improving facilities provided by the Antarctic Division, Macquarie Island has the potential for playing a major role in future global atmospheric research.

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