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# Carbon isotope measurements of atmospheric $CO_2$ at a coastal station in Antarctica

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ABSTRACT

A 3-years series of weekly 3-days integrated $CO_2$ samples analysed for $^{13}C/^{12}C$ isotopic ratio is presented, as well as a 2-years continuous sample series of 1–2 weeks integration time, analysed for $^{14}C$ activity. The samples have been collected at the German Georg von Neumayer research station situated at the Antarctic coast at 70°S, 8°W. The $^{13}C$ measurements show a slightly decreasing trend in $\delta^{13}C$ of $-0.038 \pm 0.009$‰/year as is to be expected due to the global fossil fuel $CO_2$ release. A significant seasonal variation is not detectable in the $^{13}C$ record. The $\Delta^{14}C$ activity of the atmospheric $CO_2$ samples is constantly *lower* by $-11 \pm 2$‰ if compared to the northern hemisphere clean air level. The opposite effect should be expected from an observed $CO_2$ concentration difference of about 3–4 ppm between the two hemispheres if due to $^{14}C$ free fossil fuel $CO_2$. A total (supposedly regional) $\Delta^{14}C$ depletion of about $-20$‰ to $-25$‰, compared to low latitudes in the southern hemisphere, can be attributed to exchange of atmospheric $CO_2$ with low $^{14}C$ surface water ($\Delta^{14}C \approx -100$‰) due to upwelling in the circum polar region between 50°S and 70°S. This effect can qualitatively be reproduced by a simple one-dimensional atmospheric model.

## 1. Introduction

A permanently manned German Antarctic station was established at Atka Bay (Weddell Sea) in 1981. Soon after, we commenced sampling atmospheric $CO_2$ for $^{13}C$ and $^{14}C$ analysis in addition to measurements of other trace gases and aerosols (Wagenbach et al., 1983). The principal aim was to obtain background values for the isotopic abundances of atmospheric $CO_2$ at a prominent remote site providing most valuable input data to global carbon cycle models. Moreover, combined with other trace gases and aerosol species, they give insight to the geochemical and meteorological regime of this coastal Antarctic site.

In order to allow for a comprehensive interpretation of the (regional) $CO_2$ system, the total set of $CO_2$ data should be available. That, of course, includes continuous $CO_2$ concentration data. Unfortunately, due to funding constraints, only the isotopic analyses could be established. Nevertheless, the isotopic data alone provide useful information on the $CO_2$ budget at the sampling site.

The biogenic contribution to the southern hemisphere yearly $CO_2$ concentration cycle can be determined by $^{13}C$ measurements only. Due to strong isotopic fractionation during $CO_2$ uptake by living plants (isotopic depletion of $^{13}C$ by $\Delta\delta^{13}C \approx -18$‰), and no fractionation during $CO_2$ release from the biosphere, $CO_2$ concentration variations caused by biogenic activity are strongly correlated to corresponding $^{13}C$ variations. This is not true for concentration variations caused by ocean-atmosphere exchange, showing only very little kinetic fractionation (Siegenthaler and Münnich, 1981). However, the strong temperature dependence of the oceanic equilibrium fractionation factor (about 0.13‰ per °C; Mook et al., 1974) may cause an additional isotopic signal at this coastal Antarctic station.

The radioactive carbon isotope $^{14}C$ (half life

5730 yrs) is produced naturally in the lower stratosphere and in the upper troposphere, as well as anthropogenically by atmospheric nuclear weapon testing. On the other hand, carbon derived from fossil fuels contains no $^{14}C$. Thus, observation of the $^{14}C$ in atmospheric $CO_2$ can be used as an indicator of stratospheric air masses or air influenced by fossil fuel combustion. In addition, relaxation of the $^{14}C$ levels following nuclear weapon testing is an important parameter in carbon cycle modelling.

## 2. Sampling site

The German Antarctic station, Georg von Neumayer (GvN), is situated on the Ekström ice-shelf at 70°37′S and 8°22′W, about 6.5 km from the ice-edge. At this coastal site winds are constantly from the east; even during midwinter the sea ice cover is not complete, large polynias offshore the station possibly provide access to surface ocean water, as is indicated by continuous sea-salt measurements (Wagenbach et al., 1983).

The $CO_2$ sampling is performed at some distance from the GvN station (500–1500 m) in a movable van with electrical power supply. Local fossil $CO_2$ contamination originating from fuel consumption at the main station as well as from polar vessels and surface and air traffic (in summer only) is excluded by locating the van upwind of the potential contaminating sources, in addition the integrity of the air is monitored and controlled by wind direction and by the output of a condensation nucleus counter (Wagenbach et al., 1983).

## 3. Experimental

$^{14}C$ measurements have been performed on one- to two-weekly integrated samples. The sampling and analysis technique is the same as used in the study described in an accompanying paper by Levin (1987). All $^{14}C$ activities are expressed as permill deviation ($\Delta^{14}C$ (‰)) from the NBS oxalic acid activity standard, corrected for decay (Stuiver and Polach, 1977). The precision ($1\sigma$) of a single $^{14}C$ analysis is typically $\pm 3$‰.

$^{13}C$ analysis has been made on weekly samples continuously collected over 3 to 4 days. The sampling technique has been described by Dörr and Münnich (1980). All $\delta^{13}C$ values are expressed relative to the PDB standard (Craig, 1957). Due to the specific sampling technique of adsorption in sodium hydroxide solution, the values need not be corrected for $N_2O$ contribution. The mean standard deviation for the chemical extraction and mass spectrometric analysis is typically $\pm 0.05$‰. The long delay time (about 18 months) between preparation of the sample bottles and their extraction and analysis in the laboratory leads to an additional error (correction for storage blank) of $\pm 0.07$‰ in the samples collected before 1983. This problem has, however, been solved meanwhile, by using special rubber gaskets.

## 4. Results and discussion

### 4.1. $^{13}C/^{12}C$ ratio in atmospheric $CO_2$

Fig. 1 shows a quasi-continuous three years $\delta^{13}C$ record of atmospheric $CO_2$. We observe a slightly decreasing trend in $\delta^{13}C$ of $-0.038 \pm 0.009$‰/year as to be expected due to the global fossil fuel consumption still increasing slightly. (Similarly to living plants, fossil fuel $CO_2$ is depleted in $^{13}C/^{12}C$ ratio by about $-20$‰; Tans, 1981). A significant seasonal variation is not detectable in the $^{13}C$ record. It should be mentioned that, contrary to the flask sample programs widely employed using spot samples

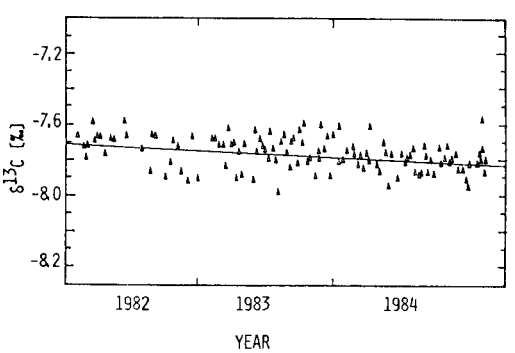


*Fig. 1.* $^{13}C/^{12}C$ isotopic ratio in atmospheric $CO_2$ observed at the German Georg von Neumayer Antarctic station in 70°S, 8°W. The straight line is a least squares fit through the observed data. The decreasing trend of $\delta^{13}C = -0.038 \pm 0.009$‰/yr is due to the global fossil fuel $CO_2$ release.

collected only during special meteorological conditions, the quasi-continuous $CO_2$ isotopic measurements presented here characterize the *true mean* conditions at the sampling site.

The yearly mean $^{13}C/^{12}C$ isotopic ratio in atmospheric $CO_2$ observed at 70°S is higher by $\delta^{13}C = +0.15‰$ to $+0.17‰$ if compared to the yearly mean $\delta^{13}C$ observed at the Schauinsland station (48°N, 8°E; Levin, 1987). The latter data have been corrected for fossil fuel $CO_2$ ($\delta^{13}C = -27.5‰$) and local biogenic perturbations ($\delta^{13}C = -25‰$) according to the concentration deviation estimated by Levin (1987; Figs. 5B and 5C). Similar north-south gradients of $\delta^{13}C$ in atmospheric $CO_2$ (measurements performed in Groningen) have been quoted by Heimann (pers. comm.). However, the absolute values observed by the two different laboratories, Groningen and Heidelberg show a significant deviation of about $\delta^{13}C = 0.2‰$ in the southern as well as in the northern hemisphere data after correction of the Groningen data for $N_2O$ contribution according to Mook and van der Hoek (1984).

Intercalibration with pure $CO_2$ standard gases, on the other hand, did not show a significant effect within the range of $\delta^{13}C = \pm 0.1‰$ (Gonfiantini, 1984). To solve this discrepancy an intercomparison based on new standard gases is presently being undertaken.

### 4.2. $^{14}C$ *in atmospheric* $CO_2$

The $^{14}C$ activity in atmospheric $CO_2$ samples from the GvN is plotted in Fig. 2. The $\Delta^{14}C$ level decreases slightly by $12.9 \pm 1.0‰$ per year—nearly the same rate as observed in the northern hemisphere data discussed in the accompanying paper by Levin (1987). There is little scatter in the data which demonstrates the elimination of contamination during sample collection. There is, however, a significant deviation from the northern hemisphere clean air $^{14}C$ level (Levin et al., 1985) by $-11 \pm 2‰$. An *opposite* effect of roughly the same size would be expected from a $CO_2$ *concentration* difference of 3–4 ppm which is observed between the two hemispheres, if we assume this gradient being exclusively due to the northern hemisphere fossil fuel consumption excess (Rotty, 1983). Indeed, measurements on atmospheric $CO_2$ samples from *low* latitudes of the southern hemisphere performed in Pretoria, South Africa (26°S, 28°E), indicate this *positive* deviation if compared to the northern hemisphere $^{14}C$ level (Vogel, pers. comm.).

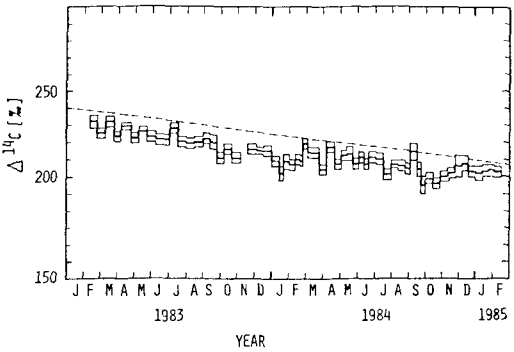


*Fig. 2.* $^{14}C$ in atmospheric $CO_2$ observed at the German GvN Antarctic station. We find a slight decrease in $^{14}C$ activity of $\Delta^{14}C = -12.9 \pm 1.0‰$/yr, nearly the same as observed in northern hemisphere $CO_2$ data (dashed line; Levin et al., 1985). There is, however, a significant $^{14}C$ depletion in the data observed at 70°S of $\Delta^{14}C = -11 \pm 2‰$ if compared to the northern hemisphere clean air level.

Assuming the *low latitude* southern hemisphere $^{14}C$ level being higher than the northern hemisphere clean air level, a total (supposedly regional) $\Delta^{14}C$ depletion at 70°S of about $-20$ to $-25‰$ has to be explained. We attribute this effect to an exchange of atmospheric $CO_2$ with low $^{14}C$ surface water ($\Delta^{14}C \approx -100‰$; Weiss et al., 1979) in the upwelling zone of the circum polar region between 50°S and 70°S, and we try to reproduce it by a one-dimensional atmospheric model.

### 4.3. *Description of the model*

As a first step to a more quantitative discussion of the experimental results we use a one-dimensional (averaged zonally and vertically) multiple box model of the atmosphere with a meridional sine $\phi$ grid ($\phi$ = degrees of latitude) meaning equal zonal surface increments. It can be shown easily (Machta, 1958) that in this case a *uniform* meridional diffusion constant $D$ is represented by a box to box transfer velocity $w(x)$ defined as:

$$w(x) = D(1 - x^2)/H. \quad (1)$$

The following notation has been used in eq. (1):

$x$ = $\sin \phi$

$\phi$ = latitude to be taken at the individual box boundary

$H = r\Delta x$ = box width
$r$ = earth radius
$\Delta x = 2/N$ = dimensionless box width
$N$ = total number of boxes
The net flux $j(x)$ at grid point $x$ then becomes

$$j(x) = w(x)(c(x - \tfrac{1}{2}H) - c(x + \tfrac{1}{2}H)). \quad (2)$$

(Note that the flux transfer velocity is defined at the grid point $x$ whereas the concentration for each individual box for simplicity is always defined in the centre at $x \pm \frac{1}{2}H$.)

The rate of change of concentration within a box is then

$$\Delta c(x \pm \tfrac{1}{2}H)/\Delta t = (j(x) + j(x \pm H))/H + Q(x \pm \tfrac{1}{2}H) \quad (3)$$

with $Q(x \pm \frac{1}{2}H)$ representing the source/sink term of the entire box right or left (i.e., north or south) of the respective grid point $x$.

In our special case we use a grid of 10 *meridional* boxes, but *no* subdivision in the vertical (troposphere and stratosphere being represented by *the same* single box). The horizontal diffusion constant $D$ is set to $3 * 10^{10}$ cm$^2$/s, but with two exceptions:

(1) Setting $D$ constant means, of course, e.g., a negligible additional north-south transfer resistance $R = 1/w(x = 0)$ at the equator (ITC), which, as we know, is not realistic. A reasonable hemispheric residence time of $\tau = 1.5$ years is realised by introducing a transfer velocity of $w(x = 0) = r/\tau$ at the equator.

(2) At the south polar front (due to the relatively coarse box grid in our model at 53°S) we have increased the box-to-box transfer resistance (1) by a factor of two, leading to an increased residence time in the "circum polar atmosphere".

For validation of these assumptions we have tested the model to reproduce the meridional $^{85}$Kr profile as observed in the lower troposphere (Weiss et al., 1983; with extended data from Weiss, pers. comm.). To do this we have assumed a yearly global $^{85}$Kr production of 1.73 pCi m$^{-3}$ yr$^{-1}$ to be released in the latitude belt between 37° and 53°N exclusively (Weiss, pers. comm.). The model is able to describe the north-south gradient at the ITC of about 2.5 pCi/m$^3$ correctly, but produces a lower $^{85}$Kr concentration in the northern hemisphere source region than actually observed there at ground level. This is due to the fact that we implicitly assume complete vertical mixing in the atmosphere, which for ground level sources, ground level observation, and the time span considered is not true, of course.

### 4.4. *Model estimates*

We now discuss the atmospheric $^{14}CO_2$ distribution obtained with the meridional mixing parameters described above and validated by the $^{85}$Kr profile.

The model is designed to reproduce the meridional distribution of $^{14}$C. In the following we will test the model to give (1) the correct natural $^{14}$C level in the atmosphere, and (2) the correct time evolution of the present day nuclear test induced $^{14}$C level. The model can, however not reproduce, e.g., any seasonal $^{14}$C variation such as the one induced by pile-up of nuclear test $^{14}$C in the seasonal ocean mixed layer producing a re-influence on the atmosphere (Broecker and Peng, 1980). Anyway, the absolute size of this specific effect is barely noticeable nowadays, because it is proportional to the atmosphere-ocean level difference which today is nearly an order of magnitude smaller than it was immediately after test stop.

*4.4.1. Pre-industrial $^{14}CO_2$ profile.* For natural $^{14}$C we now have to define the distribution of the sources and sinks: we assume that the steady state $^{14}$C level is not influenced by the biosphere (this is not true for the nuclear test transient time, see below). The influence of atmosphere-ocean gas transfer, however, is considerable. A *mean* residence time of 7.5 years for atmospheric $CO_2$ is used (e.g., $Q_{\text{gas-exchange}}(x \pm \frac{1}{2}H) = a(x \pm \frac{1}{2}H)$ $1/7.5$ $(c_{\text{atm}}(x \pm \frac{1}{2}H) - c_{\text{ocean}}(x \pm \frac{1}{2}H))$ with $a(x \pm \frac{1}{2}H)$ = normalized ocean surface of an individual meridional box), assuming that in the southernmost circum polar box the exchange rate is higher by 50% due to higher wind velocities (Peng et al., 1979). With these gas transfer rates and a mean surface ocean $^{14}$C content ($c_{\text{ocean}}$) of $\Delta^{14}C = -50‰$ for all boxes except the southernmost circumpolar box ($\Delta^{14}C = -100‰$ according to the observed values; Weiss et al. (1979)), the pre-industrial *mean* atmospheric $^{14}$C level ($c_{\text{atm}}$) of $\Delta^{14}C = 0‰$ is reproduced by the model.

(The natural $^{14}C$ source by cosmic ray production is symmetrical with respect to the equator.)

The *pre-industrial* meridional profile produced by the model is plotted in Fig. 3A. The north-south $\Delta^{14}C$ difference of $-4‰$ due to the north-south asymmetry of ocean surface is in very good agreement with the pre-industrial gradient observed from tree ring data. Lerman et al. (1970) found a significant $\Delta^{14}C$ depletion of $-4.5 \pm 1‰$ in southern hemisphere preindustrial tree rings when compared to the northern hemisphere level. Similar gradients have been observed by Vogel and also by Stuiver (Stuiver, pers. comm.).

*4.4.2. Present state $^{14}CO_2$ profile.* To describe the *present state* profile correctly we have to introduce all anthropogenic sources: the $^{14}C$ free (mainly northern hemisphere) fossil fuel source is placed between 37° and 53° N according to Rotty (1983). The prescribed surface ocean $^{14}C$ content

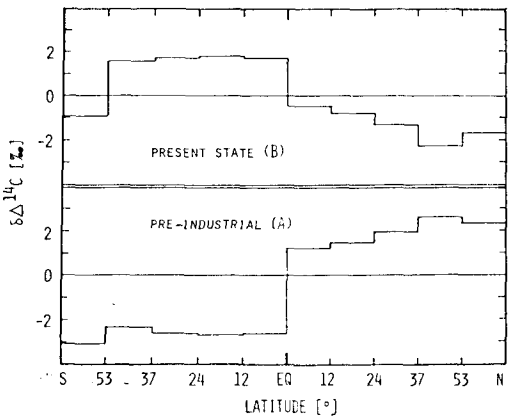


*Fig. 3.* (A) Meridional profile of $^{14}C$ in atmospheric $CO_2$ as calculated by the one-dimensional transport model for pre-industrial time. The plotted $^{14}C$ activities are the deviations from the global mean in the Δ-Scale ($\delta\Delta^{14}C$ /‰/). Due to a larger ocean surface the southern hemisphere $^{14}C$ activity is depleted by about $\Delta^{14}C = 3$–$4‰$ if compared to the northern hemisphere level. (B) Meridional profile of $^{14}C$ in atmospheric $CO_2$ as calculated by the model for the present state. The plotted activities are deviations from the global mean (e.g., 1984: $\Delta^{14}C = 215‰$). The difference between southern and northern hemisphere (now $\Delta^{14}C = +3‰$) if compared to pre-industrial time has changed sign due to the anthropogenic northern hemisphere fossil fuel source. The Antarctic atmosphere $^{14}C$ level, however, still remains low, reflecting gas exchange with low $^{14}C$ surface water in the circum polar zone.

is changed as well, due to bomb $^{14}C$ being already introduced into the ocean. In the model, we use the mean meridional $^{14}C$ concentration distribution observed during the GEOSECS survey from 1972 to 1974 in the Pacific and Atlantic oceans (Stuiver and Östlund, 1980; Östlund and Stuiver, 1980).

In order to reproduce the bomb $^{14}C$ still remaining in the atmosphere at a given time, we start the $^{14}C$ model with an initial atmospheric $^{14}C$ activity of $\Delta^{14}C = 472‰$ (the level of 1972; Levin et al., 1985), homogeneously distributed. A field observation to be modelled correctly then is the present yearly decrease of the global atmospheric $\Delta^{14}C$ level by about $-14‰$, as a result of atmosphere-ocean equilibration removing the pre 1962 nuclear weapon $^{14}C$ spike. In our model described so far the atmospheric $^{14}C$ decline from 1972 to 1984 is faster than actually observed. In part this is due to some post 1962 atmospheric weapon testing (particularly in 1973, 1976, and 1980). More important, however, is that the model contains no biosphere. Contrary to the natural steady state situation the biosphere today releases $^{14}C$ to the atmosphere which has been taken up earlier during the nuclear weapon transient. The first step to make allowance for this is to just increase the apparent size of the atmosphere (a factor of 1.4 must be used to model the observed atmospheric $^{14}C$ decline correctly).

In any case, after a few years running time the model correctly shows the equatorial north-south $\Delta^{14}C$ increase of $3‰$ due to fossil fuel $^{14}C$ dilution in the northern hemisphere (Fig. 3B). The Antarctic draw-down of atmospheric $^{14}C$ by gas exchange with the "old" ($\Delta^{14}C = -100‰$) ocean water due to upwelling is, however, smaller than observed. A larger meridional mixing resistance at the polar front than assumed above would not be in accordance with the $^{85}Kr$ data. Another fact, however, not considered at all so far in the model, is the incomplete vertical mixing of the polar atmosphere. It is easily seen that, restricting the vertical mixing to a more realistic layer thickness of, e.g., 2 km (instead of 8 km) will multiply the effect on the air $^{14}C$ level induced by the "old" $CO_2$ released by the polar ocean with a factor of four. The observed value is, therefore, quite easily explained.

An effect so far not included in the model assumptions is the (regional) influence of strato-

spheric air being particularly enriched in natural $^{14}C$. Stratospheric air masses can partly compensate for the $^{14}C$ depletion produced by atmosphere-ocean gas exchange. A slight indication of this effect is seen in Fig. 2: the $^{14}C$ draw-down in 1984 was smaller than in the year before. A presumably stratospheric influence at 70°S particularly in 1984 is also suggested by other stratospheric tracers (e.g., $^{7}Be$).

## 5. Conclusions

As has been illustrated by the carbon isotope measurements presented here the southern hemisphere $^{14}C$ levels seem to be strongly influenced by ocean-atmosphere interaction. This effect is seen best at a coastal Antarctic site at 70°S filling the gap in $CO_2$ observations between lower southern latitudes and the South Pole station. Future effort will be taken to extend the observation period, hence providing more representative input data for a more sophisticated carbon isotope model (including a horizontal grid of higher resolution as well as a more realistic vertical structure). This model will be valuable also in describing the meridional profile of the $^{13}C$ data.

## 6. Acknowledgements

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