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Radon-calibrated emissions of CO_2 from South Africa

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ABSTRACT

Atmospheric CO_2 and ^{222}Rn have been monitored at Amsterdam Island since 1980. Data were selected in order to eliminate any local influence. Typical CO_2 concentrations of the subantarctic marine atmosphere can be determined by selecting those values for which ^{222}Rn radioactivity was particularly low: less than 1 pCi m^{-3}. ^{222}Rn concentrations higher than 2 pCi m^{-3} are mainly due to injections into the subantarctic atmosphere from the continental source of South Africa. The passage of air masses under continental influence also shows typical CO_2 variations, well correlated with ^{222}Rn variations. From the knowledge of the global continental fluxes of ^{222}Rn, it has been possible to estimate CO_2 fluxes into the atmosphere from South Africa. The mean CO_2 flux corresponding to a 6-month period from May to October is about 5 millimole m^{-2} h^{-1}. Continental CO_2 emissions reach a maximum in August.

1. Introduction

Most CO_2 flux evaluations from various parts of the globe have been made by direct measurements over small areas, typically from a few m^2 to a few km^2 in various ecosystems (de Jong et al., 1974; Ripley and Saugier, 1974; Whittacker and Likens, 1975; Verma and Rosenberg, 1976; Desjardins et al., 1982, 1985; Dörr et al., 1983; Dörr and Münnich, 1987). The aim of this paper is to estimate the global emission of CO_2, from the southern part of the African continent, using comparisons between atmospheric concentration variations of both CO_2 and ^{222}Rn measured in a remote station, Amsterdam Island (37°47′S; 77°31′E). ^{222}Rn has been monitored there since 1967. Since 1980, CO_2 has also been continuously monitored by means of a non-dispersive infrared analyzer (URAS 2T). The results given in the present study are expressed in the WMO 1985 mole fraction in air scale.

The principle of the method is to determine the ^{222}Rn flux emitted into the atmosphere from the soil surface, and in a second step to compare the variations in time of the atmospheric concentrations of both ^{222}Rn and CO_2, then to assume that the CO_2 variations, relatively to those of ^{222}Rn, are proportional to the ratio of their fluxes. This method was successfully utilized on a regional scale. Dörr and Münnich (1987) routinely determined fluxes of CO_2 from soils and respiration of vegetation near Heidelberg, Germany. They found a strong correlation between CO_2 fluxes and soil temperature. CO_2 flux reached maximum values in summer of about 12 millimole m^{-2} h^{-1}, for temperatures close to 20°C, while minimum values of about 2 millimole m^{-2} h^{-1} were reached in winter for temperatures close to −1°C. By using a similar process, Levin (1987), documented CO_2 fluxes in West Germany. The same method used at Porspoder, Brittany, France (unpublished results of our laboratory), gave a nocturnal flux of 11 millimole m^{-2} h^{-1} at the end of the summer 1986. Bonsang and Lambert (1985) similarly used the mean continental ^{222}Rn emission calculated by Lambert et al. (1982) to evaluate the global emission of propane from all the continents.

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In the present study the line of argument followed is slightly different. Individual air masses are considered at Amsterdam Island, in which ^{222}Rn concentrations are high enough, compared to usual figures, to be ascribed to a South African origin. The correlated CO_2 variations are interpreted in term of fluxes, assuming that all dilution processes were identical for both gases from the continent surface. This method will be detailed and discussed further.

2. Atmospheric CO_2 background concentrations over the southern ocean

Even though local influences are weak at Amsterdam Island, they yield CO_2 variations comparable in magnitude to that of air masses from remote continental areas. Therefore they must be clearly identified.

Fig. 1 shows typical examples of local contamination. A high pressure situation prevailed up to 20 June 1983, 3 a.m., with very weak winds. Thus, in that particular case, the first peak of CO_2, labelled "A", corresponded to a non-ambiguous local influence. Simultaneously, ^{222}Rn remained very low. This indicates that the soil of the island, which is able to influence CO_2 concentration, can be insignificant for ^{222}Rn. A different example of local influence is also given in Fig. 1. After 21 June, 3 a.m., the barometric pressure increased whereas both temperature and relative humidity decreased, winds rotated and air crossed the island. Then, the CO_2 peak "B" is also attributed to local influences.

Such local influences can be discarded. When winds arrive at our sampling point from a sector 260° to 300° with velocities higher than 8 m s^{-1}, and from a sector 300° to 360° and 0° to 40° with velocities $\geqslant 5$ m s^{-1}, it has been verified that corresponding CO_2 records are free from any local perturbation (Gaudry et al., 1983; Monfray et al., 1987). However, because of the small size of the island, ^{222}Rn concentration is not an unambiguous indicator of local influences.

It is presently widely admitted that low ^{222}Rn concentrations, less than 1 pCi m^{-3}, characterize "oceanic" air masses, meaning those not influenced by continental emissions for more than one week (Lambert et al., 1982; Polian, 1984). CO_2 concentration values measured during such

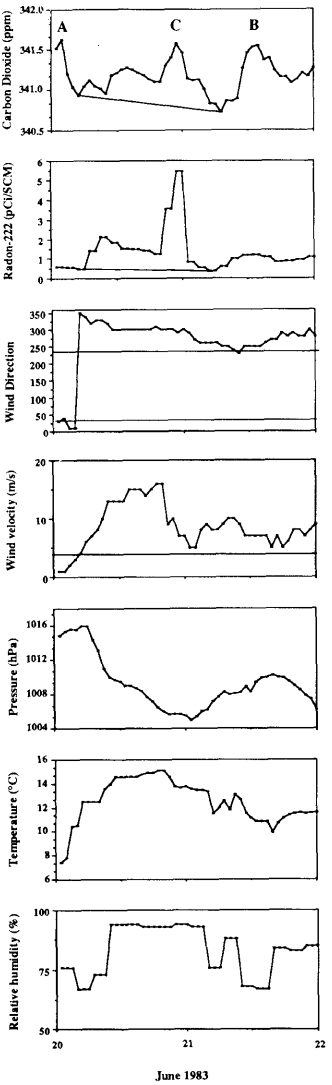


Fig. 1. From top to bottom. Evolution of atmospheric CO_2 recorded at Amsterdam Island between 20 June, 0 a.m. and 21 June, 12 p.m., 1983. Among the 3 peaks labelled "A", "B" and "C", two, "A" and "B" are of local origin. Peak "C" is attributed to a recent continental advection. Evolution of ^{222}Rn, recorded simultaneously. The 5.5 pCi m^{-3} peak of ^{222}Rn corresponds to a clear recent transport of air from South Africa. An excellent correlation between CO_2 peak "C" and ^{222}Rn peak is shown. Evolution of the corresponding meteorological parameters, simultaneously recorded: wind direction and velocity, barometrlc pressure (hPa), air temperature (°C), and relative humidity (%). See text for the use of these parameters to differentiate local from remote events.

periods can be considered as representative of the "oceanic atmosphere", even though the air mass involved may have been influenced by continental emissions several weeks earlier. For recent continental influences on air masses sampled over oceans, ^{222}Rn is a good tracer of their continental origin (Polian et al., 1986).

In Fig. 2, it may be seen that during 2 days corresponding to the passage of a storm, from 30 August 1986, 4 a.m., to the end of 31 August 1986, the ^{222}Rn level remained low, denoting that the air mass did not recently cross a continent, whereas the CO_2 remained practically constant. Just before this event, on 29 August 1986, 0 to 4 a.m., a short local influence is clearly revealed by looking at the meteorological parameters.

However, the CO_2 concentration can also vary, even when the sampled air mass is typically "oceanic". For instance, Fig. 3 shows a significant trough of CO_2, linked to a cyclonic perturbation revealed by a sharp decrease of the barometric pressure, on 28 July 1982, and confirmed by synoptical maps. This meteorological event was not accompanied by a significant ^{222}Rn variation and the synoptical maps confirmed the absence of any injection of "continental air". In fact, variations of approximately 1 pCi m^{-3} above the background concentrations of ^{222}Rn could be attributed as well to a radon release from the sea surface, due to high winds. Thus, this monotonous depletion of CO_2 is attributed rather to its absorption by superficial sea water. It is worth noting that the whole period of July 1982 displayed this kind of decrease, 5 times. During the 7-year data collection such evolutions were very frequent.

The long-term monitoring of CO_2 made it possible, after selection of "oceanic" air masses, to evaluate monthly mean "oceanic" values of CO_2 which are shown in Fig. 4. These oceanic background values differ slightly from monthly mean values already published by Gaudry et al. (1987). However, the increase rates shown in Fig. 4 are practically unchanged. The standard deviation on these monthly mean "oceanic" background concentrations range between 0.16 and 0.79 ppm, depending on the season. This includes the variations during an "oceanic" period, corresponding to a cyclonic perturbation, as well as the variability of CO_2 between different "oceanic" periods, within the same month.

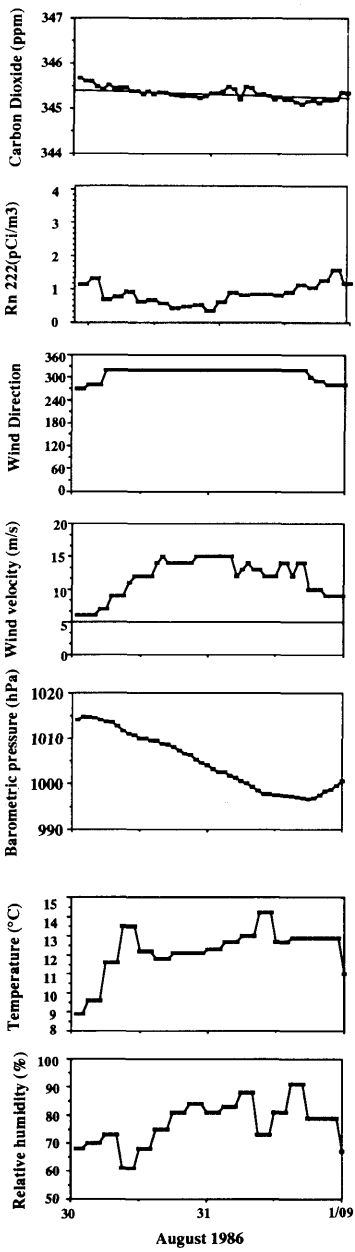


Fig. 2. Passage at Amsterdam Island of a cyclonic perturbation beginning on 30 August 1986, 6 a.m. and ending on 31 August 1986, 12 p.m. From top to bottom. Simultaneous variations of CO_2, ^{222}Rn, and same parameters as in Fig. 1. It can be noted that for this event, the absence of any ^{222}Rn peak identifies an "oceanic" air mass, that has not recently crossed a continental area. The CO_2 variability is very low.

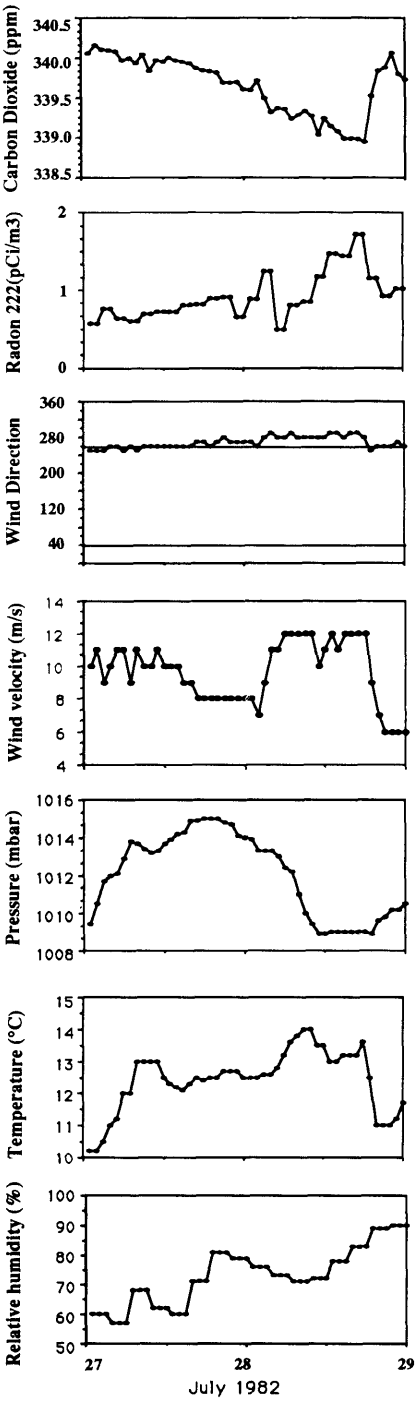


Fig. 3. Passage at Amsterdam Island of a cyclonic perturbation, beginning on 27 July 1982, at 6 p.m. and ending on 28 July 1982, at about 8 p.m. From top to bottom. Variations of the same parameters as in Fig. 2. However, although the ^{222}Rn variability is insignificant which is a characteristic of an "oceanic" air mass, the CO_2 variability is significantly monotonously decreasing by 1 ppm. This shows that ocean is a strong CO_2 sink during this period.

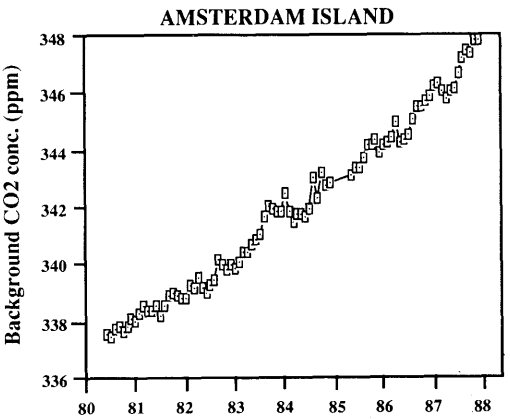


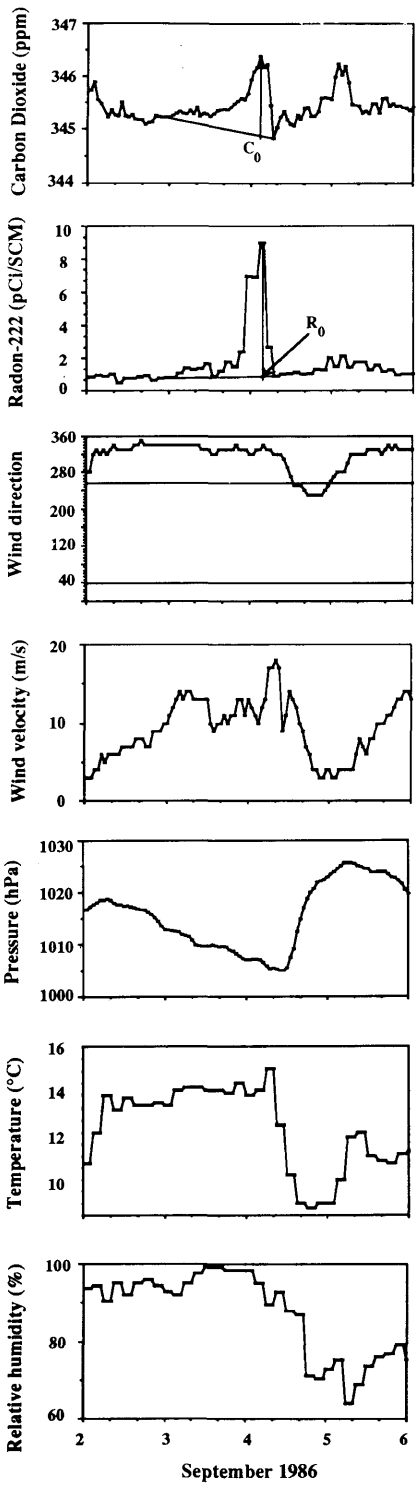
Fig. 4. Monthly mean atmospheric CO_2 concentrations recorded at Amsterdam Island, between May 1980 and November 1987, corresponding to "oceanic" air masses, with $^{222}Rn \leqslant 1$ pCi m^{-3}.

3. Principle of CO_2 emissions evaluation

The basic idea is that we frequently observe at Amsterdam Island peaks of ^{222}Rn well correlated with CO_2 variations (more frequently maxima), for which any local influence can be ruled out. Such events have been interpreted as advections of air masses from South Africa, including Madagascar (Gaudry et al., 1983; Polian et al., 1986). For instance in Fig. 1, beside the already mentioned local peaks A and B, a third peak C is attributed to continental injections.

Fig. 5 shows a clear positive correlation between ^{222}Rn and CO_2 related to the advection of remote "continental" air, transported by a cyclonic perturbation, on 3 to 4 September 1986.

The South African origin of such air masses is supported by several considerations. An examination of the 3-day back-trajectories calculated from Amsterdam Island by the Air Resources laboratories of the NOAA, Silver Spring, Maryland, for the period 1981–1984, has shown the absence of direct westward transports from Australia. However, very scarcely, a mineral-



ogical analysis by Gaudichet et al. (1987) of collected aerosols have shown possible indirect long-range transports from Australia. In these air masses the ^{222}Rn level was too low to be interpreted in term of recent continental advection.

When an air mass first arrives over Africa, the concentrations of ^{222}Rn and CO_2 are typically the above-mentioned "oceanic" values, which can be referred to R_0 and C_0 respectively. "Oceanic" background ^{222}Rn concentration in the subantarctic areas ranges from 0.5 to 1 pCi m^{-3} (Lambert et al., 1982). For CO_2, it may be assumed that the atmospheric concentration of marine air is very close to the oceanic values recorded at Amsterdam Island. Practically, C_0 at this maximum time is determined by linearly interpolating the "oceanic" concentration of atmospheric CO_2, preceding and following any ^{222}Rn maximum as shown in Fig. 5. Above continents, the atmospheric ^{222}Rn concentration increases because of the continuous emissions from the soil surface. The atmospheric ^{222}Rn concentration does vary with wind speed and eddy diffusion, although ^{222}Rn emissions do not vary rapidly in space and time.

For CO_2, the concentration can either increase or decrease according to the direction of Carbon exchanges between atmosphere, soil and vegetation. During the night, the atmospheric CO_2 concentration increases, because of the respiration of the soil and vegetation. During the day, a depletion of CO_2 occurs due to photosynthesis. In winter, photosynthesis is very weak and does not balance the emissions from the soil, so that the winter CO_2 concentrations by day also remain above the oceanic background value. A campaign performed in 1986, between 28 August and 11 September, at Cape Saint Francis, located at the south coast of South Africa, enabled us to verify that continental concentrations were always greater than, or close to those measured in

Fig. 5. Passage at Amsterdam island of a cyclonic perturbation between September 1986, 5 a.m. and 4 September, 1986, 4 p.m. From top to bottom. Variations of the same parameters as in Fig. 2. This figure displays clear positively correlated peaks of CO_2 and ^{222}Rn, attributed to a long distance transport from South Africa. It can be observed, as an example, how the values C_0 for CO_2 and R_0 for ^{222}Rn are determined (see text).

air coming from the sea, even by day. It can be assumed that these conditions prevail during the whole winter. These exchanges of both CO_2 and ^{222}Rn obviously stop as soon as an air mass leaves the continent, with characteristic concentrations C_c and R_c. At that moment, we can assume that the ratio of the differences between the continental and oceanic values: $\Delta C = C_c - C_o$ and $\Delta R = R_c - R_o$ is proportional to the ratio of the fluxes of these gases:

$$\frac{\Delta C}{\Delta R} = \frac{\phi_c}{\phi_R}. \quad (1)$$

The fluxes ϕ are positive for emissions. ΔR is always positive; ΔC can be either positive or negative depending on the prevalence of Carbon emission or absorption processes by the soil and vegetation.

During the transport between South Africa and Amsterdam Island, the concentration differences ΔC and ΔR, similarly evolve following identical atmospheric processes, since the sea surface exchanges can be considered negligible for both gases. It was thus shown by Broecker et al. (1967) and by Hoang and Servant (1972), that the emission rate of ^{222}Rn from the sea surface is about 100 times less than that coming from continental surfaces. For CO_2, the air/sea exchanges depend on the wind strength and on the CO_2 partial pressure differences (ΔpCO_2). According to Monfray (1987) and Etchetto and Merlivat (1988) using the experimental relation of Liss and Merlivat (1986), for $\Delta pCO_2 = 10$ ppm, the exchanged flux is about 0.05 to 0.1 millimole $m^{-2}\ h^{-1}$. Typical values in various continental areas are 10 to 100 times higher.

Moreover, during the transit of an air mass, ^{222}Rn decays with a 3.8-day half-life T_m. Consequently, after correction by a factor $\mu = \exp(0.693t/T_m)$, where t is the transit time, the ratio of these concentration differences, actually measured at Amsterdam Island, $\Delta C^*/\mu\Delta R^*$ should be equal to the initial value of $\Delta C/\Delta R$.

Finally, we have:

$$\phi_c = \phi_R \frac{\Delta C^*}{\mu \Delta R^*}. \quad (2)$$

In eq. (2), ϕ_c is in millimole $m^{-2}\ h^{-1}$, ΔC^* in millimole m^{-3}, ΔR^* in atom m^{-3}, ϕ_R in atom $m^{-2}\ h^{-1}$ and μ a dimensionless factor.

The conversion factors are:

$$\Delta C^* \text{ (mmole m}^{-3}) = \Delta C^* \text{ (ppm)} \times 1/22.4$$

$$\Delta R^* \text{ (atom m}^{-3}) = \Delta R^* \text{ (pCi m}^{-3}) \times 3.7 \times 10^{-2} \times 3.8 \times 86{,}400/0.693.$$

The continental ^{222}Rn flux has been estimated by various methods, between 0.5 and 2.5 atom $cm^{-2}\ s^{-1}$ (Israel, 1951; Servant, 1964; Wilkening et al., 1972; Turekian et al., 1977; Polian, 1984; Mochizuki and Sekikawa, 1978). Polian et al. (1989) proposed an average ^{222}Rn flux of 0.9 atom $cm^{-2}\ s^{-1}$, or 32×10^6 atom $m^{-2}\ h^{-1}$, from an estimate by Lambert et al. (1982) of the global atmospheric pool of ^{222}Rn. It has been shown by Polian et al. (1986) that the transit time (t) between South Africa and Amsterdam Island is relatively constant, about three days. Consequently, $\mu = 1.73$. The possible variability of this factor will be discussed later.

Finally, by taking $\mu = 1.73$ and $\phi_R = 32 \times 10^6$ atom $m^{-2}\ h^{-1}$, we can write:

$$\phi_c \text{ (mmole m}^{-2}\ \text{h}^{-1}) = 47 \frac{\Delta C^* \text{ (ppm)}}{\Delta R^* \text{ (pCi m}^{-3})}. \quad (3)$$

4. Results

During the period under study (May 1980 to November 1987), 94 events were selected, for which peaks of ^{222}Rn and CO_2 variations (peak or dip) were well correlated. In every case, we evaluated the values of ΔC^* and ΔR^* between the maximum and the base of the variations of CO_2 and ^{222}Rn, measured at Amsterdam Island. For the calculations, we preferred to take into account only clear events of ^{222}Rn, well defined by a peak-to-noise ratio higher than 3, the noise varying between 0.6 to 0.9 pCi m^{-3} according to the seasons. Sometimes, when the cyclonic perturbations lasted a long time, several significant peaks of ^{222}Rn occurred during one event, enabling as many estimates of CO_2 fluxes.

All results are listed in Table 1 and summarized by months in Table 2. Table 1 gives a list of all the cyclonic perturbations under study with their timing, as well as the estimates for ΔC^* (in ppm), ΔR^* (in pCi m^{-3}), and ϕ_c (in millimole $m^{-2}\ h^{-1}$). It can be observed that these results only apply to the seasons of fall, winter and early spring (April to November). The lack of results for the complementary period is due to the fact that no ^{222}Rn peak occurred at Amsterdam

Table 1. *Measured variations of ^{222}Rn and CO_2 at Amsterdam island, associated to cyclonic perturbations and corresponding fluxes*

Chronology of the events		Total			
Beginning Year/month/day:hour	End Year/month/day:hour	duration (hours)	ΔC^* (ppm)	ΔR^* (pCi m^{-3})	CO_2 flux (mmole m^{-2} h^{-1})
80/05/29:20	80/05/30:04	8	−0.41	2.7	−7.1
80/06/16:12	80/06/16:19	7	1.06	4.3	11.6
80/07/25:07	80/07/26:07	22	0.4	3.8	5.0
80/07/29:01	80/07/31:02	49	0.65	6.9	4.4
			0.76	9.0	4.0
			1.0	9.8	4.8
80/08/04:12	80/08/05:12	24	0.56	4.6	5.8
80/08/12:16	80/08/13:17	23	1.1	4.4	11.8
80/08/16:17	80/08/18:13	44	0.47	2.7	8.2
			0.76	4.0	8.9
80/11/25:09	80/11/25:22	13	0	5.6	0
80/11/28:04	80/11/28:15	11	0	3.2	0
81/05/29:05	81/05/29:18	13	0.59	5.0	5.6
81/06/23:12	81/06/25:00	36	0	2.9	0
81/08/12:01	81/08/12:09	8	0.88	11.9	3.5
81/08/13:22	81/08/17:06	80	1.41	8.8	7.5
			0.76	4.3	8.3
			0.82	4.8	8.1
81/09/06:22	81/09/09:09	59	0.3	7.8	1.8
81/10/01:12	81/10/02:09	21	0.53	6.21	4.0
81/10/05:00	81/10/05:09	9	−1.42	4.09	−16.5
81/10/06:15	81/10/10:09	96	1.76	5.76	14.4
			0.35	3.64	4.5
			0	2.5	0
			0	3.9	0
82/05/25:20	82/05/26:20	24	1.17	3.2	17.2
			0	2.35	0
82/06/07:06	82/06/08:06	24	2	8.57	11
			0.8	8.93	4.2
82/07/01:01	82/07/03:01	48	1.41	6.4	10.4
			1.2	10.1	5.6
			0	9.6	0
			1.52	9	7.9
			0.7	11	3.0
82/07/26:03	82/07/26:19	16	0.94	9.41	4.7
			0.35	10	1.7
82/08/23:00	82/08/24:12	36	0.5	2.6	9
82/09/24:04	82/09/25:01	21	0.7	3.5	9.3
83/05/05:04	83/05/08:18	82	0	2	0
83/05/25:21	83/05/27:03	30	0.47	4.1	5.4
83/06/03:03	83/06/06:02	70	0.8	3.7	10.2
			0.41	2	9.7
			1.5	5	14.1
			1.1	5.6	9.2
83/06/20:04	83/06/21:00	20	0.59	5.1	5.5
			0.29	1.7	8
83/06/24:01	83/06/25:03	26	1.53	20.5	3.5
83/09/04:02	83/09/05:12	34	0.4	18.5	1.1
84/05/21:00	84/05/21:13	13	0.35	5	3.3
85/05/12:13	85/05/13:02	13	−0.29	3.4	−4.0
85/06/15:09	85/06/15:23	14	0.41	3.5	5.2

Table 1 *continued*

Chronology of the events		Total duration (hours)	ΔC^* (ppm)	ΔR^* (pCi m^{-3})	CO_2 flux (mmole m^{-2} h^{-1})
Beginning Year/month/day:hour	End Year/month/day:hour				
85/07/23:09	85/07/25:00	39	1.52	2.9	24.6
			0.11	1.2	4.3
85/07/06:05	85/07/07:16	34	0.17	3.2	2.5
			0.41	4.1	4.7
85/08/23:20	85/08/24:16	20	0.59	6	4.6
			0.53	9.2	2.7
85/08/04:08	85/08/06:19	35	0	3.2	0
85/09/02:12	85/09/05:00	64	0.23	4	2.7
			0.23	3.5	3.1
			0.65	8	3.8
85/09/22:00	85/09/22:12	12	0.23	3.2	3.4
86/04/26:05	86/04/26:14	9	−0.48	2	−11.2
86/05/13:14	86/05/14:10	20	0.35	6.8	2.4
			0.35	4.1	4.0
86/06/02:00	86/06/02:12	12	0.94	3.17	2.8
86/06/27:00	86/06/28:09	33	0.7	1.8	18.3
			0.76	2.5	14.3
86/07/28:18	86/07/31:23	78	0.7	7.5	4.3
			0.47	5.8	3.8
86/08/08:05	86/08/09:00	19	0.65	6.8	4.5
86/08/11:00	86/08/11:05	05	0.94	2.5	17.7
86/08/27:12	86/08/29:15	51	0.23	1.5	7.2
			0.65	3.3	9.2
86/09/03:12	86/09/04:12	24	1.29	8.1	7.4
86/10/23:06	86/10/24:04	22	0.41	3.5	5.5
86/11/14:00	86/11/14:23	23	−0.35	2.5	−6.6
87/05/23:01	87/05/25:00	47	0.77	3.2	11.3
			0.83	8.1	4.8
87/06/18:02	87/06/19:04	26	0	16.4	0
			0.65	16.8	1.9
			0.48	7.9	2.8
87/06/25:12	87/06/26:12	24	0.61	17.5	1.6
87/06/27:12	87/06/29:04	[illegible]	0.89	22.9	1.9
			0.42	5	4.0
87/07/03:00	87/07/04:04	28	0.83	3.9	10.0
87/07/08:06	87/07/09:18	36	0.42	3.6	5.5
87/07/16:04	87/07/16:18	14	0.48	5.7	4.0
87/07/20:04	87/07/21:04	24	0.54	15.5	1.6
87/08/02:20	87/08/03:12	16	0.83	4.5	8.7
87/08/10:00	87/08/11:17	31	1.19	14	4
87/08/14:06	87/08/15:07	25	0.71	6.4	5.2
87/09/12:00	87/09/12:16	16	0.95	4.5	9.9
87/09/17:11	87/09/18:00	13	0.53	4	6.2

First and second column: Dates of the beginning and the end of the passage of cyclonic perturbations studied.

Third column: Total duration of such events.

Fourth column: CO_2 concentration offsets in ppm, above the "oceanic" conditions and due to a remote continental influence (see text).

Fifth column: ^{222}Rn, same as for CO_2.

Sixth column: CO_2 fluxes calculated by means of relation (3) in millimole m^{-2} h^{-1}. The existence of several estimates for a same meteorological perturbation means that several ^{222}Rn peaks have occurred, permitting several calculations.

Table 2. *CO_2 African net fluxes in millimole m^{-2} h^{-1}, month by month*

Apr.	May	June	July	Aug.	Sept.	Oct.	Nov.
−11.2	−7.1	11.6	5	5.8	1.8	4	0
	5.6	0	4.4	11.8	9.3	−16.5	0
	17.2	11	4	8.2	1.1	14.4	−6.6
	0	4.2	4.8	8.9	2.7	4.5	
	0	10.2	10.3	3.5	3.1	0	
	3.3	9.7	5.6	7.5	3.8	0	
	−4	14.1	0	8.3	3.4	5.5	
	2.4	9.2	7.9	8.1	7.4		
	4	5.5	3	9	9.9		
	11.3	8	4.7	4.6	6.2		
	4.8	3.5	1.7	2.7			
		5.2	24.7	0			
		2.8	4.3	4.5			
		18.3	2.5	17.7			
		14.3	4.7	7.2			
		0	4.3	9.2			
		1.9	3.8	8.7			
		2.8	10	4			
		1.6	5.5	5.2			
		1.9	4				
		4	1.6				
Monthly average -11.2	3.5	6.7	5.6	7.1	4.9	1.7	−2.2
Standard deviation	6.7	5.2	5.0	3.8	3.1	9.4	

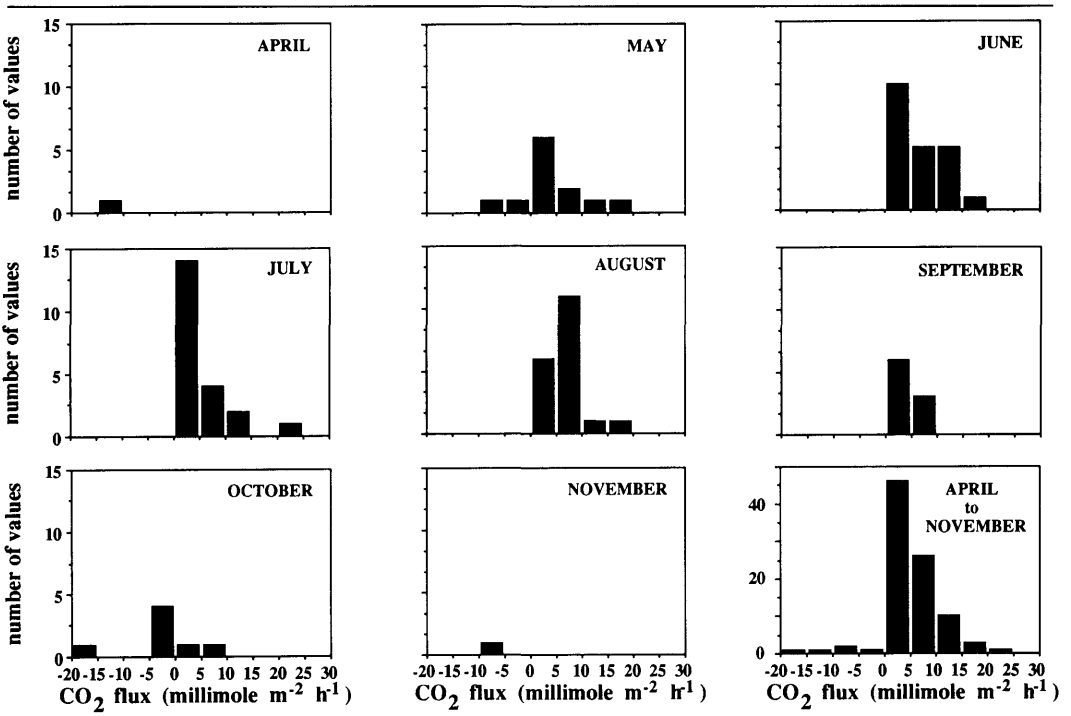


Fig. 6. Histograms of the number of CO_2 fluxes, observed by intervals of 5 millimole m^{-2} h^{-1}, between −20 and +25 millimole m^{-2} h^{-1}, from April to November, for the total data set over 8 years.

Island in summer and late spring, as previously observed by Polian et al. (1986).

In Table 2, the values of CO_2 fluxes range from -16.5 to $+24.7$ millimole m^{-2} h^{-1}. Some negative CO_2 fluxes appear in April, May (austral fall), October and November (austral spring), showing that, in these seasons, the CO_2 absorption by the vegetation prevailed over emission processes. By contrast, in winter the CO_2 flux is always positive. As expected, the average flux is larger in austral winter (June, July and August) than for other seasons.

The 6-month period from May to October corresponds, for the continent, to a significant net release of CO_2 with an average flux of 5.5 millimole m^{-2} h^{-1}. This flux represents the sum of the emissions from soil and vegetation as well as anthropogenic sources. However, the anthropogenic flux, evaluated by Rotty, (1987), is only 0.4 millimole m^{-2} h^{-1} for South Africa. This leads to admit a flux of about 5 millimole m^{-2} h^{-1} for natural processes during this colder season.

Histograms of frequencies of CO_2 fluxes by intervals of 5 millimole m^{-2} h^{-1}, are drawn every month in Fig. 6. In May, data are very spread out, in the range of -10 to $+20$ millimole m^{-2} h^{-1}. No negative value is observed from June to September. For June and July, data are significantly centered on the 0 to 5 millimole m^{-2} h^{-1} interval. The highest value is found in July. In August, data are significantly centered on the 5 to 10 millimole m^{-2} h^{-1}. This month corresponds to the highest mean net release of 7.1 millimole m^{-2} h^{-1}.

5. Discussion and conclusion

Large uncertainties are introduced into our calculations at different steps.

First of all, the precision on ΔR^* and ΔC^* can be estimated at about 20 and 15% respectively. Moreover, transit times of the air masses between South Africa, and Amsterdam Island vary somewhat: 3 ± 1 days, giving an additional uncertainty of about 20% on μ. All these uncertainties lead to a total of about 50% for every ratio $\Delta C^*/\mu\Delta R^*$, but they are random and partly compensated by averaging data.

Second, all CO_2 fluxes in Table 2 are proportional to the global average ^{222}Rn flux of 0.9 atom cm^{-2} s^{-1}, known with an uncertainty of about 25%.

Third, the continental sources of ^{222}Rn and CO_2 are obviously spread differently. However, it may be assumed that as observed from Amsterdam Island, South Africa, including urban areas, could be considered rather homogeneous. Anyway, it is not possible to give any estimate for this kind of uncertainty.

Finally, the mean CO_2 fluxes of 5 millimole m^{-2} h^{-1} should be estimated with a total uncertainty of about 30%.

The large monthly deviations are accounted for by the large variability of CO_2 emissions according to a great number of parameters such as the availability of water and light.

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