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Concentrations and $\delta^{13}C$ values of atmospheric CO_2 from oceanic atmosphere through time: polluted and non-polluted areas

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ABSTRACT

CO_2 is one of the primary agents of global climate changes. The increase of atmospheric CO_2 concentration is essentially related to human-induced emissions and, particularly, to the burning of fossil fuel whose $\delta^{13}C$ values are quite negative. Consequently, an increase of the CO_2 concentration in the atmosphere should be paralleled by a decrease of its $\delta^{13}C$. Continuous and/or spot measurements of CO_2 concentrations were repeatedly carried out during the last decade and in the same period of the year along hemispheric courses from Italy to Antarctica on a vessel of the Italian National Research Program in Antarctica. During these expeditions, discrete air samples were also collected in 4-l Pyrex flasks in order to carry out precise carbon isotope analyses on atmospheric CO_2 from different areas, including theoretically 'clean' open ocean areas, with the main purpose of comparing these open ocean results with the results obtained by the National Oceanic and Atmospheric Administration/World Meteorological Organization (NOAA/WMO) at land-based stations. According to the data obtained for these two variables, a relatively large atmospheric pollution is apparent in the Mediterranean area where the CO_2 concentration has reached the value of 384 ppmv while quite negative $\delta^{13}C$ values have been measured only occasionally. In this area, southerly winds probably help to reduce the effect of atmospheric pollution even though, despite a large variability of CO_2 concentrations, these values are consistently higher than those measured in open ocean areas by a few ppmv to about 10 ppmv. A marked, though non-continuous, pollution is apparent in the area of the Bab-el-Mandeb strait where $\delta^{13}C$ values considerably more negative than in the Central and Southern Red Sea were measured. The concentration of atmospheric CO_2 over the Central Indian Ocean increased from about 361 ppmv at the end of 1996 to about 373 ppmv at the end of 2003 (mean growth rate of about 1.7 ppmv yr^{-1} in excellent agreement with the NOAA/WMO data from land-based stations). Simultaneously, the mean $\delta^{13}C$ value of atmospheric CO_2 over the Central Indian Ocean (Equator) decreased from $-7.92‰$ at the end of 1998 to $-8.22‰$ at the end of 2003; the mean decrease rate being of about $-0.06‰$ yr^{-1}. This rate as well as that calculated at $12°S$ ($-0.05‰$ yr^{-1}) are not far from the rates that may be calculated according to the data from the nearest NOAA sites (Crozet and Mahe islands); the rates calculated South of Australia and between Tasmania and N.Z. are almost identical to those calculated according to the data from Cape Grim NOAA site (Tasmania).

1. Introduction

Despite the massive production of CO_2 related to anthropogenic activities, the CO_2 growth rate in the atmosphere is consistently lower than expected. In fact, recent studies (Houghton et al., 2001; Sarmiento and Gruber, 2002) estimated the average fossil fuel emissions during the 1980s at 5.4 petagrams of carbon per year (1 petagram is synonymous with 1 gigaton and is equal to 10^{15} g: the addition of 1 PgC to the atmosphere increases the concentration of CO_2 by about 0.47 ppmv). The atmospheric growth rate in the same period was 3.3 ± 0.1 PgC yr^{-1}. During the 90s, more than 6 PgC yr^{-1} of man-made CO_2 were added to the atmosphere (Battle et al., 2000): in the same period, the increase of the atmospheric burden of CO_2 was about 2.8 PgC yr^{-1}. The ocean and the land biosphere took up the balance of the CO_2 (IPCC, 1995). The increase of the atmospheric CO_2 concentration is mainly related to the burning of fossil fuel. The $\delta^{13}C$ of fossil fuels is very negative so that the increase of the atmospheric CO_2 concentration should be paralleled by a ^{12}C enrichment. Since 1750, the atmospheric CO_2 concentration has increased by 31%, whereas its $\delta^{13}C$ has decreased by about 2.3‰ (IPCC, 2001).

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During the last few years, continuous and/or spot measurements of atmospheric CO_2 concentrations were repeatedly carried out during ship routes of the vessel 'Italica' (Italian National Research Program in Antarctica) from Italy to Antarctica, along with measurements of the $\delta^{13}C$ of atmospheric CO_2 obtained from discrete air samples collected in 4-l Pyrex flasks. We report here the results obtained between Italy and New Zealand. The results obtained between New Zealand and Antarctica are very different from those reported here and will be discussed in a further paper. The main purpose of this study was to obtain a series of measurements carried out through time along the same ship route and always in the same period of the year. This made it possible to calculate the average growth rate of the CO_2 concentration over the oceans and to compare these data with those obtained at NOAA land-based stations. From the results obtained, the average yearly growth rate of CO_2 concentration can be measured with relatively good reliability, particularly in the central section of the Indian Ocean. However, our attention was particularly drawn by the $\delta^{13}C$ values obtained from the discrete air samples. Light isotopic values can be commonly measured in polluted areas, where the contribution of anthropogenic activity is clearly responsible for anomalous results. The $\delta^{13}C$ values obtained from samples collected in open ocean, far away from continental areas and from possible sources of anthropogenic pollution, should be considered a direct indication of the global conditions of the atmosphere and should be taken as background values for each time interval considered. The comparison among the results obtained in different years can thus outline the possible evolution through time of these parameters, and allow models for their evolution in the near future.

2. Analytical technique

Measurements of the CO_2 concentrations were carried out by means of a Siemens Ultramat 5E analyser assembled for shipboard use (Ori et al., 1996; Lenaz et al., 2000; Longinelli et al., 2001). The Siemens Ultramat 5E measurement system is based on the comparison between the signals from two infrared adsorbing cells, one filled with a flux of synthetic air with constant CO_2 concentration and the other filled with the air sample. Our self-assembled 'marine' version is enclosed in a thermostatic box mounted on suitable shock absorbers and dampers and uses air samples dried by means of a cold trap ($t < -50°C$). The calibrations, using the WMO-X85 scale, were performed with two working standards (345 ppmv and 375 ppmv) whose concentration was determined by comparison with primary standards normally used at the Monte Cimone Observatory (Sestola, Modena, Italy), a baseline site of the Italian meteorological service. Processing of raw CO_2 concentrations was carried out according to the criteria reported by Ori et al. (1996). A numerical smoothing filter was used, according to the suggestion by Thoning et al. (1989) in order to obtain steady-state values ex-

cluding CO_2 values that are short-term variations in the data record. The collection of a continuous record of the atmospheric CO_2 concentrations was planned for all the four expeditions, along the course from Italy to Antarctica. Unfortunately, while the CO_2 analyser yielded excellent results during the first two expeditions (1996–1997 and 1998–1999), during the 2001–2002 expedition a strong transient in the ship power line damaged the Siemens analyser. In order to obtain at least some data along the ship course, it was decided to carry out discrete measurements at a few selected points. During the 2003–2004 expedition, further minor instrumental problems were experienced during the central section of the expedition, preventing the collection of a continuous record between about the equator and 40°S in the Central Indian Ocean. However, the overall trend of the values obtained allows the evaluation of a reliable mean growth rate during the last 7 yr (1996–2003).

Four-litre Pyrex flasks, carefully evacuated to about 10^{-3} mmHg, were used for the collection of discrete air samples. The flasks were opened on the windward side of the ship, about 15 m above sea level, then closed by means of high-vacuum greaseless stopcocks, and stored at room temperature until the docking of the ship at the final harbour in Italy. The separation of the CO_2 was carried out by slow pumping of the flask air at a rate of 7/8 ml m^{-1} through a Pyrex high-vacuum line by means of a Swagelock micro-valve and a liquid nitrogen cooled spiral trap in which H_2O and CO_2 were frozen. The flask was pumped down to about 10^{-3} mmHg and the trap was then isolated by means of high-vacuum stopcocks and heated to about $-80°C$ by means of an ethyl alcohol–liquid nitrogen slush. The evolving CO_2 was collected in a sample tube and measured at the stable isotope laboratory of the Department of Earth Sciences of the University of Parma in a Finnigan Delta S mass spectrometer against a standard CO_2 obtained from a very pure Carrara marble (MAB 99) whose isotopic composition is –2.43 ($\delta^{18}O_{vs.PDB-1}$) and +2.45 ($\delta^{13}C_{vs.PDB-1}$), calibrated against NBS-19 and NBS-20 standards whose isotopic values used for this calibration are: NBS-19, -2.20 ($\delta^{18}O$) and $+1.95$ ($\delta^{13}C$).; NBS-20, -4.14 ($\delta^{18}O$) and -1.06 ($\delta^{13}C$). The standard deviation of the $\delta^{13}C$ measurements was determined by means of repeated measurements on air samples collected at the same place, at the same time, with the same procedure, and treated in the same way. The standard deviations obtained ranged from ±0.02 to $\pm0.04‰$ (1σ). The row isotopic results obtained from flask air samples have been corrected for the N_2O effect according to the equation reported by Mook and Van der Hoek (1983):

$$\delta^{13}c - \delta^{13} = (343 \pm 6)\rho E$$

where $\delta^{13}c$ is the true isotopic value of the CO_2 and δ^{13} is the measured isotopic value calculated as if N_2O was absent, ρ is the N_2O/CO_2 concentration ratio and E is the ratio between the ionization efficiencies for N_2O and CO_2 in the mass spectrometer. We have assumed an N_2O concentration of 316 ppb (mean

value of the data reported by NOAA for the Crozet and Mahe sites, Indian Ocean, in that period) and a CO_2 concentration of 375 ppmv. The value of ρ is therefore 0.842×10^{-3}. The ratio between the mass spectrometer efficiencies at equal inlet pressure of N_2O and CO_2 has been calculated equal to 0.6161.

The theoretical correction for the presence of N_2O is then $+0.1779 \pm 0.003$. The value used is $+0.18‰$.

3. Results

The various sets of measurements of CO_2 concentration (as well as the discrete air samples) were obtained along the same route of the ship: from Mediterranean to the Red Sea, Indian Ocean, Southern Ocean, Tasman Sea, New Zealand (Fig. 1). All the expeditions took place during the same period of the year, from about mid-November to about mid-December. The results obtained measuring the concentration of atmospheric CO_2 during

the four expeditions (1996 to 2003) from Italy to New Zealand are reported graphically in Fig. 2. Through the Southern Equatorial belt (0° to about 30° S in the Central Indian Ocean, apparently one of the most 'clean' oceanic areas), we have mean values of the growth rate of the CO_2 concentration between 1996 and 1998 and between 1998 and 2001 of about 1.8 ppmv yr^{-1}. This value does not agree with the high value, measured in 1998–1999 in the Northern Hemisphere at the Mt. Cimone Observatory [2165 m.a.s.l., Sestola (Modena) Italy], of close to 3 ppmv yr^{-1} (Colombo et al., 2000), in agreement with the NOAA/CMDL global CO_2 measurements for that latitudinal belt (Tans, 2000). This means that different latitudinal belts do not behave in the same way, relatively large differences being apparent among different latitudinal areas. These differences have been already pointed out (Global view-CO_2, 2004) and related to interannual variations in the imbalance between sources and sinks and to variations in atmospheric transport. However, in this case the
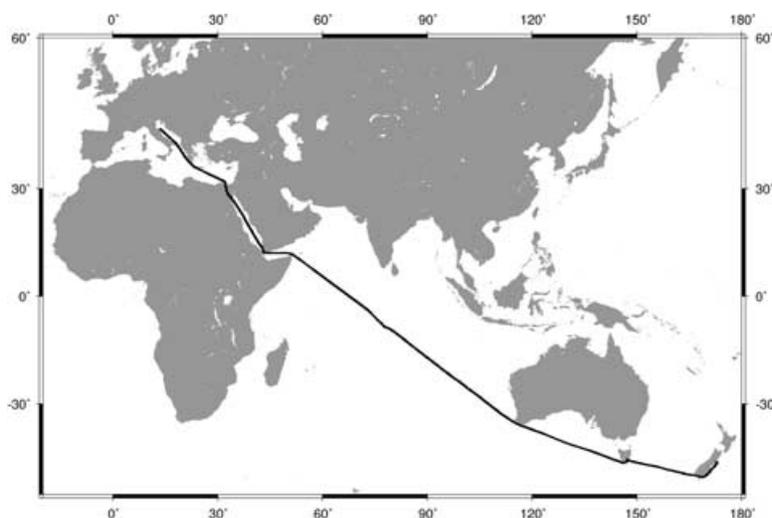
Fig 1. Course of the ship *Italica* from Ravenna, Italy to Littleton, New Zealand.
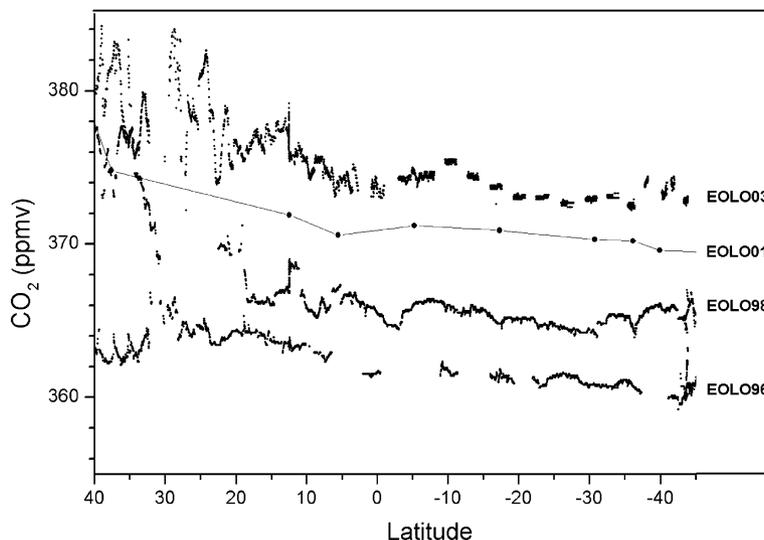
Fig 2. Concentration of atmospheric CO_2 (ppmv) measured between Ravenna (Italy) and Littleton (New Zealand) during four expeditions from 1996 to 2003. In the Indian Ocean section, the 2003 line is fuzzy and incomplete because of instrumental problems.

Table 1.   Carbon isotopic composition of CO_2 and position of the collection sites of discrete air samples between Italy and New Zealand during three expeditions from 1998 to 2003*

1998–99	2001–02	2003–04		Position (2003–04)
		−9.08	44°11.65′N	(off Otranto)
			18°40.81′E	
−8.72	−8.57	−8.51	36°42.16′N	(off Greece)
			21°33.50′E	
−9.08	−8.53	−8.48	31°48.25′N	(North of Port Said)
			31°50.25′E	
−8.23		−8.36	22°03.38′N	(Central Red Sea)
			37°46.88′E	
		−8.39	18°07.78′N	(Southern Red Sea)
			40°12.23′E	
−9.09	−10.03	−9.19	13°00.00′N	(Bab-el-Mandeb)
			43°08.16′E	
		−8.42	12°33.96′N	(S–E of Bab-el-Mandeb)
			43°24.16′E	
−8.18	−8.11	−8.63	12°25.70′N	(Cape Guardafui)
			50°47.91′E	
−7.92	−8.12	−8.22	00°00.30′N	(Indian Ocean)
			67°42.25′E	
	−8.14	−8.25	12°00.12′S	(Indian Ocean)
			83°10.13′E	
−8.12	−8.20	−8.22	35°41.16′S	(South of Australia)
			117°18.89′E	
−8.15		−8.27	45°01.66′S	(between Tasmania and New Zealand)
			158°11.90′E	
−8.05		−8.20	43°50.90′S	(S–E of Littleton, New Zealand)
			173°08.31′E	

*The samples were collected during each expedition between mid November and mid December.

observed difference can be at least partially related to the fact that our measurements were carried out at sea while NOAA/CMDL measurements refer to stations located on land and working normally under more stable atmospheric conditions than at sea. The large CO_2 growth rate measured in the Northern Hemisphere was obviously related to the anthropogenic activity and to meteorological conditions, however, a significant contribution was referred to the most severe and persistent El Niño event of the last years, which took place in 1998. However, if much uncertainty surrounds the interactions between El Niño and long-term global change (Huber and Caballero, 2003), the ocean contribution to the interannual variability of atmospheric CO_2 concentration is also poorly established (Gruber et al., 2002). In some oceanic areas (North Atlantic Ocean), the interannual variability was mainly related to physical processes such as changes in the mixing depth of the ocean surface layer when atmospheric conditions favour the formation of deep surface-mixed layers determining surface ocean temperatures with variable CO_2 uptake (Quay, 2002). In the Equatorial Pacific, there is a general inverse relationship between the maximum CO_2 flux and

the temperature minimum. This relationship has been related to the upwelling and advection of CO_2-rich waters during non-El-Niño periods (Feeley et al., 1999). It is important for the reliability of our measurements to evaluate the mean CO_2 growth rate in the Central Indian Ocean area throughout the 7 yr from 1996 to 2003; the calculated overall growth rate is 1.7 ppmv yr^{-1}, and this value is in very good agreement with the mean growth rate updated in 2000 by the WMO of 1.6 ppmv yr^{-1} (WMO, 2000).

The $\delta^{13}C$ of CO_2 from the discrete air samples collected between Italy and New Zealand (three expeditions) are reported in Table 1 versus PDB-1 standard, along with the position of sample collection (not exactly identical for the samples coming from the same area and collected during different expeditions but fairly representative of the location). PDB-1 standard is used since our working standard (CO_2 from Carrara marble) has been periodically calibrated, throughout the last 45 yr, versus NBS-19 and NBS-20 which, in turn, were calibrated directly versus the original PDB-1. The samples from the Mediterranean, and particularly the first one, collected only a few miles

offshore, show a rather large variability through time, probably related to local meteorological conditions and, particularly, to the wind speed and direction rather than to an evolution of the local situation. However, despite the massive production of CO$_2$ related to anthropogenic activities, the δ^{13}C values obtained from the Mediterranean samples are considerably less negative than expected with the only exception of the 2003–2004 sample collected off Ravenna. The presence of an oil refinery near the Ravenna harbour is probably responsible for this negative value.

The samples collected North–North-West of Port Said show only slightly more negative than expected atmospheric values despite the intense ship traffic in the area of the Suez Canal, the huge urban concentration in the area of the Nile delta (cities of El Iskandaria, El Qahira, El Mansura, El Mahalla, Tanta, Port Said, etc.), and the apparent huge atmospheric pollution in that area. Samples from Central and Southern Red Sea, collected 2 yr apart, show values which may be compared with the δ^{13}C results obtained from the Central Indian Ocean, that yielded carbon isotope values with less of a fossil fuel signal. On the contrary, the Bab-el-Mandeb Strait shows considerably more negative carbon isotope values than expected even though large differences are found between different years. The most reasonable explanation of these results is a heavy contribution to the atmosphere by the discharge from the oil refineries in Djibuti, contribution that North–North-East of Djibuti is variable, probably according to the wind speed and direction. The sample collected S–E of Bab-el-Mandeb is not particularly affected by atmospheric pollution, while the sample collected off Cape Guardafui is slightly more negative than the previous one and consistently more negative than the samples collected 2- and 5-yr earlier at the same location. Also, in this case, the minor variations observed can very probably be related to differences in the meteorological conditions and, particularly, to wind speed and direction since the possibility of a significant contribution from local anthropogenic pollution in this area is rather unlikely. The two samples from the Central Indian Ocean and the sample collected South of Australia are by far the most interesting from the point of view of the evolution through time of the atmospheric CO$_2$ concentration and its progressive enrichment in ^{12}C. In fact, the δ^{13}C values measured during the 1998–1999 expedition at these three locations were close to –8.00 $\pm$ 0.1‰ while during the 2001–2002 and the 2003–2004 the δ^{13}C of the samples that were collected in the same open ocean areas were lower than −8.20‰. Even assuming the most favourable error, the calculated shift of the δ^{13}C values in the case of the samples from the Central Indian Ocean is of about −0.06‰ yr^{-1}, in agreement with the increasing CO$_2$ concentrations, and with the rate (−0.070‰ yr^{-1}) that can be calculated for the same period using data from one of the nearest NOAA Indian Ocean sites (Crozet island, 46°27′S, 51°51′E) (White and Tans, 2005). For the same period (1998–2003) using data from the second nearest NOAA Indian Ocean site (Mahe island, 4°40′S, 55°10′E), a rate of −0.036‰ yr^{-1} can be calcu-

lated (White and Tans, 2005). However, it should be pointed out that the difference between this value and our value is potentially less than the reported standard deviation of our measurements (±0.02‰ to ±0.04‰). It should also be noted that we are attempting to make comparisons between two data sets that differ greatly in spatial and temporal coverage. The samples collected South of Australia, between Tasmania and New Zealand and SE of Littleton (N.Z.) show a considerably smaller shift of the carbon isotope values between 1998 and 2003. This shift ranges from −0.020‰ yr^{-1} to −0.030‰ yr^{-1} in very good agreement with the mean yearly isotopic decrease (−0.026‰ yr^{-1}) that can be calculated for the same period according to the data from the NOAA site at Cape Grim (Tasmania, 40°41′S, 144°41′E) (White and Tans, 2005). The ultimate cause of the differences between the Central Indian Ocean and the Southern Ocean is not clear. A plausible explanation can be related to differences in the dynamic of air masses that is very difficult to reconstruct and to understand without specific measurements and data.

4. Summary and conclusions

Measurements of CO$_2$ concentrations carried out from December 1996 to December 2003 in the Central Indian Ocean allow the calculation of its mean growth rate through time. This value (1.7 ppmv yr^{-1}) is very close to that updated in 2000 by the WMO (1.6 ppmv yr^{-1}) and related to land-based stations. The shift of the δ^{13}C of the atmospheric CO$_2$ in the same area from 1998 to 2003 is very close to that calculated from the data of the NOAA site at Crozet island for the same period and not very far from the same value calculated from the data of the NOAA site at Mahe island (Seychelles). The shift through time of the δ^{13}C calculated for an area South of Australia and in the Tasman Sea is in very good agreement with the isotopic shift calculated from the data of the NOAA site at Cape Grim (Tasmania) for the same period. In the optimistic hypothesis of a yearly increase of the CO$_2$ concentration and a carbon isotope shift remaining equal or very close to the values measured during the last years, in about 30 yr the former variable will exceed 420 ppmv and the latter variable will be close to −10‰. However, more pessimistic estimates have been made by the Hadley Centre model (Cox et al., 2000) according to changes that the carbon system may undergo in response to warming. The expected values of CO$_2$ concentration at the end of this century may range from 713 to 980 ppmv according to different models (Cox et al., 2000; Friedlingstein et al., 2003, respectively).

5. Acknowledgments

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