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## A study of seasonal and yearly modulation of carbon dioxide sources and sinks, with a particular attention to the Boreal Atlantic Ocean

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## Abstract

With the intention of identifying and monitoring space and time patterns of carbon dioxide sources and sinks, the seasonal fields of atmospheric  $CO_2$  concentration over an area covering Europe, the Boreal Atlantic, and North Africa have been computed by using  $CO_2$  observations measured at one or two remote sites in conjunction with the backward air trajectories crossing the same observation sites.

The air trajectories have been calculated by means of the wind speed fields provided by the ECMWF (European Centre of Medium-range Weather Forecast, of Reading, UK) analyses (T213/L31 model) on a regular grid, while the atmospheric  $CO_2$  concentrations have been measured at two alpine European stations, located in the free atmosphere, far from the influence of local industrial pollution.

A modified version of the statistical receptor-to-source-oriented-model (hereafter, source-oriented model) of Stohl (Atmos. Environ. 30 (1998) 947), using the above-mentioned air trajectories, has then been applied to reconstruct the spatial distribution fields of atmospheric CO<sub>2</sub>.

This source-oriented methodology belongs to a family of models which are simpler and easier to use than the more powerful and widespread inverse models and can allow a reliable deduction of the location of sources and sinks of gas tracers.

We have applied this kind of model in order to identify source and sink macro-regions of  $CO_2$  over the abovementioned area in the period 1993–1998. The  $CO_2$  observing stations of Plateau Rosà (3480 m a.s.l., in the western Alps) and Zugspitze (2937 m, in the eastern Alps) have been considered particularly fit for this purpose, because of their location in high orography areas, allowing to monitor values of atmospheric  $CO_2$  concentrations representative of fairly well-mixed air, not affected by some local influences (industries, urban emissions, etc.). In this way, it can be assumed that possible maxima or minima observed in the trend of measured gas concentration can be due to contaminations of the air mass during its whole travel, at some specific locations identified by the source-oriented model.

The most interesting result obtained in this study is the seasonal cycle of the atmospheric  $CO_2$  concentration found over the mid- and sub-tropical latitudes of the Boreal Atlantic Ocean and evident in all simulations. This cycle appears to be clearly related to the seasonal trend of the SST, particularly in the tropical and subtropical Atlantic regions, and is particularly evident during the warmest months (during Spring, Autumn and particularly in Summer).  $\bigcirc$  2002 Elsevier Science Ltd. All rights reserved.

Keywords: Long-range transport; Backward trajectories; SST; Seasonal cycle; Source-oriented relationship; Carbon dioxide

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## 1. Introduction

The atmospheric concentration of  $CO_2$  depends on the balance of the  $CO_2$  global mass in the environmental and climate system of the Earth. The role of the oceans is fundamental for understanding this balance; in fact, depending on a variety of factors, among which the seasonal modulation of the sea surface temperature (SST) takes on a particular relevance, the oceans can absorb or release  $CO_2$ , thus regulating the trend of the  $CO_2$  annual variation.

The study of the ocean contribution to the  $CO_2$  cycle as a function of the seasonal cycle of its SST can be accomplished with a variety of methods.

A method frequently used is the cluster-analysis: it is based upon a correlation between groups of trajectories with similar characteristics (path shape and type, associated potential or pseudopotential temperature) and the mean values of gases, measured at the observation site at the moment of arrival of air trajectories. With this method, the patterns of the trajectories transporting high or low quantities of greenhouse gases are identified (Longhetto et al., 1995, 1997; Brankov et al., 1998; Cape et al., 2000). However, the identification of the corresponding sources or sinks with this method is not always an easy task (Dorling et al., 1992a, b).

Another, and more powerful, method is based on atmospheric transport models, used to constrain sources and sinks of  $CO_2$  by requiring that the modeled spatial and temporal concentration patterns are consistent with the observations (inverse problem, see Masarie and Tans, 1995).

A third kind of approach, based on a receptor-tosource-oriented model (hereafter source-oriented model) and meant to identify in a simpler and faster way source areas of atmospheric gas tracers, was developed by Ashbaugh (1983) and Ashbaugh et al. (1985). This approach allows synoptic inspection of anomalies of CO<sub>2</sub> concentration patterns, which represents the main objective of this study. It has been improved by Seibert et al. (1994) and by Stohl (1996, 1998) who applied this methodology to calculate the sulphate concentration over Europe, while Charron et al. (2000) searched for possible sources of acid rain over France. Using this model, the geographic area covered by air trajectories is split into a grid of cells. For each cell, the concentration field of the studied greenhouse gas is evaluated using an iterative statistical algorithm.

In the present study, a relationship between ocean surface temperature and atmospheric values of  $CO_2$ observed at remote stations has been investigated. The geographic area of this study covers Europe, northern Africa and the Atlantic Ocean in the Northern Hemisphere. The  $CO_2$  concentrations data used refer to two alpine remote stations, located, respectively, at Plateau Rosà (Italy) for the period March 1993–November 1998, and Zugspitze (Germany), for the period March 1996– February 1997. Backwards 3-D trajectories starting at the two monitoring sites have been calculated and a source-oriented model has been developed to compute the  $CO_2$  concentration field. Then, the seasonal fields of  $CO_2$  concentration have been calculated and their trends and variations have been analysed.

### 2. The site locations and the CO<sub>2</sub> concentration data

The first station site, belonging to the Institute of Cosmo-Geophysics, National Research Council (ICG-CNR), Turin, is located at Plateau Rosà (western Alps, 7°42′E, 45°56′N, 3480 m a.s.l.). Considering its elevation and position (East of the slope of Mount Cervino, in the free atmosphere upon an extended snow-clad, bare mountain plateau, far from urban and polluted zones), Plateau Rosà has been considered as a station located in a remote site, suitable for being included in the worldwide network (World Meteorological Organization (WMO)) devoted to the monitoring of greenhouse-gas concentrations uncontaminated by local anthropogenic effects. Here, the main greenhouse gas  $(CO_2)$  has been regularly measured by Centro Elettrotecnico Sperimentale Italiano (CESI), since April 1989. Weekly data, obtained with the flask method, were collected from April 1989 to December 1997. Since March 1993, a continuous time-series of half-hourly data is available. Other greenhouse gases, like N2O, O3, CFCl3, CH4, have been measured only during shorter and sporadic periods, wherefore any meaningful comparison with these longer lived species was not feasible.

In this study, half-hourly concentration values of  $CO_2$  measured from March 1993 to November 1998 have been used; these values are automatically averaged every 30 min by the software of the  $CO_2$  analyzer on the basis of the instantaneous data collected every 2 s. The  $CO_2$  concentrations are referred to the international X85 WMO scale and expressed in ppmv units. Each data is the result of the analysis of air samples performed by a non-dispersive infrared analyser (model Siemens UL-TRAMAT—5E) working at a sampling rate of 0.5 Hz in the range 335–385 ppmv with a precision of 0.05 ppmv at 360 ppmv.

The  $CO_2$  analyser of Plateau Rosà is calibrated with the international primary, secondary and tertiary standards, respectively once a year, every 3 days and every 6h. Before their processing, data have been averaged by us over periods of 6h in order to filter out short-term and random local and regional sources and sinks. In addition, they have been also seasonally detrended (see Section 4) in order to smooth the vegetation cycle effects due to alpine European forests around the observation sites. The second station is located at Zugspitze  $(10^{\circ}59'E, 47^{\circ}25'N, 2937 \text{ m a.s.l.})$  near the highest peak of the German Alps and, for this reason, is also a remote station, included in the WMO network. The data used in this paper belong to the Umweltboundesamt Federal Environmental Agency (WMO: WDCGG CD-ROM no. 5, March 1999), and cover a shorter period than Plateau Rosà, from March 1996 to February 1997. The instrument used for the measurements is the URAS 3G analyser, and the values have been calibrated according to the scale X85 WMO, and expressed in ppmv unit.

By the way, the  $CO_2$  concentration data collected during 1993 and 1994 years at the Plateau Rosà station have been compared with other European station: Monte Cimone (44°11′N, 10°42′E, 2165 m a.s.l.) and Izana (28°18′N, 16°29′W, 2367 m a.s.l.). In both comparisons the datasets appear to be homogeneous and consistent.

Regarding the Plateau Rosà station, an inspection of the 12-months-running-mean of the  $CO_2$  concentration time series in the period 1989–1999 (Fig. 1), reveals an evident change of slope in the yearly trend during the years 1992–1993. In fact, the observed trend during the decade 1989–1999 is 1.45 ppmv/year while in the period 1993–1998 is 1.77 ppmv/year. These two trends are separated by a decreasing of concentrations.

One possible explanation for this fact could base itself on secondary effects of the Pinatubo volcanic eruption in 1991. Isotopic data suggest that the low growth rate resulted from fluctuations in the exchanges of  $CO_2$ between the atmosphere and both the ocean and the terrestrial biosphere, possibly resulting from climatic and biospheric variations following the eruption (Houghton et al., 1996; Parker et al., 1996). These variations could also support the assumption that the drop in the mean atmospheric temperature (due to a decrease of the solar radiation, brought about by increased atmospheric turbidity) could slow down the  $CO_2$  production from the vegetation.

Of course, other effects could have contributed to lower the  $CO_2$  concentration in that period (for instance, the ENSO cycle).

An empirical orthogonal function analysis of monthly mean Pacific SST (Newell and Weare, 1977) has shown a spatial and temporal correspondence between changes in Pacific SST and changes in atmospheric  $CO_2$ , after seasonal trends have been removed.

It has been suggested (Newell and Weare, 1977; Newell et al., 1978), that this could be related to the oceanic upwelling, whose effect is the increasing uptake of atmospheric  $CO_2$  by the nutrient-rich cool water from intermediate layers. The annual increase of the atmospheric  $CO_2$  during the anomalous El Nino-years, when upwelling is replaced by warm tropical waters is about twice as large as in years with strong upwelling.

As a matter of fact, a comparison we made with the cold (la Nina) and warm (El Nino) episodes calendar did not reveal any sure relationship between these episodes and our calculated atmospheric  $CO_2$  concentrations.

#### 3. The trajectory model

3-D backward trajectories have been computed in order to reconstruct the path of an air parcel arriving at the  $CO_2$  monitoring sites.

The trajectory model has been developed at the Department of General Physics of the University of Torino (Anfossi et al., 1988). Trajectories are computed using the tri-dimensional wind fields provided by the T213/L31 model running at the ECMWF (European Centre of Medium-range Weather Forecast of Reading, UK). The dataset used in this study had the following characteristics: four analyses per day (at 00, 06, 12, 18



Fig. 1. Monthly average (thin line) and running monthly average of 12th order (thick line) of  $CO_2$  concentrations measured at Plateau Rosà, Italy (45°54′N, 7°42′E, 3480 m a.s.l.) from April 1989 to December 1999.

GMT and at 10 pressure levels: 1000, 850, 700, 500, 400, 300, 250, 200, 150, 100 hPa), a grid step of  $0.5^{\circ} \times 0.5^{\circ}$  in latitude and longitude, and an extension of the considered geographical area from  $5^{\circ}$ N to  $70^{\circ}$ N in latitude and from  $60^{\circ}$ W to  $45^{\circ}$ E in longitude.

Four trajectories were calculated daily at the synoptic hours (00, 06, 12, 18 GMT). Trajectory points were computed every 36 min by interpolating the wind speed fields. The interpolators used are a bicubic relation on the horizontal plane (as suggested by Walmsley and Mailhot, 1982), a linear relation on the vertical and a parabolic one in the time (Longhetto et al., 1995, 1997). In this study, only trajectories composed by at least 200 points, i.e. 5 days were considered.

For each trajectory, a statistical analysis of its pressure (p) and pseudopotential temperature  $(\vartheta_p)$  at every time step has been performed, in order to estimate both the fluctuations of the flight altitude and the extent of loss and uptake processes.

The results of this analysis, given in terms of standard deviation of p and  $\vartheta_p$  following the motion of each individual air mass, show that the most frequent value for  $\sigma_p$  was 80 hPa and 3.5 K for  $\sigma_{\vartheta_p}$ , all through the 5-day period. This shows that, on the average, most air trajectories undergo noticeable vertical displacements, mainly due to the Alpine complex terrain, while their mixing with other air masses along their path is limited.

### 4. The source-oriented model

The source-oriented model, originally developed by Ashbaugh (1983) and Ashbaugh et al. (1985), then improved by Seibert et al. (1994) and by Stohl (1996), was meant for assessing sulphate concentration patterns. In the present paper, it has been adapted and optimized to consider the previously mentioned  $CO_2$  concentration measurements. As a first step, the data measured every half an hour at the monitoring sites have been processed in order to remove the annual trend and the seasonal variations. The following relation is used:

$$c_l = c_{l0} \frac{\overline{c_{21}}}{\overline{c_{\text{period}}}},\tag{1}$$

where  $c_l$  is the concentration corrected for the annual variation,  $c_{l0}$  is the concentration measured at the trajectory arrival on a site (averaged over the 3h preceding and following the trajectory arrival),  $\bar{c}_{21}$  is the 21 days running mean concentration, and finally  $\bar{c}_{\text{period}}$  is the mean concentration computed over the whole considered period. Hereafter, in this paper, the measured values detrended according to Eq. (1) will be referred as "CO<sub>2</sub> concentrations".

The considered region has been split into  $m \times n$  cells; each cell has a spatial extension of 1.5° in longitude and latitude. The logarithmic concentration field, weighted with the residence time  $\tau_{nnl}$  in the grid element (m, n) of the *l*th trajectory, has been calculated in each cell according to the following relation:

$$\bar{C}_{mn} = \frac{\sum_{l} \tau_{mnl} \operatorname{Log}(c_{l})}{\sum_{l} \tau_{mnl}},$$
(2)

where  $\tau_{mnl}$  is the residence time in the grid element (m, n) of the *l*th trajectory.

The final field is then calculated following an iterative procedure composed by eight steps:

- (1) The trajectory l is split in  $N_l$  points.
- (2) The mean concentration in the grid cell hit by the *i*th point of the *l*th trajectory is computed by means of the relation

$$X_{il} = 10^{C_{mn}(i)}.$$
 (3)

(3) The average of the mean concentrations (3) over all grid cells hit by the points of the *l*th trajectory is calculated as

$$\bar{X}_{l} = \frac{\sum_{j=1}^{N_{l}} X_{jl}}{N_{l}}.$$
(4)

(4) The concentrations (4) are redistributed along the trajectory as

$$c_{il} = c_l \frac{X_{il} N_l}{\sum_{j=1}^{N_l} X_{jl}}.$$
 (5)

- (5) The process from points (1) to (4) is recursively repeated for all trajectories.
- (6) The new logarithmic concentration fields are computed using the equation:

$$\bar{C}_{mn} = \frac{1}{\sum_{l=1}^{M} \sum_{i=1}^{N_l} \tau_{mnli}} \sum_{l=1}^{M} \sum_{i=1}^{N_l} \log(c_{il}) \tau_{mnli}.$$
 (6)

- (7) A nine point smoothing to the gridded values is applied. The value over each grid point is calculated as the weighted average between the considered point and its surrounding points; the central point has a weight of 1.0, the horizontally and vertically nearest ones have a weight of 0.5, and the corner points have a weight of 0.25.
- (8) The process is iterated until one or more maxima (minima), whose intensity equal to c<sub>l</sub> + 2σ (c<sub>l</sub> - 2σ) are present. This value can be considered, with good approximation, as the absolute maximum (minimum) of all measured values.

An analysis of sensitivity and errors made by this method in the concentration maps was made before applying it to this study (Ferrarese, 2002). In this paper, the model sensitivity has been estimated by simulating some sets of schematic air trajectories, and by assigning "a-priori" both the position of some sources in given cells of an horizontal grid, and the atmospheric concentrations of the tracer associated to each trajectory crossing pre-assigned receptor points. The conditions allowing the model to locate correctly the sources, together with a useful rule for dealing with the grid cells crossed by a limited number of trajectories have been established. These conditions have been used in this study.

Anyway, it must be remarked that, besides the already mentioned filtering effect of the data averaged over a period of 6 h, the model itself contributes to smooth short-term, sporadic local or regional sources and sinks. In fact, when the trajectory sample is large enough (ideally, infinite; in practice, made up of a number of trajectories allowing at least one intersections at each grid cell during the observation period), sporadic uptakes or losses of  $CO_2$ , do to the changes of altitude of the trajectory or to the passage over short-term local sources or sinks, are filtered, and only stationary or slowly changing sources and sinks of  $CO_2$  are recorded as high or low local signals of features, which are analysed as meaningful strengths of emission of sequestration of  $CO_2$ .

## 5. Results

The source-oriented model has been applied to the seasonal sets of trajectories crossing the monitoring station of Plateau Rosà. At the beginning, seasonal year by year analyses from 1993 to 1998 have been performed and they are discussed in Section 5.1. In this work, seasons are defined in the following way: Spring refers to the months March-May; Summer to June-August; and so on. In order to reduce the number of figures, only the set of analyses made in 1994 is reported here. Afterwards, the analysis has been applied to the whole period, from 1993 to 1998, obtaining four CO<sub>2</sub> patterns, each of them representing a 6-years average. The choice of 1994 was motivated by the fact that this year exhibited a particularly clear seasonal trend of CO<sub>2</sub> concentration, but the same features of the seasonal cycle are also evident in the others years, as it can be noticed when examining the graphics referring to the whole period.

## 5.1. Analyses carried out at Plateau Rosà year by year

The concentration levels, referring to the borderline of the isocontoured regions of Figs. 2a–d (as well as the zones near to blank areas without intersecting trajectories) must be considered not significant, and will not be considered in the discussion because the trajectory overlapping in these regions is too sparse. The  $CO_2$  computed fields relevant to Spring, Summer, Autumn and Winter 1994 are reported in Figs. 2a–d, respectively.

The concentration field of  $CO_2$  in Spring (Fig. 2a) shows medium-low values over the Atlantic Ocean at latitudes lower than 50°N, medium values at latitudes higher than 50°N and two distinct maxima: the first one over the centre of Italy (spreading from the Tyrrhenian Sea to the Balkans), and the second one located North of Scotland. In Fig. 2b, relative to Summer, considerable maxima in the southern extratropical Atlantic Ocean are present. The Atlantic basin appears then separated into two distinct latitudinal regions: the first one, below 50°N, with the maxima, and the second one, above 50°N, with the minima. In Fig. 2c, showing the concentration fields during Autumn, the area with the Summer maxima seems to be dissolved, and only an isolated maximum, residual of the large area in the Summer, is persisting on the southern extratropical Atlantic Ocean. Other maxima are present over the South of Iceland, over Germany and over the Mediterranean Sea (even if, in this last case, we must remember that this region is located at the borderline). Finally, Fig. 2d, relative to Winter, shows a field that looks similar to the one observed in Spring (Fig. 2a). The minima are located at a latitude below 50°N, while at the higher latitudes there are some local and relative maxima; a maximum is still persisting over the Mediterranean Sea.

By analysing these fields, we can conclude that the concentration values of CO2 over the Atlantic Ocean in the Northern Hemisphere seem well related with the seasonal surface temperatures. Indeed, during Spring, there are medium-low concentration values over the southern part of the extratropical Atlantic Ocean; they become high during Summer; during Autumn (when the sea surface cools down with a delay with respect to atmosphere) they remain medium-high, and they are again medium-low in Winter. The maxima and minima over the continental region during the whole year, and over the Atlantic Ocean in the Northern Hemisphere in Spring and Winter, are probably caused by the continental anthropogenic activity or by the long-range transport across the Ocean from the heavily urbanized areas of the North America.

Concerning the maxima and minima over the continental region all the year round and over the Boreal Atlantic Ocean in Spring and Winter, it is important to observe that the seasonal detrend operated over the observed data at Plateau Rosá and Zugspitze rules out the effect of alpine vegetation cycle. Of course, this remark does not apply to the Summer maxima over the southern extratropical Atlantic Ocean, because the vegetation cycle is out-of-phase in this season.



Fig. 2. (a) Simulated concentration fields of  $CO_2$  using 257 backward trajectories started at Plateau Rosà during Spring 1994. The position of Plateau Rosà is shown by a black point, the trajectory length is 5 days. (b) Simulated concentration fields of  $CO_2$  using 160 backward trajectories started at Plateau Rosà during Summer 1994. The position of Plateau Rosà is shown by a black point, the trajectory length is 5 days. (c) Simulated concentration fields of  $CO_2$  using 211 backward trajectories started at Plateau Rosà is shown by a black point, the trajectory length is 5 days. (d) Simulated concentration fields of  $CO_2$  using 238 backward trajectories started at Plateau Rosà during Winter 1994. The position of Plateau Rosà is shown by a black point, the trajectory length is 5 days. (d) Simulated concentration fields of  $CO_2$  using 238 backward trajectories started at Plateau Rosà during Winter 1994. The position of Plateau Rosà is shown by a black point, the trajectory length is 5 days.

# 5.2. Analysis carried out at Plateau Rosà on the entire 6-years period

The same analysis has been repeated for the whole period (6 years), and the corresponding fields are displayed in Figs. 3a–d.

In Fig. 3a, relative to all Springs, it is possible to distinguish two zones separated by an imaginary line starting from Ireland and crossing the left boundary of

the grid at 35°N. Medium-high values are prevailing northwest of this line, while the minima prevail southeast of it, with the only exception of the maximum off the Gibraltar coast. Medium values are reported over the western Mediterranean Sea, the Denmark and the northern Germany, while some maxima are present over the Black Sea and the eastern Europe. Fig. 3b, relative to all Summers, allows to identify two regions over the Atlantic Ocean. The first one is characterized by



Fig. 3. (a) Simulated concentration fields of  $CO_2$  using 1712 backward trajectories started at Plateau Rosà during Springs from 1993 to 1998. The position of Plateau Rosà is shown by a black point, the trajectory length is 5 days. (b) Simulated concentration fields of  $CO_2$  using 1362 backward trajectories started at Plateau Rosà during Summers from 1993 to 1998. The position of Plateau Rosà is shown by a black point, the trajectory length is 5 days. (c) Simulated concentration fields of  $CO_2$  using 1567 backward trajectories started at Plateau Rosà during Autumns from 1993 to 1998. The position of Plateau Rosà is shown by a black point, the trajectory length is 5 days. (d) Simulated concentration fields of  $CO_2$  using 1290 backward trajectories started at Plateau Rosà during Winters from 1993 to 1997. The position of Plateau Rosà is shown by a black point, the trajectory length is 5 days.

maximum values at the latitude below 50°N, while in the second one (above 50°N) there are some minima, with the only exception of some maxima located near the Greenland, from the eastern Iceland to Scandinavia and Finland. Fig. 3c, showing all Autumns is a transition figure. Over the southern belt of the extratropical Atlantic Ocean, the Summer maxima are being substituted by the Winter minima. Over the continent, some

maxima are appearing over northern Germany, over the Mediterranean Sea and over the western coasts of the Black Sea. Fig. 3d, relative to all Winters, shows a non-homogeneous field. Medium-high values are prevailing over the Atlantic Ocean above the latitude  $50^{\circ}$ N, while the minima are located at a latitude lower than  $50^{\circ}$ N. Some relative maxima are present also over the northern Atlantic Ocean between the latitudes  $55^{\circ}$ N -  $60^{\circ}$ N, over



Fig. 4. (a) Simulated concentration fields of  $CO_2$  using 643 backward trajectories started at Plateau Rosà and Zugspitze during Spring 1996. The position of Plateau Rosà is shown by a black point and that one of Zugspitze is shown by a black square; the trajectory length is 5 days. (b) Simulated concentration fields of  $CO_2$  using 620 backward trajectories started at Plateau Rosà and Zugspitze during Summer 1996. The position of Plateau Rosà is shown by a black point and that one of Zugspitze is shown by a black square; the trajectory length is 5 days. (c) Simulated concentration fields of  $CO_2$  using 586 backward trajectories started at Plateau Rosà and Zugspitze during Autumn 1996. The position of Plateau Rosà is shown by a black point and that one of Zugspitze is shown by a black square; the trajectory length is 5 days. (d) Simulated concentration fields of  $CO_2$  using 630 backward trajectories started at Plateau Rosà and Zugspitze during Winter 1996. The position of Plateau Rosà is shown by a black point and that one of Zugspitze is shown by a black square; the trajectory length is 5 days. (d) Simulated concentration fields of  $CO_2$  using 630 backward trajectories started at Plateau Rosà and Zugspitze during Winter 1996. The position of Plateau Rosà is shown by a black point and that one of Zugspitze is shown by a black square; the trajectory length is 5 days. (d) Simulated concentration fields of  $CO_2$  using 630 backward trajectories started at Plateau Rosà and Zugspitze during Winter 1996. The position of Plateau Rosà is shown by a black point and that one of Zugspitze is shown by a black square; the trajectory length is 5 days.

England, Denmark and Russia, while an absolute maximum is located over Scandinavia and another one is present over the Adriatic Sea, stretching from Greece to the central zone of the Mediterranean basin.

This analysis involves a longer period and a wider spatial area with respect to the previous one; for this reason, the results are more reliable. The results have confirmed the preliminary conclusions drawn after examining the year 1994. Furthermore, the region of the Atlantic Ocean located between 25°N and 50°N is influenced by the seasonal surface temperature trend. In this area, some minima during Spring, some maxima during Summer, isolated maxima during Autumn and some minima during Winter are clearly distinguishable. The continent and the northern Atlantic Ocean, on the contrary, are characterized by zones of minimum or maximum due to other phenomena (perhaps anthropogenic activity or transport).

## 5.3. Analyses carried out at Plateau Rosà and Zugspitze during year 1996

The concentration fields of  $CO_2$ , calculated using the observations collected at the two remote stations of Plateau Rosà and Zugspitze during the unique overlapping year 1996, are shown in Figs. 4a–d. In these figures, the  $CO_2$  concentration scale has been enlarged for an adequate representation of the field.

In Fig. 4a, relative to Spring, an isolated maximum located over the Atlantic Ocean (near 35°N) and another one located northwestwards of Scotland are present. Over the continent, the maxima are placed between France and Germany, in Poland and over the Mediterranean Sea. During Summer (Fig. 4b), the maximum located in Spring at the latitude 35°N grows up over a wide region covering the whole Atlantic Ocean below the latitude 50°N, while the other maxima are located over the western Mediterranean Sea, Central Italy and eastern Europe. In Autumn (Fig. 4c), the values of the CO<sub>2</sub> concentration fields over the Ocean decrease; over the continent, and especially over Central and eastern Europe, on the contrary, the concentrations grow. The maxima located over Spain and the Balkans are not significant, because they are near to zones crossed by too few air trajectories, as already mentioned before (beginning of Section 5.1). Finally, Fig. 4d, relative to Winter, shows a more homogeneous field than the one for Autumn; then, it is possible to observe that, on average, the values over the Atlantic Ocean are higher at latitudes above 45°N than at latitudes below 45°N.

By comparing Figs. 4a-d with the previous ones (in which the concentrations have been calculated by using only the data of Plateau Rosà station), it is possible to observe that the same annual cycle is present over the southern extratropical Atlantic Ocean: minima in Spring, maxima in Summer, isolated maxima in Autumn and minima in Winter. The only difference between the two figure series is in the absolute values of CO<sub>2</sub> concentrations. This difference is due to the detrending procedure: in fact, the values of Figs. 4a-d have been detrended only referring to the year 1996, while the other values have been detrended on the whole 6-year period, and then they are referring to the year 1995. By using the CO<sub>2</sub> concentration trend observed during the decade 1993–1998 at Plateau Rosà (1.77 ppmv/year, see Fig. 1), an average difference of about 1-2 ppmv between the two figure series can be expected.

#### 6. Conclusions

The results of this study have shown that the  $CO_2$  concentration fields, calculated in the recent years over an area including Europe, northern Africa and the Boreal Atlantic Ocean, display a seasonal cycle that seems well related with the seasonal variations of SST.

In particular, over the Boreal Atlantic basin, the  $CO_2$  concentration field seems to be particularly subject to seasonal variations that are correlated with the seasonal cycle of the SST. In fact, during Spring, the fields exhibit medium and low values, but during Summer the increase of the SST (stronger near the Tropic of Cancer) seems in good agreement with the appearance of intense maxima of atmospheric  $CO_2$  concentration over the southern part of the extratropical Atlantic Ocean.

The increase in the SST during warmer months (due to intensification of solar radiation, connected with the seasonal variations of the solar elevation) explains why Autumn concentration fields show their maxima in the southern belt of the extratropical Atlantic Ocean, and the values of these maxima are weaker. In Winter, the fields show low values except for the northern region of Atlantic Ocean, where the  $CO_2$  concentration fields might be influenced not only by the SST, but also by other unknown factors.

In any case, if the existence of a relationship between the  $CO_2$  concentration fields and the SST is assumed, it is expected to observe, in the intertropical region of the Northern Hemisphere, a zone of maxima oscillating, during the year, from North (in Summer) to South (in Winter) and vice versa. This study has demonstrated the presence of this oscillation during the first three seasons of the year (Spring, Summer and Autumn), and particularly during Summer, when the largest maxima are located in the southern belt of the Boreal Atlantic Ocean involved in our analysis.

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