Localization of source and sink regions of carbon dioxide through the method of the synoptic air trajectory statistics

F. Apadula\textsuperscript{a,*}, A. Gotti\textsuperscript{a}, A. Pigini\textsuperscript{a}, A. Longhetto\textsuperscript{b}, F. Rocchetti\textsuperscript{a}, C. Cassardo\textsuperscript{b}, S. Ferrarese\textsuperscript{b}, R. Forza\textsuperscript{b}

\textsuperscript{a}CESI SpA, Centro Elettrotecnico Sperimentale Italiano, via Rubattino 54, Milan 20134, Italy
\textsuperscript{b}Department of General Physics, University of Turin, Turin, Italy

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Abstract

The main purpose of this paper is to contribute to the improvement of the present knowledge concerning the transient components of the global carbon cycle, superimposed to the periodic seasonal oscillation and to the yearly trend. This purpose has been achieved through the comparison among the calculated concentration fields of atmospheric CO\textsubscript{2} and its comparison with the sea-surface temperature patterns, forestation maps, forest fires, and the anthropogenic emissions extracted from Edgar V.2.0 database. In order to identify with high spatial resolution the most relevant areas of CO\textsubscript{2} sources and sinks, we have applied a methodology based on a statistical analysis of simulated back-trajectories related to atmospheric concentration values measured at some receptor sites where the back-trajectories originate.

In particular, we have used a 2-year time series (1996 and 1997) of CO\textsubscript{2} concentration data observed in three receptor sites located in high mountain areas, in order to reduce significantly the effects due to local influences (such as emissions from industries and urban areas or the absorption processes due to the vegetation). The back-trajectories were computed by means of the wind fields provided by the ECMWF analysis (T213/L31 model) on a regular grid. The area investigated was from 11°W to 36°E in longitude and from 30°N to 57°N in latitude. The final concentration field was computed by means of a statistical source–receptor model, based on a methodology developed by Stohl (Atmos. Environ. 30 (1996) 579) and adapted here with some modifications in the pre- and post-processing phases.

Before applying the model, a careful evaluation of its sensitivity to the input data has been performed, followed by an analysis to identify the optimal configuration of the model.

The results have shown a satisfactory accuracy in the identification of the major sources and sinks considered.

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

1. Introduction

The growth of the atmospheric concentration levels of greenhouse gases such as carbon dioxide, methane, nitrous oxide, halocarbons (so-called CFCs) and tropospheric ozone since the industrial period (1750) has been identified as one of the major causes of the general warming of the Earth’s surface. Recent studies (IPCC WGI, 2001) have demonstrated that, in 1999–2000, the CO\textsubscript{2} alone has been responsible for about 60% of the increase in radiative forcing due to the emissions of all greenhouse gases.
The background CO$_2$ atmospheric concentration has been steadily increasing for at least 200 years, from around 280 ppmv (parts per million by volume), corresponding at the pre-industrial period, to 367 ppmv in 1999 (IPCC WGI, 2001). The CO$_2$ growth rate has been about 1.5 ppmv (0.4%) per year over the past two decades. During the 1990s, the year to year increase varied from 0.9 ppmv (0.2%) to 2.8 ppmv (0.8%).

As mentioned before, this increase can contribute to a great extent towards the enhancement of the greenhouse effect which forces the atmospheric global warming and climatic changes. For this reason, a better understanding of the global carbon cycle and of its natural and anthropogenic sources and sinks is crucial for the prediction of the future CO$_2$ concentration levels.

It is well known today that the atmospheric CO$_2$ concentration exhibits, besides a secular trend, a seasonal cycle with a maximum occurring from late winter to early spring and a minimum in late summer. Superimposed on this seasonal cycle are present some short-term fluctuations in the CO$_2$ concentration, that prompted this study because they reflect the presence of regional or sub-regional sources and sinks (Higuchi et al., 1987), easily retrievable in ground-based or remote-sensed data inventories.

Different approaches have been used to improve the knowledge concerning both quantitative estimates of sources and sinks of carbon dioxide, and seasonal distribution patterns of their locations over the Earth.

The inversion methods are proper to the first kind of approach. This methodology is based on the use of numerical models of atmospheric transport to translate the CO$_2$ mixing ratio patterns into sources and sinks by requiring that the modelled spatial and temporal concentration distributions are consistent with the observations. In this sense, an inverse approach works in an opposite manner with respect to the traditional one, which uses the emission values as input data and applies a transport and diffusion model to calculate the concentration fields. Several methodologies exist to solve the inverse model; in one of them, the model of atmospheric transport redistributes the CO$_2$ at each time step; the sources and sinks in the bottom layer are initially set equal to their values during the previous time step. The discrepancies between model calculations and observations at the end of each time step are used to infer the necessary change of sources or sinks to reduce them. The time step is then repeated with the corrected values of sources and sinks. In a few iterations, the model converges to the sources/sinks required for each latitude (Tans et al., 1989; Conway et al., 1994; Masarie and Tans, 1995).

Serious obstacles to this approach are some specific weak points pertaining to the world monitoring network. In particular, scarcity of monitoring sites, lack of temporal continuity of the measurements in some stations, and the fact that not all monitoring stations have covered the same observation periods (Masarie et al., 2001) might be potentially misinterpreted by the transport models, producing derived source/sink scenarios that are biased.

The second kind of approaches includes different methodologies, some of which are based on back-trajectory analyses. These methodologies have been applied to study source–receptor relationships. One of these approaches, the cluster analysis, has been used by Moody and Samson (1989), Harris and Kahl (1990) and Longhetto et al. (1995, 1997). Cluster analysis is a multivariate statistical technique that splits a data set (in this case the trajectory data set) into a number of groups or clusters. All trajectories having similar characteristics (path shape and type, potential or pseudo-potential temperature, and so on) are grouped together, and a relationship is searched between each group and the mean values of the gas measured at the moment of the arrival of the back-trajectories at the observation site. This method allows one to relate air pollution data with particular trajectory clusters, but can hardly produce reliable identification of sources and sinks.

During the 1980s, Ashbaugh (1983) and Ashbaugh et al. (1985) developed a method specifically designed to identify source areas; afterwards Seibert et al. (1994) and Stohl (1996) improved this methodology. They split the domain of trajectory computation into grid cells. In each cell, the concentration field was calculated by using an interactive statistical algorithm. This methodology, which does not convert CO$_2$ atmospheric concentration data (ppmv) into CO$_2$ emission data (kg s$^{-1}$) but only takes concentration anomalies to be a proxy of sources and sinks, was applied by Stohl (1996), Ferrarese et al. (2002), Charron et al. (2000) and by Aalto et al. (2002) to localize source areas of sulphate, carbon dioxide, acid rain over Europe and France and the concentration field of several compounds over North-Europe, respectively, and by Stohl et al. (2001) to investigate the distribution and the sources of ozone (O$_3$) observed in the uppermost troposphere and lowermost stratosphere.

The last two methods can be considered as a useful tool to support the inverse model approach in the localization of sources and sinks.

In this paper, we have applied this last methodology with a few modifications regarding in particular the pre- and post-processing phases.

2. Methodology

Our methodology consists in a statistical approach (Stohl, 1996; Ferrarese et al., 2002) that combines CO$_2$ concentration data measured at some monitoring stations (also called receptors sites) with backward
trajectories crossing the same observation sites. This statistical approach acts in such a way to determine gridded concentration field maps representing, at each grid cell, the vertical average column concentration of CO₂ from the surface to the free troposphere, which make it possible to localize the spatial distribution of CO₂ sources and sinks.

This kind of approach does not allow one to distinguish between biogenical and anthropogenical sources, but it can identify, with a good spatial resolution, the localization of source and sink areas. In this work, the evaluation of the type of source has been carried out by comparing the final result with other information such as the geographical area (for example, continental or marine), emission inventory data, spatial distribution of forest fires, and sea-surface temperature (SST) patterns.

2.1. Data set and locations

We have used CO₂ concentration data measured in three different monitoring stations over Europe: Plateau Rosa, Monte Cimone and Wank Peak Zugspitze. These three receptors are located in high orography areas in order to prevent pollution and absorption processes due to local influences (such as CO₂ industrial and urban emissions, and photosynthetic activity).

Plateau Rosa station is located in the Italian Alps (7.70°E, 45.93°N, 3480m a.s.l.); it is the highest atmospheric monitoring station in Europe and belongs to the Institute of Interplanetary Space Physics, Turin Section, National Research Council. The CO₂ measurement program has been run by CESI since April 1989. The atmospheric CO₂ concentration, referred to the X85 WMO international scale and expressed in ppmv in dry air, is measured by means of a non-dispersive infrared analyzer (NDIR) working in the range 335–385ppmv with a precision of 0.05ppmv at 360ppmv.

Fig. 1 shows the time trend of the monthly mean values of the CO₂ background concentrations from April 1989 to June 2001. The solid line represents the 12-month running mean. The calculated annual trend during this period is 1.52ppmv/year.

Monte Cimone station is located in the Italian Apennines (10.70°E, 44.20°N, 2165m a.s.l.) on a mountain top, about 500m above the tree line. The CO₂ measurement programme has been run by the Italian Air Force Meteorological Service since March 1979 (Colombo and Santaguida, 2000). The continuous observation of atmospheric CO₂ concentration began with a URAS-2T non-dispersive infrared analyzer and afterwards, since November 1988, went on with an ULTRAMAT 5E analyzer. Monte Cimone measurements are referred to the X93 WMO international scale.

Wank Peak Zugspitze station (10.59°E, 47.25°N, 2937m a.s.l.) is located near the highest peak of German Alps. The instrument used for the CO₂ measurements is the URAS 3G analyzer and the concentration data, expressed in ppmv, are referred to the X85 WMO international scale. The station and the data belong to the Umweltbundesamt Federal Environmental Agency.

![Figure 1: Monthly mean and yearly trend of CO₂ background concentration measured at Plateau Rosa from April 1989 to June 2001 (source web site: www.cesi.it/greeninfo/monitoraggio).](image-url)
and the measurements program has been run since 1981 (Scheel and Sladkovic, 2001).

We have merged data measured with X85 WMO international scale and X93 international scale because CMDL has showed, through repeated intercalibrations with the primary standards of SIO, no significant offset among the WMO mole fraction scale and the X85 and X93 scales (GLOBALVIEW-CO2, 2001).

The carbon dioxide concentration data used in this study cover the period from January 1996 to December 1997, since this time interval was characterized from a maximum contemporary coverage of CO2 data at monitoring stations.

In order to prevent the influences of local sinks and according to the suggestion of Cundari et al. (1995), we have decided to discard all hourly averages of Monte Cimone station data collected from 09:00 a.m. to 09:00 p.m. in the period from mid-May to mid-September. In fact, in this period, the photosynthetic activity of the local vegetation affects the CO2 measurements, bringing about a systematic depletion of several ppmv in the CO2 diurnal cycle.

2.2. The trajectory model (TRAIEET)

The trajectory model named Tri-dimensional Atmospheric Interpolation Evaluation of Trajectory (TRAIEET), developed by Reap (1972) and later modified by Anfossi et al. (1988), has been used in this paper. The backward trajectories have been calculated using the three-dimensional wind fields provided by the T213/L31 weather prediction model on regular grid, running at ECMWF. We have used four analyses per day (0, 6, 12, 18 UTC) at ten different pressure levels (1000, 850, 700, 500, 400, 300, 250, 200, 150, 100 hPa) relative to a grid of 0.5° × 0.5° and ranging from 11°W to 36°E in longitude and from 30°N to 57°N in latitude. The time extent of each trajectory, starting from any receptor site, was 5 days back and its departure times were 0, 6, 12, 18 UTC for each day.

Starting from the horizontal and vertical wind fields of the ECMWF analysis, TRAIEET calculates the points of each trajectory every 18 min by means of a spatial and temporal interpolation code. The interpolators are: bicubic on the horizontal plane (Walmsley and Mailhot, 1982), linear in the vertical direction and parabolic in time.

2.3. The source–receptor model (ISOGASP)

The source–receptor model applied in this study, named Identification of Sources of greenhouse Gases Plus (ISOGASP), descends from the statistical methodology developed by Stohl (1996) and originally proposed by Seibert et al. (1994). This model identifies sources and sinks areas of CO2 by means of a statistical method. In particular, a cell is labelled as a source (sink) when a prevalent part of the large number of trajectories crossing the same cell from various directions, over the whole observation period (month, season, year), is associated to high (low) CO2 values detected in the monitoring stations (or receptors points). Furthermore, also the residence time of the trajectory in the cell is taken into account (Stohl, 1996; Ferrarese et al., 2002). More details on this model can be found in the above quoted papers. In this paper we only describe the procedures that we have applied to the pre- and post-processing phases.

After removing the long-term time trend and the cyclic seasonal component as suggested by Stohl (1996), in the pre-processing phase we have rejected all CO2 concentration data laying outside the range \( \bar{c}_{\text{period}} \pm 2\sigma \), where \( \bar{c}_{\text{period}} \) is the CO2 mean concentration calculated over the whole analyzed period and \( \sigma \) is the standard deviation associated with \( \bar{c}_{\text{period}} \). As it will be explained in the next section, this procedure (that actually corresponds to a pre-processing filter) has been adopted in order to prevent some anomalous episodes of short duration from obscuring continuous emissions thus affecting the final result.

With the purpose to mitigate the increasing uncertainty in the computed positions of the back-trajectories with the backward time (Stohl et al., 1995), which can affect the accuracy of the identification of source and sink areas, a nine-point smoothing filter (Stohl, 1996) has been applied to the gridded final concentration field. Therefore, the result on each grid point is a weighted average of the considered grid cell and of the surrounding cells.

In this study the central cell was given a weight of 1.0 while the other eight cells were given a weight of 0.25.

3. Test of the method

In order to test the ability of ISOGASP to correctly identify source and sink areas of carbon dioxide, we have performed several simulations introducing in the grid some selected cells representing ad hoc stationary sources and sinks of pre-assigned strength. Stationary sources and sinks are easier to be dealt with this method than non-stationary sources and sinks. We have selected stationary sources and sinks for this sensibility test because we only wanted to verify if the statistical approach performed well in this situation, and this result could be better achieved with a test of the method addressing a stationary problem. Anyway, on the basis of the considerations of the first part of Section 2.3 and as it will be quite evident in the results, this method can solve both stationary and non-stationary sources and sinks.
As we have already said before, in our simulation we have considered the whole free troposphere like a single layer. In this way, every cell corresponds to an air column and the advection from the surrounding cell (columns) is considered negligible, because it is very unlikely that this kind of phenomena can be so much systematic to become prominent.

The reasonable assumption made here is that the CO₂ concentration value associated with back-trajectories arriving to three receptor points (Plateau Rosa, Monte Cimone and Zugspitze) after crossing one of these ad hoc cells should be greater (source cell) or lower (sink cell) than the concentration value associated with every other back-trajectory that did not cross such kind of cells.

In detail, we have forced six source and six sink cells, hereafter called check points, with the geographical coordinates given in Table 1 (in total, 12 check points with assigned CO₂ emission or absorption strengths, expressed as CO₂ concentrations).

In this test, the concentration values associated with the back-trajectories crossing the other cells in the computation domain corresponded to assigned values randomly generated in the concentration range from 350 to 360 ppmv.

The final result of this simulation is represented in Fig. 2, showing that all 12 check points, i.e. the six forced sources and six sinks, are well identified. In this figure, and also in Figs. 5–7 and 11, we have represented the computed average column CO₂ concentration up to the tropopause level. Because of the large lack of homogeneity of the CO₂ values randomly assigned to the tropopause level. Because of the large lack of homogeneity of the CO₂ values randomly assigned to the tropopause level. Because of the large lack of homogeneity of the CO₂ values randomly assigned to the tropopause level.

Table 1
Geographical coordinates of the 12 check points forced

<table>
<thead>
<tr>
<th>Cell type</th>
<th>Longitude</th>
<th>Latitude</th>
<th>Concentration value assigned (ppmv)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Source 1</td>
<td>From 3.0°E to 3.5°E</td>
<td>From 53.0°N to 53.5°N</td>
<td>363</td>
</tr>
<tr>
<td>Source 2</td>
<td>From 3.0°E to 3.5°E</td>
<td>From 47.5°N to 48.0°N</td>
<td>364</td>
</tr>
<tr>
<td>Source 3</td>
<td>From 3.0°E to 3.5°E</td>
<td>From 42.0°N to 42.5°N</td>
<td>365</td>
</tr>
<tr>
<td>Source 4</td>
<td>From 9.0°E to 9.5°E</td>
<td>From 53.0°N to 53.5°N</td>
<td>366</td>
</tr>
<tr>
<td>Source 5</td>
<td>From 9.0°E to 9.5°E</td>
<td>From 47.5°N to 48.0°N</td>
<td>367</td>
</tr>
<tr>
<td>Source 6</td>
<td>From 9.0°E to 9.5°E</td>
<td>From 42.0°N to 42.5°N</td>
<td>368</td>
</tr>
<tr>
<td>Sink 1</td>
<td>From 8.5°W to 8.03W</td>
<td>From 53.0°N to 53.5°N</td>
<td>348</td>
</tr>
<tr>
<td>Sink 2</td>
<td>From 8.5°W to 8.03W</td>
<td>From 44.5°N to 45.0°N</td>
<td>346</td>
</tr>
<tr>
<td>Sink 3</td>
<td>From 8.5°W to 8.03W</td>
<td>From 42.0°N to 42.5°N</td>
<td>345</td>
</tr>
<tr>
<td>Sink 4</td>
<td>From 3.5°W to 3.0°W</td>
<td>From 53.0°N to 53.5°N</td>
<td>344</td>
</tr>
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<td>Sink 5</td>
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<td>From 44.5°N to 45.0°N</td>
<td>343</td>
</tr>
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<td>From 3.5°W to 3.0°W</td>
<td>From 42.0°N to 42.5°N</td>
<td>342</td>
</tr>
</tbody>
</table>

The iteration process due to the Stohl methodology (Stohl, 1996; Stohl et al., 2001) emphasizes the extreme values of the output field, allowing a better spatial resolution between the source and sink areas. For this reason the Stohl’s methodology is very efficient to localize source and sink region but, as already mentioned in Section 1, does not yield any quantitative CO₂ flux estimates. In this way, the CO₂ output values are connected to the emission rate, but indicate the grade of appropriate classification of the area in terms of sink, source and background region.

After several simulations and comparisons, we have identified and adopted an optimal representation scale, defined by means of the computed median and standard deviation of the final concentration field values. In particular, we have defined the following seven intervals centred with respect to the median value of each final field obtained: (−∞, −3σ), [−3σ, −2σ), [−2σ, −σ), [−σ, +σ), (+σ, +2σ], (+2σ, +3σ], and (+3σ, +∞). This scale has been utilized for all simulations performed in this study. In this way, the adopted scale is a part of our methodology, allowing the best representation of the output field.

A hint for a test concerning the accuracy of the computed trajectories has been given by the occurrence of an anomalous short-lived episode of very large fluctuations of CO₂ concentrations observed almost simultaneously at Plateau Rosa and Zugspitze. The episode took place in December 1997 and lasted only a few days, starting from the early afternoon of 30 November and ending to the late evening of 5 December: the dashed and solid curves of Fig. 3, referring respectively to Plateau Rosa and Zugspitze, clearly show that CO₂ concentrations exhibited larger fluctuations first at Plateau Rosa and then, with a
Fig. 2. Column concentration field of CO$_2$ obtained introducing 12 ad hoc sources and sinks. The simulation has involved the Plateau Rosa, Monte Cimone and Zugspitze receptor sites during 1997. The cells with less than 10 trajectories were left blank.

Fig. 3. Six-hour mean of CO$_2$ column concentration during the episode in winter 1997 after removing the secular trend, the cyclic seasonal component and subtracting the annual mean of each station.
similar shape and an average time lag of less than 1 day,
at Zugspitze.

Fig. 4 shows the pathways of four back-trajectories calculated in the middle of the episode, arriving at Zugspitze, respectively, on 2 December at 12:00 and 18:00 UTC and the day after (on 3 December) at 00:00 and 06:00 UTC. This figure emphasizes that the same air masses that crossed Plateau Rosa during the episode reached Zugspitze later, with a delay of the order of 12–18 h.

Fig. 5 shows the final concentration field of carbon dioxide during 1997. The simulation has been conducted with three receptor sites (Plateau Rosa, Monte Cimone and Zugspitze) and with all the CO2 data measured in these three monitoring stations (including the data of the above-quoted anomalous episode). Fig. 6 represents the same simulation but adopting the pre-processing filter consisting in the exclusion of the CO2 data outside the range $\tau_{\text{red}} \pm 2\sigma$ (in this manner, the anomalous episode is rejected).

The comparison between these last two figures shows how effectively a single and isolated intensive episode is able to hide a pattern of less intense but interesting continuous and steady emissive situation.

Because in this study we are principally interested in the investigation of biogenic and anthropogenic sources and sinks, we have applied this kind of pre-processing filter in all the following model simulations.

4. Results

Fig. 7 shows the final CO2 concentration field relevant to the warm semester (from May to October) of the years 1996–1997. In this simulation, like in any other one, the resulting CO2 concentration field, according to Stohl (1996) and Ferrarese (2002), has to be interpreted with caution in all those areas that are close to the border of the resolved computation domain, because a relatively small number of trajectories have crossed these areas so that the statistical significance can be lower.

The orange- and red-colored spots in this figure, corresponding to CO2 concentration anomalies higher than 2 ppmv, reveal the likely presence of CO2 sources over a wide area that covers the Westerly Mediterranean Sea, the south-east of the Iberian Peninsula and the grazing lands of the African Northern coasts (Algeria and Morocco). A second source area is visible between
Czech Republic and Slovakia. Main regions of removal (CO₂ sinks) are located, respectively, in Eastern Germany, Ireland and the surrounding areas over the Atlantic Ocean, the alpine region between Italy and Austria and in the Northern and Central Tyrrenian Sea.

In order to interpret the positive anomalies found in this analysis, we have firstly compared our CO₂ patterns with the forest fires in the warm semester. In fact, forest fires are the second most important source of CO₂ emissions after the industrial ones (see World Fire Web on http://natural-hazards.jrc.it/fires/detection/wfw/help/).
overview.html, but, contrary to the latter, they are mainly active during the warm semester. In addition, forest fires modify the ability of the vegetation cover to act as a carbon sink.

Fig. 8 shows a composite scene of forests fires, for the period 15–21 July 1997. This map shows the presence of a great number of fires in the Iberian Peninsula and in the Morocco’s and Algeria’s coasts, that agree with the positive anomalies (source areas) depicted in Fig. 7. There are also other zones in the map of Fig. 8 characterized by a high number of fires (i.e. Sardinia and Sicily) that are not associated with high values of simulated CO₂ in Fig. 7. This missed correspondence is probably due to the fact that, in these regions, the forest fires were too scattered and sparse with respect to the former ones, and the atmospheric condition were not favourable to long-distance transport. Furthermore, it could also be attributable to a difference in the combustion efficiencies of the fires in the different regions (Matsueda and Inoue, 1999).

The other simulated positive anomalies of CO₂, visible in Fig. 7, respectively, over the eastern Atlantic Ocean of the Morocco coast and the western and south-western Mediterranean Sea, and in the area between Czech Republic and Slovakia, are not connected with forest fires. The Atlantic and Mediterranean positive anomalies

Fig. 7. Column concentration field of CO₂ during the warm semester in 1996 and 1997 as computed with the source–receptor model by using three receptor sites (Plateau Rosa–Zugspitze–Monte Cimone) and 3222 trajectories. Data outside \( r_{\text{median}} \pm 2\sigma \) have been removed by applying the pre-processing filter. Those cells which have been crossed by less than 20 trajectories are left blank.

Fig. 8. Fire map during 15–21 July 1997. The red colour represents the burnt area (source: JRC; see web site http://natural-hazards.jrc.it/fires/detection/wfw/).
could be related (Ferrarese et al., 2002) to the positive anomaly of the SST. In fact, the sea can behave as a source or a sink according to the period of the year. SST plays an important role in the complex mechanisms that regulate the gas fluxes at the water–air interface, and it is common knowledge that the higher the SST value, the larger the $\Delta pCO_2$ (gradient of partial pressure between CO$_2$ in water and in air) and the greater the CO$_2$ emissions from the sea (Intergovernmental Panel on Climate Change (IPCC), 2001).

The map of Fig. 9 shows the field of SST monthly mean values relevant to the period August–November 1996 and 1997. An extended relative maximum, with values between 22°C and 23°C, is visible in the western Mediterranean Sea, in substantial agreement with the concentration field depicted in Fig. 7. Such agreement was not found for two distinct sinks in the Northern and Central Tyrrhenian Sea (above 39°N).

Instead, the positive anomaly between Czech Republic and Slovakia could be a consequence, maybe, of combustion processes from industrial sources.

As to the simulated sinks located in the alpine region between Italy and Austria and between Italy and Switzerland, a comparison with the 1998 European forestry map, shown in Fig. 10, has revealed the high extent to which these areas and the wider European forestry area overlap each other. So, we can suppose that such sinks, typical of the warm semester, are associated to the CO$_2$ removal processes due to the photosynthetic activity of the local vegetation.

Likewise the previous figures, Fig. 11 depicts the final CO$_2$ concentration field relevant to the cold semester (from November to April) of the years 1996–1997. The broad source area, covering the Westerly Mediterranean Sea and the south-east of the Iberian Peninsula during the warm semester, has now disappeared. Considering that the main causes of the emissions over this area (warm semester) have been identified in the forest fires...
and in the high SST values, both of them being not very active during the cold semester, the disappearance of the positive CO2 anomalies during the cold semester can be seen as an indirect confirmation of our previous hypothesis.

Moreover, the simulation relevant to the cold semester reveals the presence of a greater number of sources, but of smaller extent with respect to those observed in the warm semester. In particular, the major sources are located in the Czech Republic, the Eastern Germany, a region between Netherlands, Belgium and Germany, the Northwest France, the Northern England, and in an area, around 39°N, from Portugal to the western border of the resolved domain, whereas the major sinks are situated in the Tyrrhenian Sea, the Atlantic Ocean between 42°N and 51°N, and in front of the Algerian coasts.

These patterns are in fairly good agreement with the emissions due to the anthropogenic activities and depicted in Fig. 12. These data have been extracted from EDGAR Ver.2.0 database and are relevant to the 1990 (Oliver et al., 1996). The CO2 emission data, on a 1° × 1° regular grid, include many different sectors like fossil-fuel combustion, bio-fuel combustion, industrial processes, land use and waste treatment, and air traffic emissions.

The comparison between the two maps (Figs. 11 and 12) reveals some interesting similarities. In particular, we can observe a significant matching among the source areas located in Eastern Germany, the industrialized region between Netherlands, Belgium and Germany, the Northern England and in the Czech Republic close to Slovakia. A weaker agreement can be noted with regard to the source areas in Portugal and Northwest France, while there is no overlapping regarding the source areas around Paris, Madrid and in the Po Valley (northern Italy).

5. Conclusions

A modified version of the Stohl methodology has been applied to carbon dioxide concentration data measured in three different sites in high mountain areas in Europe, with the aim to locate the most significant source and sink areas of this important greenhouse gas. The investigation has regarded the period of two years 1996 and 1997, and the most important results obtained concern the warm (from May to October) and cold (from November to April) semesters. The CO2 measurement data used in this paper regard the stations of Plateau Rosa and Monte Cimone in Italy, and Wank Peak Zugspitze in Germany. The investigated area stretched from 11°W to 36°E in longitude and from 30°N to 57°N in latitude, and included Western Europe, a part of the Eastern Atlantic Ocean, and the marine coasts of North-West Africa.

The above-mentioned methodology applied in this paper to simulate the CO2 concentration fields has been tested both for the trajectory computations, through an
analysis of a case study relevant to an anomalous event, and for the statistical redistribution of the concentration values over the trajectories. This second test has been performed by forcing six ad hoc sources and six sinks in pre-assigned cells of the integration domain. The positive results of these two tests have confirmed the reliability of the methodology used in this study.

The assumption that the computed CO$_2$ concentration fields can show the source and sink regions of this gas (Stohl et al., 2001) has also been tested and validated. The test has been performed separately for different kinds of supposed emission processes of the CO$_2$.

For the warm semester of the year, when possible sources of CO$_2$ can be represented by biomass combustion processes or mechanisms that regulate the fluxes of this gas at the water–air interface as a function of the surface water temperature, the computed CO$_2$ concentration fields have been compared to the maps of remotely sensed sea-surface temperature values and of the forest fires. Whereas, concerning the sinks areas, that are likely to be connected to summer photosynthetic activity, the comparison has been made with the forestry map of Europe.\footnote{Map published by Ufficio delle pubblicazioni ufficiali delle Comunità europee. Boîte postale L-2985 Luxemburg. Servizio responsabile: Commissione Europea DG VI “Agricoltura”, DG X “Informazione, comunicazione, cultura, audiovisivo”, Rue de la Loi 200, B-1049 Bruxelles. Copyright ©: 1 trimester 1997 European Commission.}

Regarding the cold semester, the computed fields of CO$_2$ have been compared to the EDGAR V.2.0 emission inventory database, so as to check the anthropogenic emissions areas.

The agreement with the simulated concentration fields has been satisfactory, demonstrating how properly the CO$_2$ removal and emission processes, particularly those of biogenic origin or due to forestry fires, show a clear seasonal dependence. Moreover, this study has permitted one to identify with high spatial resolution some source and sink areas, highlighting the great importance of each kind of process linked to emissions or removals of this important greenhouse gas.

Finally, it must be remembered that the methodology applied in this paper is of a statistical type, so that it gives better results and accuracy if used with a great number of receptor sites, uniformly distributed over the computation domain.

As this important condition was fulfilled only to a limited degree, much evident matching has not been found for some sources and sinks.

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References


