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Sensitivity tests on the criterion of potential vorticity index for discriminating the location of ozone sources and sinks over large continental areas

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Summary. — This paper presents the results of a sensitivity analysis of a statistical-dynamic model (ISOGASP, standing for Identification of SOurces of greenhouse GAS Plus), developed by our research group to reconstruct 3D concentration patterns of greenhouse gases in large and deep atmospheric regions over continental or oceanic areas and extending vertically from the lower troposphere to the lower stratosphere. The results of this analysis have shown the ability of the ISOGASP model to discriminate the locations of ozone sources, according to the geographical distribution patterns of atmospheric O_3 concentration inside a limited number of atmospheric layers at different heights above sea level, reconstructed through the method of backward trajectories simulating the travel of air parcels from each different layer to the receptor points at their own height. The potential vorticity index has been used to discriminate the sub-sets of trajectories belonging to stratosphere or troposphere.

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PACS 92.60.Sz – Air quality and air pollution.
PACS 92.70.Gt – Climate dynamics.

1. – Introduction

The Earth's atmosphere is a "unique recipe", a mixture of many different gases in a delicate equilibrium with each other and with the whole Planet; it is the result of millions of years of geological and biological evolution which, in a feedback mechanism, itself influences the habitat of the various life forms on Earth. Geological records have shown that the atmosphere's composition has varied in time, and we know that it is varying today.

What is radically new, at the present time, is that one single biological species, mankind, is helping to cause a rapid—on a geological time scale—and a measurable change of the relative abundances of several greenhouse trace gases which influence the atmosphere's radiative equilibrium. These trace gases make up an extremely small proportion, approximately 0.04% by volume, of the dry atmosphere; this is why even relatively small changes in their absolute quantities will result in noticeable relative variations of their concentrations. Recent studies summarised in IPCC Third assessment report [1] have been evaluating the impact of these changes and have made us aware that changes in the concentrations of these trace gases may markedly affect the variations of the Earth's climates. For this reason it is very important to collect extensive, reliable and accurate data on the concentrations of these trace gases and on their evolution in space and time.

One important and exceedingly active greenhouse gas is ozone, whose absolute concentration, even if much lower than the CO_2 concentration, contributes to some extent to the greenhouse effect.

While it is the greenhouse effect itself which makes planet Earth inhabitable for mankind, an enhancement of this effect could result in noticeable variations of the Earth's climates. Variations and climates have been written intentionally because it may not be very meaningful only to speak of a unique variation of one single Earth's climate: for example we could observe simultaneously quite different regional climate anomalies over the continents of the northern hemisphere and over the oceans in the southern hemisphere.

The anomalies of the surface mean temperature are systematically positive in the last 20 years and their intensity is increasing (`http://lwf.ncdc.noaa.gov/img/climate/research/2000/aug/lo_aug00_pg.gif`). The enhanced greenhouse effect is caused by an increased concentration of the radiatively active greenhouse gases (`http://www.ipcc.ch/present/graphics/2001syr/large/06.01.jpg`).

The radiative forcing due to changes in solar radiation or to changes in the well-mixed greenhouse gases is applied to the atmosphere on a global scale. In contrast the radiative forcing, due to increases in aerosols or greenhouse gases such as tropospheric ozone that are variable in their distribution, varies greatly on the regional scale. In these cases the global average values of radiative forcing provide indications of their individual magnitudes, but they cannot be simply added or subtracted to obtain their total effect on the climate [2].

2. – The MOZAIC program

Ozone data for the boundary layer, collected by instruments installed near the ground, are available today with sufficient coverage in space and time. Only scattered data for the upper troposphere and the lower stratosphere, in the past years, has been available either with remote soundings (*e.g.*, satellite data), or with *in situ* measurements, *e.g.*, by balloon sondes during specific campaigns.

In order to fill this gap, the MOZAIC program (Measurement of OZone and wAter vapour by Airbus In-service aircraft, MOZAIC in the references) has been launched in 1993 with the aim to provide collections of regular and frequent *in situ* data on the upper troposphere and lower stratosphere, resulting in a good coverage in space and time.

It involves a strong cooperation between two different communities:

– European scientists specialized in atmospheric chemistry and dynamics, coordinated by the Laboratoire d'Aérologie/CNRS in Toulouse;

– the European Aeronautics Industry, with the aircraft manufacturer Airbus Industrie and originally four, now three important airlines: Lufthansa, Air France, Austrian

Airlines and Sabena.

Since the beginning of MOZAIC and until March 2003, 18500 flights (*i.e.* 135000 flight hours) were thus made over the continents (Europe, North America, Asia, South America, Africa) and the Atlantic ocean.

MOZAIC consists of automatic and regular observations of ozone and water vapour by five long range passenger airliners flying all over the world. The aim is to create a large database of measurements to validate global chemistry transport models, providing ozone and water vapour climatologies at 9–12 km, which is a very critical domain still imperfectly described in existing models, and requiring improved knowledge about the processes occurring in the upper troposphere/lower stratosphere and the model treatment of near tropopause chemistry and transport.

The present study addresses a sensitivity analysis of a model, developed by our research group, which combines the tool of backward trajectories analysis with a statistical redistribution in space of the observed O_3 concentrations at different points, so as to obtain accurate maps of ozone concentration patterns over large continental and oceanic areas.

Sensitivity analysis of our combined model represents the mandatory first step to assure its ability to identify source and sink regions of ozone not only over extended areas, but also in different vertical atmospheric layers, such as the upper troposphere and lower stratosphere.

3. – Method for the computation of sinks and sources

Sinks and sources are computed with a model which is adapted and modified from the ISOGASP (Identification of SOurces of greenhouse GAS Plus) model, developed from 1997 onwards by the GEOFIT Group of the Dipartimento di Fisica Generale, University of Turin [3-5].

ISOGASP models the source-receptor relationship of a set of backward trajectories associated to a set of receptor points. It is based on a method developed by A. Stohl in 1996 [6]and originally proposed by Seibert *et al.* [7], for the statistical analysis of backward trajectories. Trajectories have been computed with the TRAIETN (TRidimensional Atmospheric Interpolation Evaluation of Trajectories, New version.) model [8], based on a method proposed by Reap in 1972 [9].

First of all, why do we use backward trajectories? With a model of backward trajectories we can reconstruct, backwards in time, the path of an air parcel that has reached our receptor point at a given time and place; the physical phenomenon is the air parcel which follows its forward trajectory and reaches our receptor at the same given time and place; the model of backward trajectories retraces backwards the air parcel's trajectory, and reconstructs its position step by step from the meteorological data provided by the ECMWF with a resolution of $0.5° \times 0.5°$.

Furthermore it is not sufficient to measure the concentration of—in our case—ozone at the receptor points and to calculate the backward trajectories from each receptor point to be able to reconstruct a map of ozone concentration over the region covered by the backward trajectories. It is also necessary to weigh and redistribute the concentrations measured at the receptors over the covered geography according to the specific characteristics of the backward trajectories, *e.g.*, their density over a given area, their residence time in a given grid cell and other meteorological parameters, like the potential vorticity, which are conserved during the motion (along each trajectory) [3-5].

An important parameter calculated by the model is the residence time τ_{mnl} of the l-th trajectory in the grid cell of coordinates (m, n). In fact, air parcels "pick up" concentration differences along their trajectories more efficiently in the regions where they reside longer. For each grid cell the model then calculates the residence coefficient defined as the ratio of the number of trajectory points present in the cell to the number of trajectories crossing the same cell.

For this analysis the values of the geographical and model parameters are the following:

Index	Parameter	From	to	Step
m	lat.	80 N	5 N	1.5 degrees
n	long.	150 E	90 W	1.5 degrees

The receptor points where we measured the ozone concentration and from which we defined the backward trajectories lie along the aircraft's route and were selected for this study at 10-minute intervals. The length of the backward trajectories was chosen at 5 days which was found to be a good balance between good geographical coverage and reliable reconstruction of the trajectory.

4. – Potential vorticity

In principle, the above-mentioned receptor points could be located either in the troposphere or in the stratosphere, depending on whether the aircraft is flying in tropospheric or stratospheric air. This situation requires a criterion to distinguish them effectively.

The criterion we selected to discriminate tropospheric from stratospheric locations of the receptor points is their Ertel potential vorticity in adiabatic, frictionless flow, defined as

$$PV = g(\zeta_\theta + f)\left(-\frac{\partial\theta}{\partial p}\right), \tag{1}$$

where g is the gravity acceleration, f the Coriolis parameter, θ the potential temperature, p the pressure and ζ_θ is the vertical component of relative vorticity on an isentropic—or constant θ—surface. The term $-\partial\theta/\partial p$ tells us how stable the vertical air stratification is [10]. In words: PV is equal to the product of the absolute vorticity on an isoentropic surface by the static stability.

PV is normally positive in the northern hemisphere and its units are: 10^{-6} K $\mathrm{kg}^{-1}\,\mathrm{m}^2\mathrm{s}^{-1}$.

Increasing or decreasing static stability means decreasing or increasing the pressure difference between constant-θ surfaces, therefore shortening or stretching the vortex tubes. On an isentropic surface PV is conserved, so a stretching of the vortex tube means the absolute vorticity must increase, and vice versa.

The typical value of potential vorticity in troposphere is $PV < 2$, in stratosphere $PV > 2$ as in [11]. In our case, the aircraft cruise normally between 9000 m and 12000 m, so the computed trajectories run at the boundary between troposphere and stratosphere. For this reason in the following the computed concentration maps have been marked with UT (Upper Troposphere) when $PV \leq 2$.

5. – The results of ISOGASP sensitivity test

5˙1. *The ozone concentration maps.* – We could identify several recurring features in the July UT ozone maps we reconstructed for the years 1995 to 2002. We show the example of year 1999 because its main patterns are common to the other years (fig. 1a).

- Sources:

  – every year, one or several broad ozone maxima, above 120 ppbv, are located over northern and eastern Europe, and particularly over Siberia;

  – almost every year, a smaller recurring maximum is found over the Atlantic, approximately in the region of the Azores islands;

  – a region of intermediate or intermittent ozone concentrations is observed, linking the Azores maximum with the eastern European/Siberian maximum;

  – an intermittent ozone peak, not present every year, is found over the region around Labrador in north eastern Canada.

- Sinks:

  – We observed lower than average ozone concentration regions in the following locations: a belt of low and very low ozone concentrations, respectively below 50 ppbv and below 30 ppbv, over tropical Africa, the Indian ocean and particularly Indochina; these strong sinks could be explained by three concurring reasons: 1) with the summer monsoon over these regions, which would wash down photochemical ozone precursors and inhibit photochemical activity, 2) with the maritime boundary layer below or near these regions, where O_3 concentrations are low [12] because of photochemical destruction, 3) furthermore at these low latitudes the tropopause rises to over 16–17 km, and we are unlikely to find intrusions of stratospheric ozone at 9–12 km height.

  – a region of intermediate to low concentrations over the North Sea.

5˙2. *Sensitivity tests.* – The above-mentioned recurring features observed in the July UT ozone maps cannot, by themselves, provide reliable unambiguous pictures of the rich variety of atmospheric processes responsible of transport and mixing of the ozone, that only a dynamical approaches can simulate. Nevertheless, as these maps represent the unique reference database against which to compare (and accept or reject) model simulations, a sensitivity analysis of the ISOGASP model, from which they have been obtained, must be performed.

For this purpose we have performed several sensitivity tests on the ISOGASP maps resulting from the receptor points, and their associated backward trajectories, belonging to the UT group of the month of July 1999. The reason for selecting this particular month is that both the number of trajectories and the geographical coverage of the grid area are better than average; furthermore the features of the ozone concentration patterns were well developed and challenging to investigate in greater detail.

Subsets of receptor points and backward trajectories were therefore selected according to the criteria indicated in the following paragraphs, and the resulting ozone concentration maps were compared with each other, in order to investigate possible correlations, *e.g.*, with ozone precursors or synoptic-scale meteorological patterns.

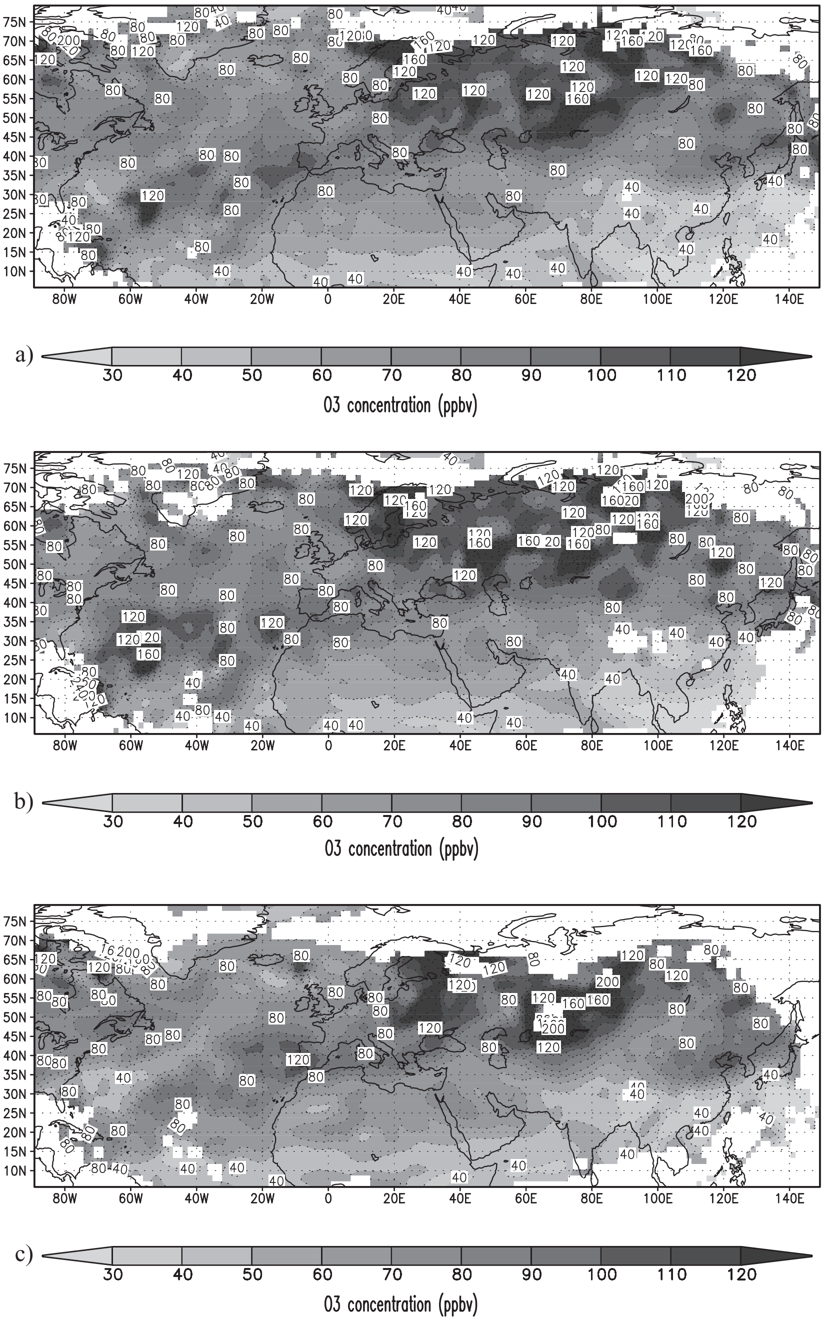


Fig. 1. – a) Concentration map of ozone, 1-30 July 1999, $PV \leq 2.0$, 5897 trajectories; b) concentration map of ozone, 1-15 July 1999, $PV \leq 2.0$, 3218 trajectories; c) concentration map of ozone, 16-30 July 1999, $PV \leq 2.0$, 2679 trajectories.

5˙2.1. Splitting of one month into 15-day periods, $PV \leq 2$. The map for 1-15JUL 99, fig. 1b is quite different from both the 16-30JUL map, fig. 1c and from the 1-30JUL map, fig. 1a where we can recognize an apparent combination of the features of the two 15-day periods; the 1-15 map shows numerous well-developed maxima above 100 ppbv, particularly over the Atlantic at 60W-30N, over Scandinavia, over most of Siberia and at 120E-50N, north of the Korean peninsula. In fig. 1c, some features disappear almost completely, such as the Atlantic maximum at 60W-30N; the maximum previously over Scandinavia appears to have moved south-eastwards, over European Russia, and an extended, strong maximum remains over Siberia, centered approximately at 80E-50N; the maximum previously at 120E-50N, north of the Korean peninsula, appears to have moved to 105E-60N. A striking, small-scale feature present in the 16-30JUL map and completely absent from the 1-15JUL map, is a sharp maximum, concentration over 120 ppbv against a background of 60 ppbv, between Iceland and Scotland; it is a fully enclosed maximum in a well-sampled area, therefore statistically significant. These observations enhance the ability of ISOGASP to highlight the presence of rapidly evolving mechanisms modulating the strength of ozone sources and sinks in summer months.

5˙2.2. Selection of receptors with $PV \leq 1.0$ and $PV \leq 2.0$: a comparison. A second significant sensitivity test was reducing the receptor samples, for the whole month of JUL99, to those with $PV \leq 1.0$ (fully belonging to the troposphere), fig. 2a; the number of trajectories is reduced from 5897 to 4451; this remains an excellent sample. In the statistical analysis of the trajectories [13], it is shown that the mean standard $\overline{\sigma_{PV}}$ deviation of the PV along the UT trajectories is 0.4 PV units. This means that the backward trajectories in this subset, from receptors with $PV \leq 1.0$, are fully tropospheric because only the tail of the distribution of the PV along the trajectory, beyond 2.5 standard deviations, may have sampled air around the tropopause ($PV = 2.0$), with a statistical likelihood of about 1%. We can therefore say that, to 99% confidence level, ISOGASP respects the constraint of PV conservation along synoptic trajectories, providing information on the tropospheric or stratospheric origin of this gas. This also provides, as in the previous example, some evidence about stratospheric, or tropospheric, or near the surface location of sources or sinks of ozone. The comparison between figs. 1a and 2a shows that the amplitude and extension of the maxima in fig. 2a are reduced when compared to fig. 1a.

5˙2.3. Splitting into 15-day periods and selection of $PV \leq 1.0$. This third test is a combination of the previous two: since it appeared that we could resolve the ozone concentration maps into shorter periods, and also into purely tropospheric *vs.* upper tropospheric ozone concentrations, we then divided the receptors with $PV \leq 1.0$ into two sub-subsets: 1-15JUL99, and 16-30JUL99. The number of trajectories per period goes down to 2509 for 1-15JUL and 1982 for 16-30JUL; while these are smaller samples, we'll show at the end of this section that they remain reliable samples.

In fig. 2b the period 1-15JUL is shown; in fig. 2c the period 16-30JUL. We can compare these maps with figs. 1b and 1c, for the same periods and $PV \leq 2.0$.

- period 1-15JUL99. This period 1-15JUL99 shows one of the highest UT ozone concentration peaks of all the periods we have examined in this study: the broad ozone maxima for $PV \leq 2.0$ in fig. 1b become smaller for $PV \leq 1.0$ in fig. 2b; over eastern Siberia, in the area centered around 90E-60N, in fig. 2b the map coverage is absent, indicating that the high concentrations in fig. 2a are sampled in the UT close to the tropopause.

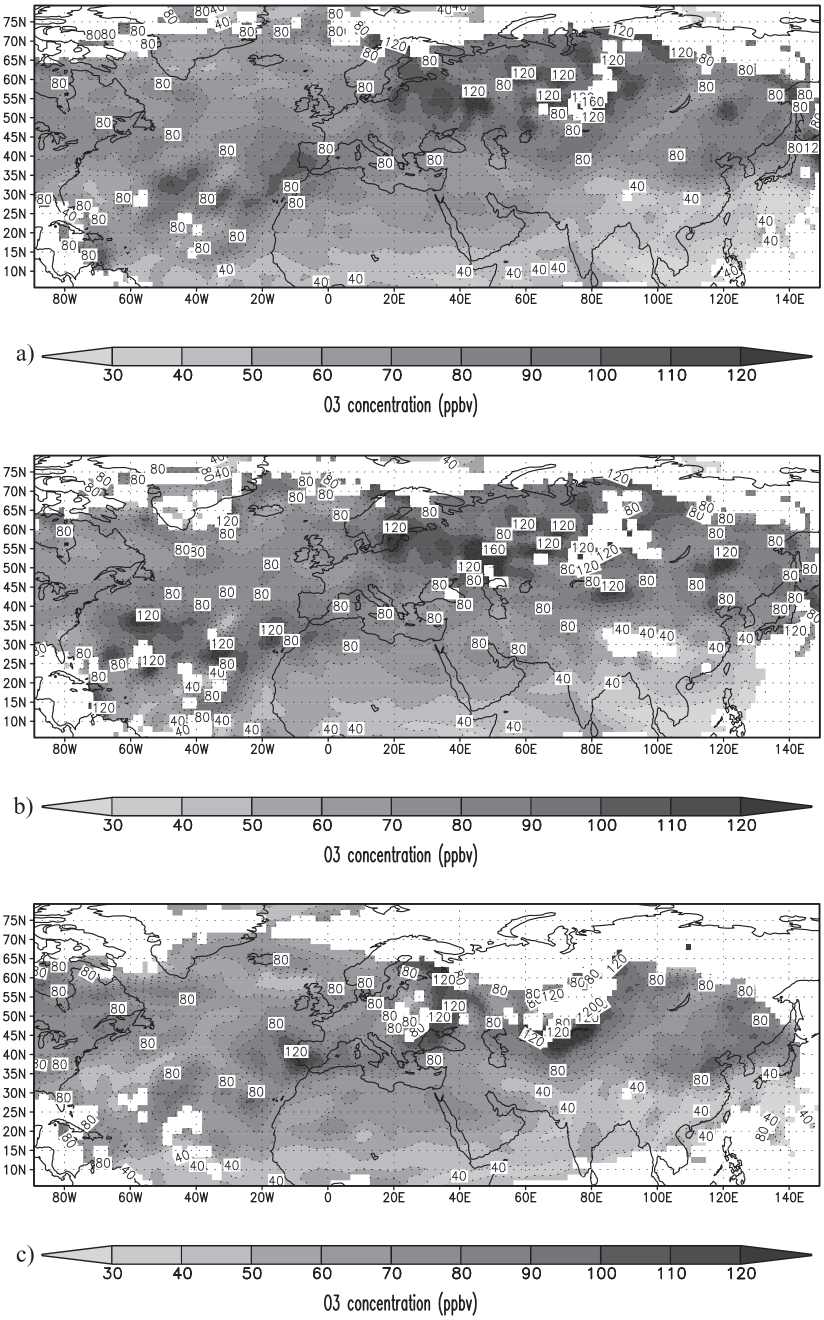
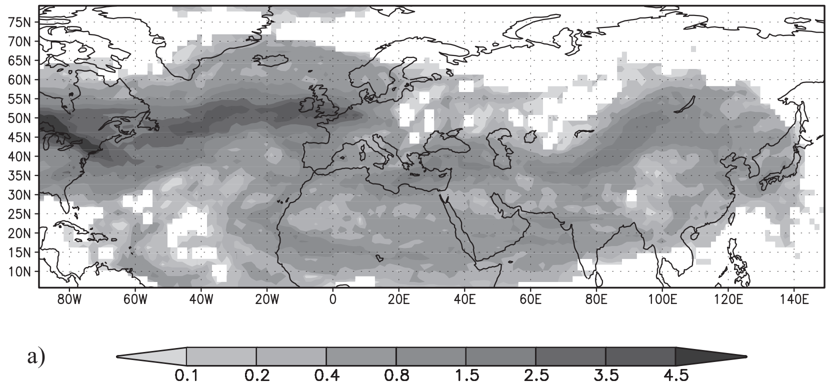


Fig. 2. – a) Concentration map of ozone, 1-30 July 1999, $PV \leq 1.0$, 4451 trajectories; b) concentration map of ozone, 1-15 July 1999, $PV \leq 1.0$, 2509 trajectories; c) concentration map of ozone, 16-30 July 1999, $PV \leq 1.0$, 1982 trajectories.



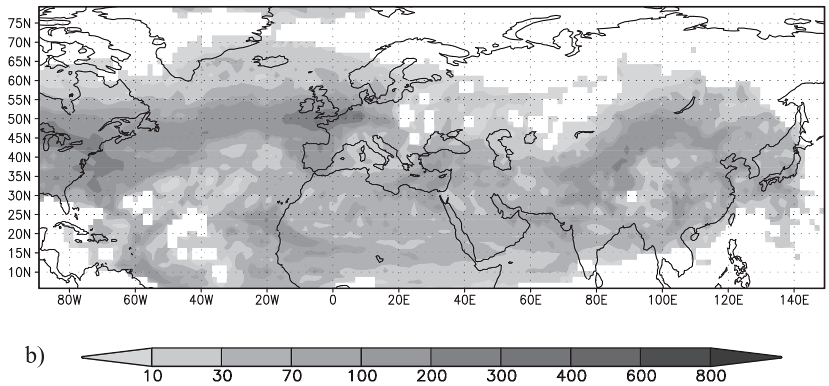
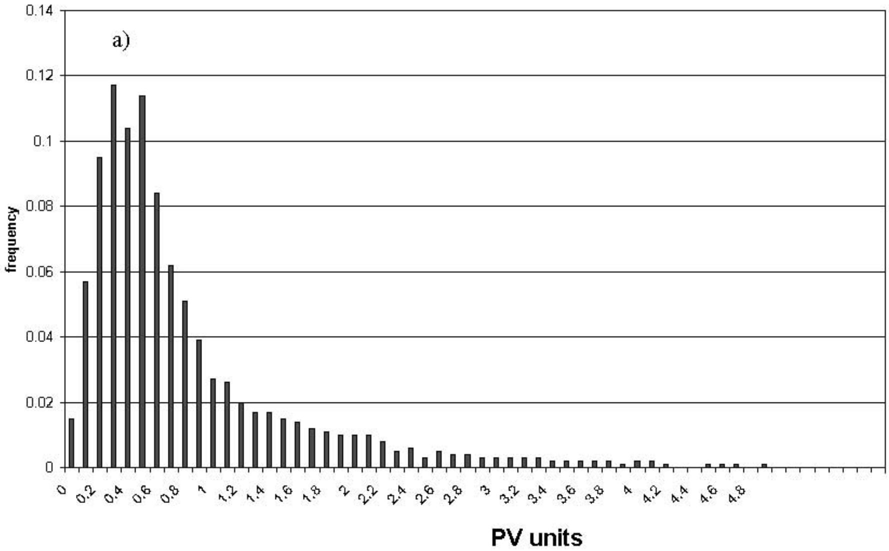


Fig. 3. – a) Distribution of trajectories, 16-30 July 1999, $PV \leq 1.0$, 1982 trajectories; b) number of trajectory points, 16-30 July 1999, $PV \leq 1.0$, 1982 trajectories.

– period 16-30JUL99: here the picture changes once more; we'll describe the main features in fig. 1c for $PV \leq 2.0$, compared to fig. 2c for $PV \leq 1.0$. The broad maximum over European Russia is reduced, but still present; the very large maximum over Siberia between 60E and 90E, 40N and 60N not only is reduced, many grid cell are empty. The small maximum over 120 ppbv at 105E-60N, north of lake Baykal, is reduced to 90 ppbv, while the other small maximum of 100 ppbv at 120E-40N is almost unchanged at about 100 ppbv.

5·2.4. A statistical test. An important additional analysis for the two ozone sources identified above was to check that the statistical trajectory samples and grid cell samples for the ISOGASP model were significant. For example we focus our attention on a source present between Iceland and Scotland and on the source over the Iberian peninsula in the 16-30JUL $PV \leq 1$ concentration map. In fig. 3a we see that the number of trajectories crossing each grid cell between Iceland and Scotland is between 0.4% and 0.8% of 1982, *i.e.* between 8 and 16 different trajectories per grid cell, and in fig. 3b that the number of trajectory points (at 36 min intervals) within each grid cell is between 10 and 30, and one grid cell has over 30 points. The statistical sample is even more significant for



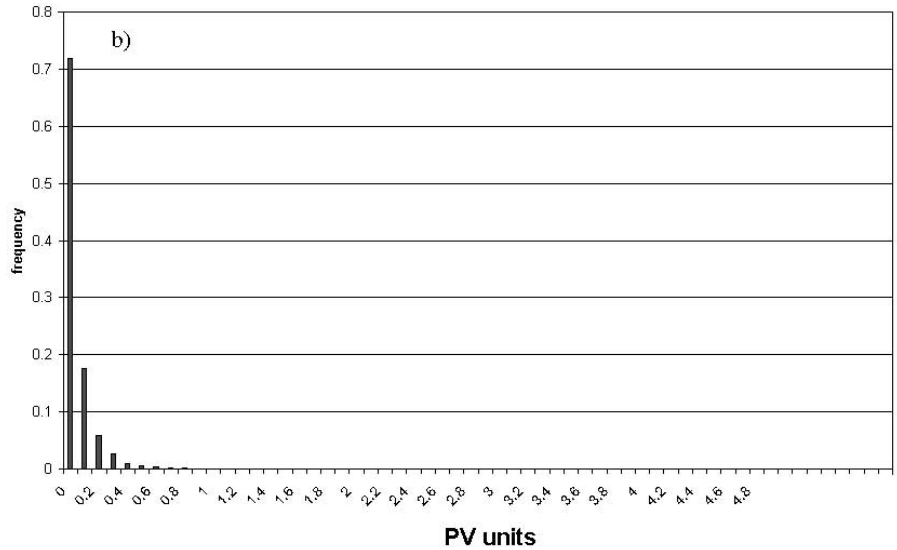


Fig. 4. – a) Distribution of mean PV along the tropospheric trajectory, UT JUL99; b) standard deviation of PV along the tropospheric trajectory, UT JUL99.

the Iberian peninsula: the ozone maximum is defined by between 16 and 30 different trajectories per grid cell, and by between 100 and 200 trajectory points per grid cell.

This statistical check whose results are summarized in fig. 4a (distribution of mean PV for tropospheric trajectories) and fig. 4b (standard deviation of PV along the tropospheric trajectories) ensures the significance of the signal over the possible background noise of single anomalous data fluctuations.

6. – Conclusions

We have presented and discussed a sensitivity analysis of ISOGASP model applied to the research of sources and sinks of tropospheric ozone.

We have used the ozone concentration data collected by the MOZAIC project and we have focused our study to the summer month of July 1995 to July 2002. In particular we have presented in this paper the results of July 1999.

To investigate the model sensitivity we have compared the results of its application in 3 different cases:

1) splitting of one month into 15-day periods, with $PV \leq 2$;

2) splitting of one month into 15-day periods, with $PV \leq 1$;

3) comparison of the selection $PV \leq 2$ with the selection $PV \leq 1$.

In all these study cases the concentrations patterns are different, so we can conclude that this study has demonstrated the sensitivity of ISOGASP in discriminating the geographical location of ozone sources according to the selection of the PV threshold; this result demonstrates the ability of the model to represent adequately processes of dispersion and transportation of ozone in the different geographical regions of our planet.

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REFERENCES

[1] IPCC, Third Assessment Report - Climate Change 2001; `www.ipcc.ch`.
[2] Houghton J., *The Physics of the Atmospheres* (Cambridge University Press) 2002.
[3] Ferrarese S., *Nuovo Cimento C*, **25** (2002) 4.
[4] Ferrarese S., Longhetto A., Cassardo C., Apadula F., Bertoni D., Giraud C. and Gotti A., *Atmos. Environ.*, **36** (2002) 35.
[5] Apadula F., Gotti A., Pigini A., Longhetto A., Rocchetti F., Cassardo C., Ferrarese S. and Forza R., *Atmos. Environ.*, **37** (2003) 27.
[6] Stohl A., *Atmos. Environ.*, **30** (1996) .
[7] Seibert P., Kromp-Kolb H., Baltensperger U., Jost D. T., Schwikowski M., Kasper A. and Puxbaum H., *Trajectory analysis of aerosol measurements at high alpine sites*, in *Transport and Transformation of Pollutants in the Troposphere*, edited by Borrell P. M., Cvita T. and Seiler W. (Academic Publishing, Den Haag) 1994, pp. 689-693.
[8] Anfossi D., Cassardo C., Giarud C., Longhetto A., Villone B., Bacci P., Bonelli P., Carboni G., Bonafè V., Bonasoni P. and Giovanelli G., *Nuovo Cimento C*, **11** (1988) 5.
[9] Reap R.M., *J. Climate Appl. Meteorol.*, **11** (1972) 8.
[10] Pedlosky J., *Geophysical Fluid Dynamics* (Springer Verlag) 1979.

[11] Stohl A., James P., Forster C., Spichtinger N., Marenco A., Thouret V. and Smit H. G. J., *J. Geophys. Res.*, **106** (2001) D21.
[12] Oltmans S. J., and Levy II H., *Atmos. Environ.*, **28** (1994) 1.
[13] Cacòpardo T., Thesis, University of Turin, available upon request at `tommaso.cacopardo@poste.it`