

# A modelling study of an extraordinary night time ozone episode over Madrid domain

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## Abstract

During the early morning hours on April 29, 2000, a time series of ozone observations from several stations showed that a unique and exceptional ozone episode occurred in Madrid city and surrounding areas, whereby monitoring stations reported ozone concentrations up to 1190  $\mu\text{g}/\text{m}^3$ . In order to investigate this phenomenon, two different air quality modeling approaches are used here:

- ✓ The FLEXTRA trajectory model was initially used, suggesting that an intrusion of stratospheric air occurred over the Madrid area and brought stratospheric air down to 1000–2000 m AGL. The local circulation system, not resolved by the FLEXTRA trajectories, subsequently brought some of this stratospheric air to the surface. However, the maximum ozone concentration that could be explained by this process is much less than the observed one.
- ✓ The OPANA Air Quality Modeling System was also employed to study air quality over the Madrid community and city domains. Results suggested that the main wind direction returned to Madrid after 180° wind change direction 1 to 2 h before the “episode”, bringing back ozone generated the day before (typical weekend day with high traffic density). On the other hand, convergence of winds along the South-West North-East axis over the Madrid community showed an important correspondence with the sequence of observations.

Preliminary conclusions show that the exceptional meteorological conditions on such a night could be reason for the occurrence of high values. Additional technical circumstances (such as technical incidences in some monitoring stations) also suggest that some instruments did not work properly under these high concentrations and, thus, real ozone concentrations may have been lower than those measured.

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## 1. Introduction

Ozone is an important secondary pollutant in the atmosphere and also a major oxidant. Ozone episodes are defined as time periods during which concentration

levels within the atmospheric boundary layer and near ground level exceed the air quality limit values that are set via legislation. High ozone concentrations are strongly related to meteorological conditions and usually occur during sunny days, when primary pollutants ( $\text{NO}_x$  and VOCs) interact photochemically, “supported” by strong solar radiation and high temperatures. During the morning hours ozone levels are usually low over large cities, the main reason being the increased

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near-ground emissions which serve as fast-acting ozone sinks. High  $\text{NO}_x$  emissions lead to a sharp rise of  $\text{NO}_2$  concentrations over the centre of the city as a consequence of the oxidation of  $\text{NO}$  to  $\text{NO}_2$  by  $\text{O}_3$ . Higher ozone levels are formed only after  $\text{NO}$  is depleted, e.g. during afternoon hours. Because of the transport and chemical transformation of the primary pollutants in central urban areas, high ozone levels are usually monitored in the suburbs where primary pollutant emissions are lower. For Madrid (Spain), the highly inhabited zone, where pollutant emissions have essentially an urban origin, stays frequently under the influence of high-pressure systems, which strongly influences the ventilation of the area and supports air pollution episodes of certain importance (Pujadas et al., 2000). This leads to ozone episodes which usually occur in the April–October period each year. Exceptional ozone episodes are considered as those occurring at “unexpected” times or seasons of the year with high values that cannot easily be attributed to anthropogenic activities. This is the case for the episode between 2h00 and 6h00 on the 29 April, 2000 over Madrid (Spain) city. Moreover, if the ozone levels reach values of  $1000 \mu\text{g}/\text{m}^3$  or more as was the case over Madrid, the explanation to be found should be somehow “exceptional”, and either deal with the “massive” failure of the monitoring network, or be correlated to rare but possible meteorological conditions. Stratosphere–troposphere exchange can play an important role when explaining these unique episodes (Liu et al., 1987; Stohl and Trickl, 1999).

## 2. The Madrid air quality monitoring network

The Madrid metropolitan area is nearly in the centre of the Iberian Peninsula. It has an average altitude of 650/700 m ASL and is bordered on the North/North-West by the Central System range. These mountains, with maximum heights of 2500 m, are located 60 km away from the city. The surrounding zone presents a smooth slope following a South-West/North-East direction with an inclination of 2% within the city. The whole region is frequently under the synoptic influence of high-pressure systems, which causes poor ventilation and air pollution episodes in winter. The Madrid metropolitan area is one of the most densely populated regions in Spain, with around 5 million inhabitants, including the capital and the surrounding towns. Its industrial activity consists essentially of light industries, and only some medium-sized factories which are located in villages 35 km towards the east and 20 km to the south, and few small chemical industries in the north. The nearest heavy industrial activities are 200/250 km away in Puertollano (Ciudad Real) to the south and in Valladolid to the North-West of Madrid. As a consequence, the Madrid plume is typically urban, which

means that it is fed mostly by traffic emissions and by domestic heating in winter.

Madrid city has a dense Air Pollution Monitoring Network with 24 automatic stations (Fig. 1). In each station the air is sampled at 2.5 m above the surface and the concentrations of  $\text{SO}_2$ ,  $\text{CO}$ ,  $\text{NO}_x$ ,  $\text{O}_3$ , VOCs and particles are continuously monitored. The network was designed for air quality control purposes while also following management criteria, so it covers a rather limited area with an inhomogeneous distribution of the stations in the city. Moreover, the measurements are generally affected from nearby emissions because most stations are located at intersections and busy streets. In addition to this network, Madrid community has nine monitoring stations (Fig. 2). Both networks are operating on a continuous basis and provide information on air pollution concentrations to the local authorities.

## 3. The OPANA model

OPANA is composed of REMEST (non-hydrostatic mesoscale meteorological model, based on the MEMO model; University of Karlsruhe, Germany, 1994) and the SMVGEAR model (University of Los Angeles, USA, 1994; see also San José et al., 1999). There are five different important modules in OPANA: REMO, DEPO, REMEST, EMIMA and CHEMA. REMO, DEPO and REMEST are part of the MEMO adapted code. REMO is a module which provides automatic land use classification based on information provided by the LANDSAT-5 satellite images. This model uses the principal component and the cluster analysis for providing this information which is particularly useful for quantifying the biogenic emissions in EMIMA (San José et al., 1995). EMIMA is a module which provides the biogenic and antropogenic emissions for 250 m grid cells for Madrid mesoscale domain and with a temporal resolution of 1 h. REMEST is the transport model itself which calculates the wind components by solving the Navier–Stokes equations in the atmospheric fluid. REMEST is a transport module which includes a full non-hydrostatic mesoscale meteorological model which provides the three dimensional wind fields and the temperature and specific humidity scalar fields. The transport model provides prognostic and diagnostic information about the three dimensional distribution of the trace gases in an Eulerian context. The model solves the Navier–Stokes equations for the meteorological variables and the Eulerian transport equation for the quantities to be transported by the atmospheric flow (Moussiopoulos, 1984). The adopted temporal discretization uses of the second-order Adams–Bashforth scheme and for the vertical diffusion, the Crank–Nicholson method is applied. The advection term is solved using a modification of the TVD (total variation)

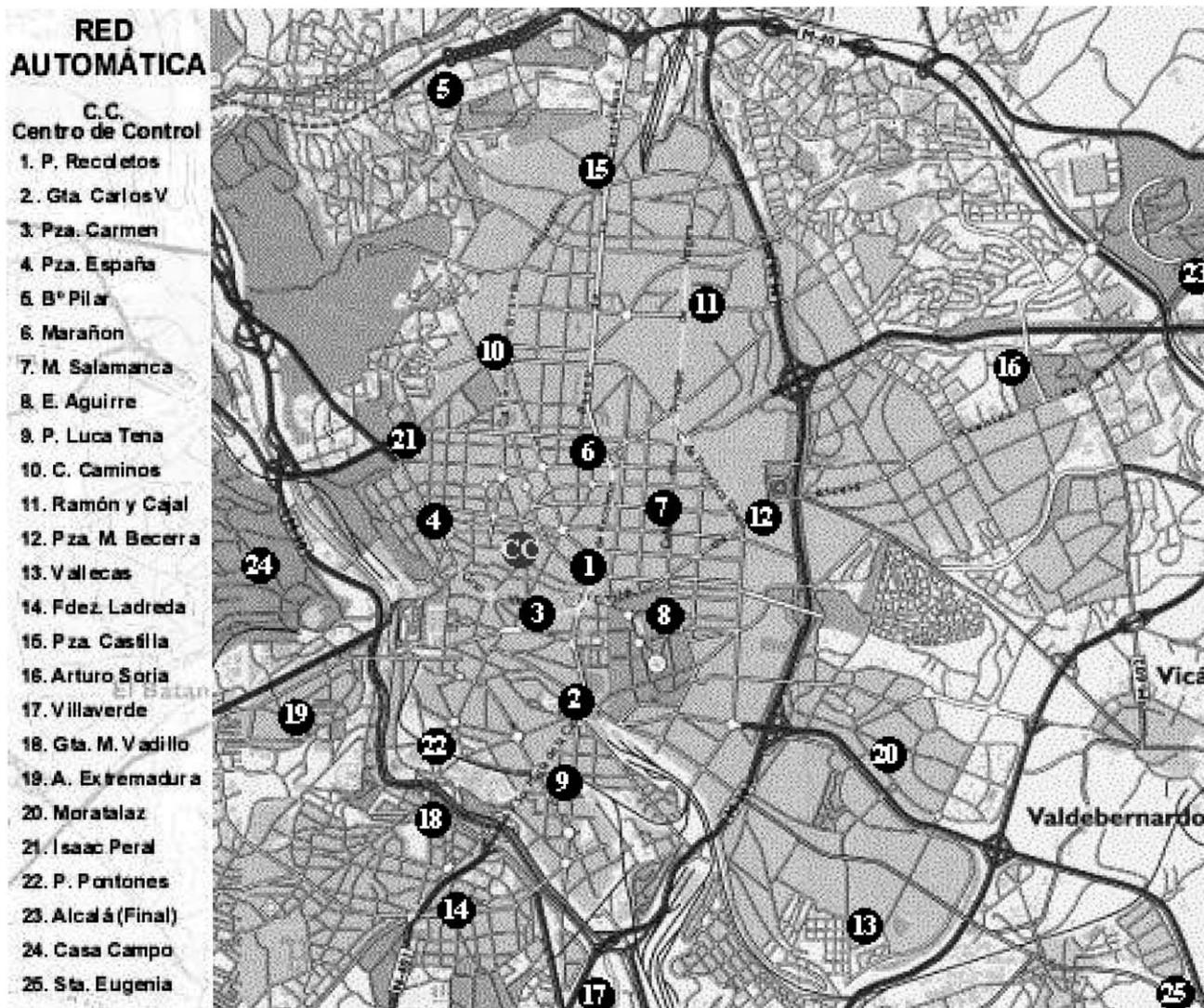


Fig. 1. Madrid city monitoring network.

method introduced by Harten (1986). The CHEMA module includes the chemical transformation of the active trace gases by solving a kinetic differential equation system based on the Carbon Bond Mechanism

for the organic species. A total of 38 species and 78 chemical reactions are incorporated to this module. The numerical method used to solve the system is based on the method proposed by Jacobson and Turco (1994). DEPO model is the deposition parameterization. It uses the resistance approach based on Wesely (1983, 1989).

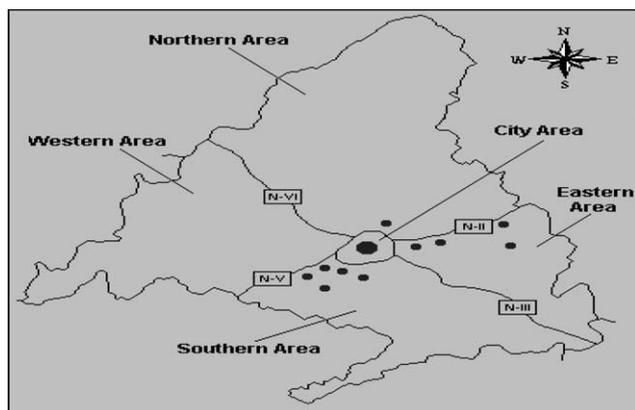


Fig. 2. Madrid community monitoring network.

#### 4. The FLEXTRA model

FLEXTRA [<http://www.forst.tu-muenchen.de/ext/lst/meteo/stohl/flextra.html>] is a Lagrangian model for calculating 3D trajectories, either forwards or backwards in time, based on high-density meteorological data with several user options to change trajectory types and initial configuration (Stohl et al., 1995; Stohl and Seibert, 1998). It is used here with meteorological data from the ECMWF with a horizontal resolution of  $1^\circ \times 1^\circ$  and 60 vertical levels. ECMWF data were

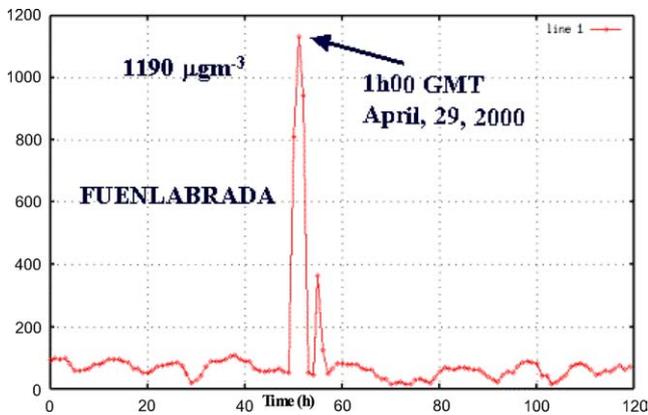


Fig. 3. Ozone concentrations in the Fuenlabrada monitoring station (Madrid community) during the ozone episode. The pattern shows data on April 27, 2000–May 1, 2000.

available every 3 h (analyses at 0, 6, 12, 18 UTC, and 3 h forecasts at 3, 9, 15, 21 UTC). The ECMWF data were interpolated to the trajectory positions using linear interpolation in time and bicubic interpolation in space.

## 5. The Madrid unique episode

In the morning hours of April 29, 2000 the air quality networks of both the Madrid community and Madrid city observed extraordinarily high values of ozone between 1h00 and 6–7h00. Figs. 3 and 4 show these concentrations for two of the Madrid community monitoring stations: Fuenlabrada and Leganés. At Fuenlabrada station (Madrid community monitoring station)—located in the southern part of Madrid—the maximum  $O_3$  concentration value ( $1190 \mu\text{g}/\text{m}^3$ ) was observed at 1h00. Two hours later, the Leganés monitoring station detected an ozone peak reaching  $642 \mu\text{g}/\text{m}^3$ . All 30 stations provide evidence of observations registering somehow the ozone peak with different maxima. In fact five urban monitoring stations (out of 23) registered

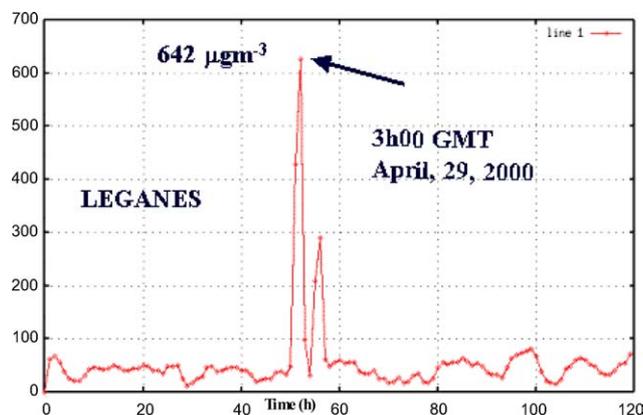


Fig. 4. Ozone concentrations in the Leganés monitoring station (Madrid community) during the ozone episode. The pattern shows data on April 27, 2000–May 1, 2000.

peak values over  $200 \mu\text{g}/\text{m}^3$  and 11 urban monitoring stations registered peak values over  $150 \mu\text{g}/\text{m}^3$ . In any case, all stations registered a relative maximum during the episode. From the community of Madrid monitoring network (7 stations), 5 stations registered peak values over  $500 \mu\text{g}/\text{m}^3$  and the remaining two stations (Móstoles and Torrejón) registered peak values of 230 and  $150 \mu\text{g}/\text{m}^3$ , respectively.

Some meteorological aspects are illustrated in Fig. 5, showing the radio soundings at Barajas airport location at 12h00 and 0h00 on April 28 and 29, 2000. At 0h00 the vertical temperature profile shows very weak inversion profile and in fact it shows a clear neutral turbulent layer which allows vertical exchange with upper layers supporting the findings of previous studies (Sørensen and Nielsen, 2001). Fig. 6 presents the analysis surface map which shows a high pressure over the Madrid area (1016 mb) but a strongly occluded cyclone approaches from the west, which causes a shift in the wind direction during the night which is further confirmed by the mesoscale modelling analysis.

In order to study the air quality of the Greater Madrid area, the OPANA modelling system was used. Simulation results reveal that the wind direction during the hours before the episode (and after the heavy traffic of the Friday evening-week end) was North-East (Fig. 7) while at about 1h00 of April 29, 2000 the wind direction turned by  $180^\circ$  to South-West at about 400 m above ground level (Fig. 8). These circumstances support the suggestion that ozone produced during Friday, 28 April, 2000 was accumulated in the residual layer and returned to Madrid after  $180^\circ$  wind change direction. However, even with such a mechanism ozone concentrations of the magnitude observed during the episode cannot be explained, as the highest ozone concentrations normally produced in European city plumes do not exceed  $400 \mu\text{g}/\text{m}^3$  (San José et al., 1997).

The second tool used is the trajectory model FLEXTRA (Stohl and Seibert, 1998). The model was applied using ECMWF datasets. Seven-day backward trajectories were calculated ending on a regular 3-d grid every  $1^\circ \times 1^\circ \times 1000 \text{ m}$ . Fig. 9 shows only those trajectories that descended by more than 5000 m and, at some point, originated in the stratosphere (potential vorticity greater than 1.6 potential vorticity units) to isolate and emphasize those trajectories that are associated with a stratospheric intrusion. It is evident that the Iberian Peninsula was affected by an air mass that originated in the stratosphere and that the deepest descent occurred just over the Madrid region. It thus seems possible that high concentrations of ozone were transported from the stratosphere to the area of Madrid. However, these trajectories did not reach the surface, but descended down only to 1000–2000 m above ground level. The presence of the stratospheric air is also seen by a layer of dry air at about 750 h Pa in the 0 UTC radiosounding

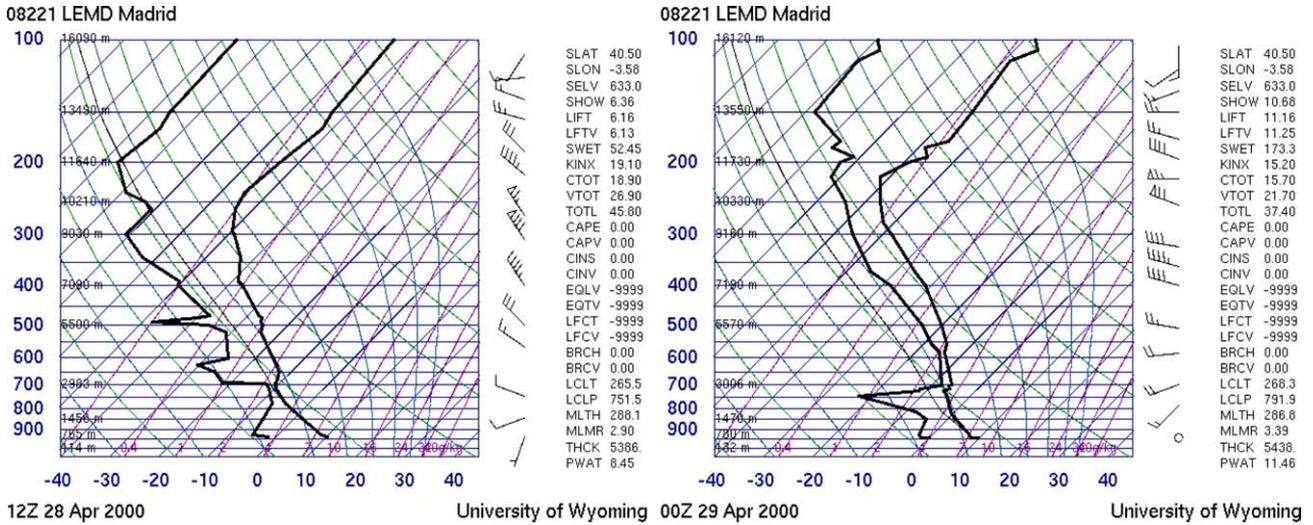


Fig. 5. Skew-T diagrams from radiosounding data from Barajas International Airport, Madrid (<http://weather.uwo.edu/upperair/sounding.html>).

(Fig. 5). Once the ozone is in the upper layers of the Madrid nocturnal boundary layer at approximately the time of the ozone episode in discussion, stability is very weak and thus turbulence may prevail throughout the night. This suggestion is supported by the REMEST (San José et al., 1997) model analysis, that shows Monin-Obukhov lengths at surface level close to the ones corresponding to neutral conditions.

### 6. Conclusions

During the early morning hours on April 29, 2000, observation data showed that a unique and exceptional ozone episode occurred in Madrid city and the surrounding area, where monitoring stations reported ozone concentration levels up to  $1190 \mu\text{g}/\text{m}^3$ . In this contribution simulation results are presented based on

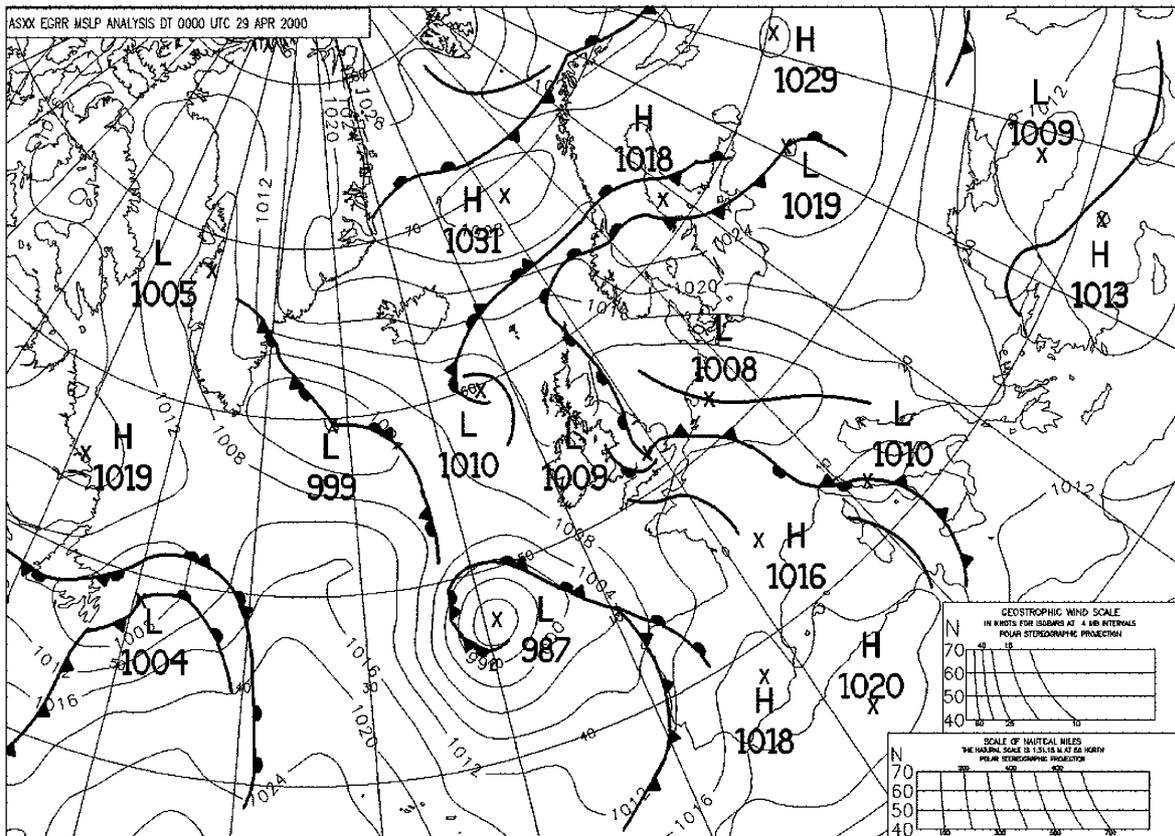


Fig. 6. Surface analysis map over Europe.

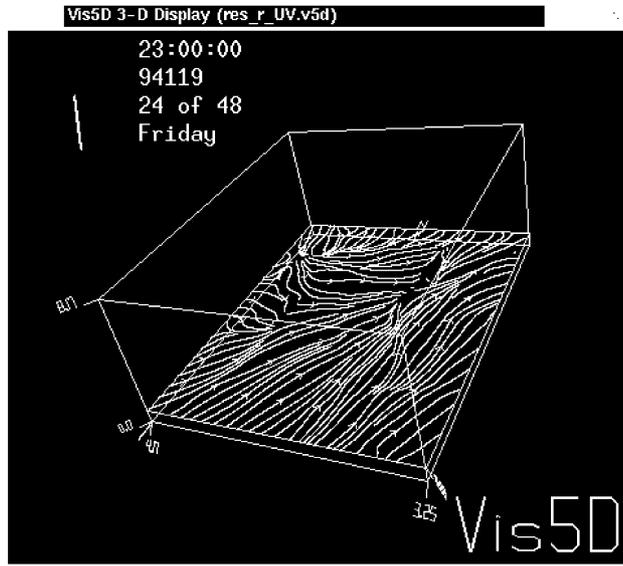


Fig. 7. Three dimensional visualization showing 400 m wind patterns on Madrid domain (OPANA domain) at 0h00 GMT, April 29, 2000. The wind direction is clearly oriented to the North-East.

two different modeling approaches. The first one is a trajectory analysis with the FLEXTRA trajectory model, and the second one is the application of the OPANA Air Quality Modeling System over the Madrid community and city domains. Trajectory analysis results support ozone intrusion from elevated layers that might have brought significant amounts of ozone from the stratosphere to about 1000–2000 m above the ground during April 28 and 29, 2000 over the Madrid domain. On the other hand, OPANA results regarding upper layer wind trajectories on late hours on April 28, 2000

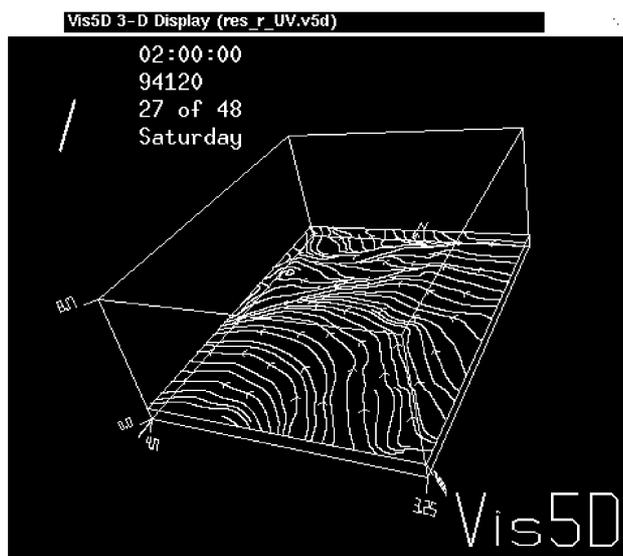


Fig. 8. Three dimensional visualization showing 400 m wind patterns on Madrid domain (OPANA domain) at 2h00 GMT, April 29, 2000. The wind direction is clearly turning back to South West.

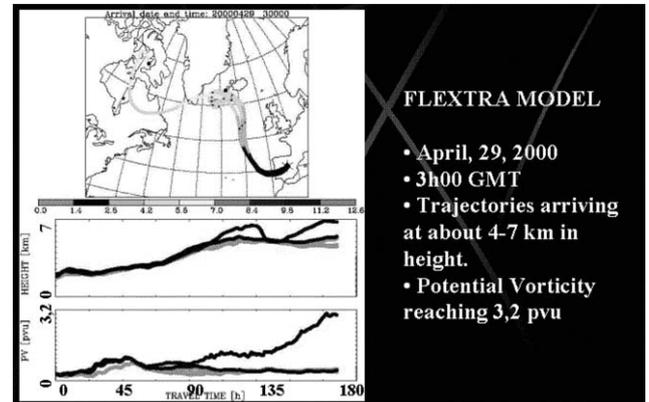


Fig. 9. Ensemble of three-dimensional trajectories depicting the intrusion of stratospheric air into the troposphere. Seven-day trajectories were started from a  $1^\circ \times 1^\circ \times 1000$  m three dimensional grid covering a domain larger than the Iberian Peninsula. Plotted here are only those trajectories that descend by more than 5000 m from the stratosphere ( $PV > 1.6$  pvu).

(Friday evening) over Madrid domain, suggest that the main wind direction, being South West, turned  $180^\circ$  1–2 h before the episode, bringing back the ozone generated on the day before (typical weekend day, high traffic rates, etc.). On the other hand, convergence of winds along the South-West North-East axis over the Madrid community shows an important correspondence with the sequence of air monitoring stations observing the high exceptional ozone values. Preliminary conclusions are that the exceptional meteorological conditions during this night were the reason for the occurrence of such high values. However, the sheer magnitude of the observed ozone values makes it difficult to explain them by any known mechanism, be it accumulation of photochemical pollution or the occurrence of a stratospheric intrusion. Technical problems (such as technical incidences in some monitoring stations) could have caused some instruments to not work properly under such high concentrations. Thus, while the measurements clearly suggest very high ozone levels, the exact magnitude of these levels may be highly uncertain.

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