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A winter ozone episode over Portugal

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1 Introduction

It is well known that air masses cross the stratospheric-tropospheric "boundary". Some of the processes responsible for this fact are well established although the mixing process that perturbs chemical species concentrations in the troposphere is not well understood (Johonson and Viezee, 1981; Vaughan *et al.*, 2001). Ozone is one of the species specially important and several studies have been carried out in order to determine the influence of stratospheric ozone in the tropospheric ozone budget. However, the quantification of the chemical fluxes between these two layers is rather difficult to settle (Monks, 2000 and references in it).

Holton et al (1995) pointed out the importance of considering all the scales involved in the Stratosphere-Troposphere exchange, namely in the Northern Hemisphere Winter. This fact is also confirmed by Elbern *et al* (1998) that also estimated an occurrence of 11 000 days of folds each year, which is quit impressive, considering that folds are the most important processes by which the stratospheric and tropospheric air masses are mixed.

Stratospheric air masses are characterized by high ozone concentration levels, low moisture content, high levels of ^{7}Be, ^{10}Be and potential vorticity (Hov *et al.*, 1998). Based on these facts it is possible to relate surface measurements with stratospheric intrusions. Several studies indicate that mountain monitoring stations have higher probability of registering intrusion events compared with lower altitude monitoring stations

(Zanis *et al.*, 2003). Although deep folds are exceptional events they can be measured sometimes.

The work presented in this abstract has the perspective of justifying the ozone values measured at an air quality station sited in the downtown of Coimbra, a city in the centre part of Portugal, as a result of strong tropopause activity between the 5^{th} and 17^{th} of January 1996.

2 Methodology

The methodology applied concerning detection of possible surface ozone with stratospheric origin included the application of several criteria to the ozone measurements at all air quality stations (belonging to the Environment Institute) between 1991 and 2001. First of all, a WMO data quality assessment (WMO, 1994) was applied to all ozone data series. A set of 165 ozone episodes were selected after the accomplishment of one of the following criteria:

- date of occurrence, namely ozone episodes occurring between November and February;
- hour of occurrence, episodes measured during the night period, considered between 21H00 and 7H00;
- number of stations in which the ozone episodes are measured simultaneously (three or more stations);
- number of regions with simultaneously occurrence of ozone episodes.

For practical purposes, an ozone episode is considered when the alert threshold of 180 μg.m^{-3} was surpassed. Nevertheless, concentrations above 100 μg.m^{-3} were also analysed (Barros e Fontes, 2003).

For a subjective analysis of the synoptical conditions this number of ozone episodes is difficult to handle. In order to diminish it, a new set of criteria was considered. Based on the ozone episodes list, meteorological episodes were established. Ozone episodes occurring in consecutive days or with one-day interval are considered to be included into the same meteorological episode.

A meteorological episode is defined starting 8 days before the ozone episode (or the first day if more than a day of exceedance occurs), including the ozone episode day (or the period between the first and the last day of consecutive days of exceedances) and adding two more days after the ozone episode. Concerning this definition the number of episodes was reduced to 100.

Furthermore, choosing meteorological episodes corresponding to an isolated ozone episode, with no overlapping with the following meteorological episode, and not occurring in summer reduced considerably the meteorological episodes to be analysed in an subjective way (Leitão e Moreira, 2004). At the end, six meteorological episodes were considered for a detailed analysis and modelling for the period 1996-2001.

The meteorological model applied in this study was the Penn State Mesoscale Model MM5 (Dudhia, 1993) with ECMWF ERA 40 database as global meteorological fields each 6 hours. The model was applied over a coarser domain and two nested domains, the inner most covering Portugal with a spatial resolution of 10 km x 10 km.

3 Case Study

The meteorological and ozone episode occurred at Coimbra on 17^{th} January 1996 at 14 UTC where 182 μg.m^{-3} were measured in the monitoring station. The simultaneous analysis of back-trajectories and dynamic tropopause (defined using the 1.5 PVU reference value) showed that in some situations the meteorological episode should be redefined, and in this case it is considered to start on the 5^{th} January.

Back-trajectories that arrive at Coimbra at low levels reach the upper troposphere close to tropopause anomalies. It is possible to consider important stratospheric intrusions in 3 key moments: i) 7^{th} January between 00 UTC and 12 UTC over North America; ii) between 8^{th} January at 12 UTC and 9^{th} January at 12UTC in North Atlantic off the coast of Newfoundland; iii) 11^{th} January between 12 and 18 UTC. During the 5 days before the ozone episode the back-trajectories lie over the Iberian Peninsula in low-pressure levels. On the other hand, back-trajectories arriving at Coimbra in high levels come from the Mediterranean region.

As it was mentioned before this meteorological event was simulated with MM5 and a high dynamic activity can be observed over the three simulated domains. This period was characterised by a strong tropopause activity and cyclogenetic activity in the North Atlantic, with frontal activity over Portugal between 5^{th} and 12^{th} January. An intense and persistent folding, lasting for more than 15 hours, creates a pool of stratospheric air at an altitude of around 3500 m over the Atlantic. These results confirm the tropopause anomaly previously detected and pointed out as one the moments where stratospheric air could have entered into the free troposphere.

Although the exceedance has been measured on the 17^{th} January, surface ozone values in Coimbra are considered high enough for this type of station and at this time of the year. Between 4^{th} and 21^{st} of January ozone concentrations over 100 μg.m^{-3} are measured at Coimbra. This air quality station is classified as urban, and in fact is measuring ozone in a busy downtown avenue. During 11 years the highest value for the median of hourly ozone concentration was 24 μg.m^{-3} calculated with ozone data corresponding to the year 2000. For the same period, a rural station over Portugal presents the highest median value of 94 μg.m^{-3} in 1996. Ozone concentrations over 100 μg.m^{-3} were also measured in other two stations in the Southwest part of Portugal in this period.

4 Conclusions

This work shows that the methodology applied to identify possible influences of stratospheric ozone on surface measurements of this chemical specie is suitable.

Based on chemical and dynamic features there are strong arguments to sustain that stratospheric ozone influenced the air quality over Portugal in January 1996. It may be speculated that different stratospheric air masses mixed with boundary layer air mass in different moments, thus possibly being present in Coimbra for more than 15 days.

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