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On the origin of elevated surface ozone concentrations at Izana Observatory, Tenerife during late March 1996.

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Abstract: The origin of relatively high surface ozone concentrations measured at Izana Observatory (Canary Islands) during the end of March 1996 is studied using a coupled chemistry-GCM (ECHAM4) at T63 resolution (1.875° x 1.875°). Meteorological fields (geopotential height, potential vorticity, specific humidity), and a model-simulated stratospheric ozone tracer as well as 3-D back trajectories, show the stratospheric origin of these relatively high surface ozone values caused by cross-tropopause exchange at the western flank of an upper level trough/cut-off low (COL) over the extratropical North–Atlantic Ocean. The good agreement between observations and model results (within 10-15%) indicates that the high resolution chemistry-GCM is a useful tool towards the understanding of natural sources controlling background surface ozone variability. The results underscore the importance of stratosphere-troposphere exchange (STE) during late winter/early spring for lower free tropospheric ozone at subtropical latitudes.

Introduction

The relative contributions of stratospheric and in situ photochemically-produced ozone in the troposphere has been the subject of scientific inquiry and debate for the past three decades (e.g. Chameides and Walker 1973; Fishman et al. 1979, 1985; Logan 1985; Liu et al 1987; Roelofs and Lelieveld 1997; Levy et al. 1997). Of particular interest is the cause of the spring maximum observed in tropospheric ozone concentrations at a number of remote sites in the northern hemisphere (e.g. Oltmans et al., 1992; 1996a; Derwent et al., 1997). STE is considered to be an important source of tropospheric ozone especially in the northern hemisphere (NH) mid-latitudes during spring and may be most clearly discerned in the middle/upper troposphere, where short-term variations occur less frequently than in the lower troposphere. However, a limited number of studies showed that air of stratospheric origin can occasionally penetrate into the lower troposphere [e.g. Davies and Schuepbach, 1994; Kunz et al., 1997; Eisele et al., 1999] producing transient high ozone concentrations at the earth's surface particularly in late winter/spring. The present study examines the origin of relatively high ozone concentrations (~ 30% higher than the monthly mean value) measured at a high-elevation remote site in the eastern subtropical Atlantic [Izana Observatory, Canary Islands (28.2°N, 16.3°W); 2.3 km altitude] during the period 23-27 March 1996, by using a high resolution coupled chemistry-GCM. We evaluate the ability of the model to reproduce the observed short-term ozone variability and examine the origin of these elevated ozone mixing ratios by 'decomposing' the modelled ozone daily variability into the contributions from two different source categories: ozone originating from the stratosphere and ozone produced within the troposphere. Hence, our goal is to obtain a quantitative understanding of the sources controlling background surface ozone. A recent study by Oltmans et al. (1996b), based on a number of O3-sonde profiles, underlined the importance of the stratospheric source to elevated ozone concentrations in the middle troposphere over the Atlantic Ocean during spring/summer, while Prospero et al. (1995) found good correlation between ozone and ^{7}Be in the free troposphere over Tenerife.

The general circulation model used in the present study is the European Centre Hamburg Model, version 4 (ECHAM4), with 19 vertical layers in a hybrid sigma-p co-ordinate system from the surface up to 10 hPa, and a horizontal resolution of 1.875° x 1.875° (T63). Tracer transport is calculated using a semi-Langrangian advection scheme (Feichter et. al., 1996), while additional vertical transports are included through parameterizations for vertical diffusion and convection (Roeckner et al., 1992; Tiedtke, 1989). The GCM is coupled to a tropospheric chemistry model that considers background CH4-CO-NOx-HOx chemistry, emissions of NO and CO, dry deposition of O3, NO2, HNO3 and H2O2 and wet deposition of HNO3 and H2O2 (Roelofs and Lelieveld 1995; 1997). Stratospheric ozone concentrations are prescribed using the results of a 2-D troposphere-stratosphere chemical model (Bruehl and Crutzen 1988). ECHAM4 considers a tracer (O3S) for ozone that originates from the stratosphere (the reactions involving O3S in the troposphere are only those that destroy ozone). The difference between calculated concentrations of O3 and O3S (hereafter referred to as O3t) represent ozone that is photochemically produced/destroyed within the troposphere due to natural and anthropogenic precursor emissions. In order to represent the actual meteorological conditions during early spring 1996, we applied a 4D assimilation technique, i.e. Newtonian relaxation, by which the model variables representing vorticity, divergence, temperature and surface pressure have been nudged towards ECMWF analysis (a detailed description/evaluation of the technique can be found in Jeuken et al., (1996)). The aforementioned technique has been previously used in ECHAM4 (at T30 and T63 resolutions) to simulate synoptic patterns associated with (real time) STE events, for which good

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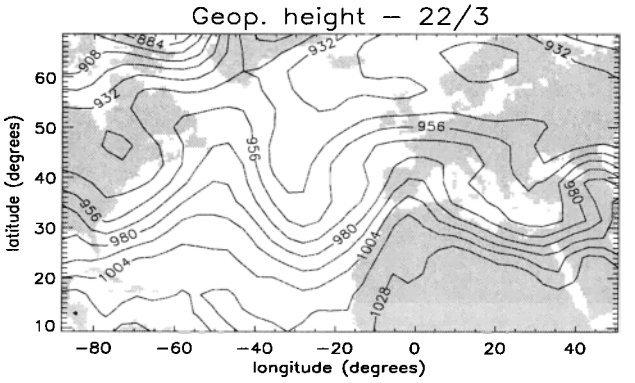

Figure 1. Modelled geopotential heights (values in dm) at model level 10 (~320 hPa) for the 22nd of March 1996 (12:00 UTC).

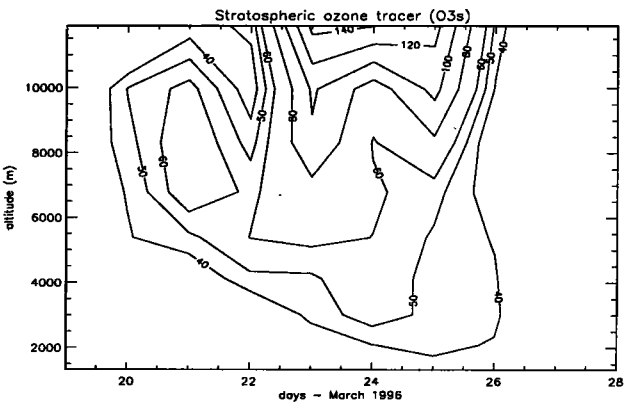

Figure 3. Vertical distribution of O3S concentrations (ppbv) for the grid-box that encompasses Canary Islands and for the period 20-28 March 1996.

agreement between model results and ECMWF analyses was found, especially at T63 resolution (Kentarchos et al., 1999; 2000).

Results - Discussion

The synoptic situation over the extratropical N-Atlantic during the second half of March 1996, was dominated by upper-level pressure anomalies and intense surface fronts. In particular, examination of the sequence of upper level geopotential heights revealed the presence of a developing trough over the N-Atlantic on the 20th of March, which deepened and moved southwards during the following two days (*Figure 1*). On the 23rd, a cut-off region of low geopotential height values formed to the north-west of the Canary Islands, and moved eastwards during the following two days after it started to decay. The corresponding surface charts showed a vigorous cold front (associated with intense cyclogenesis) passing over the Canary Islands (after the 23rd) followed by a high pressure system. The combination of such synoptic conditions has been associated with strong downward transport of air masses from the upper troposphere/tropopause region into the lower troposphere (Davies and Schuepbach 1994). The corresponding potential vorticity (PV) distributions in the upper troposphere are suggestive of the advection of stratospheric air associated with the upper level trough. (Hoskins et al., 1985). *Figure 2* shows the southward excursion of high-PV air masses (> 2 pvu) (1pvu= 10^{-6} K m^2/kg s) over the N-Atlantic during the 22nd of March. According to WMO (1986), PV values > 1.6 pvu are indicative of the stratospheric origin of air masses. Furthermore, inspection of the sequence of modelled specific humidity fields in the upper troposphere (not shown here) revealed the presence of dry air masses of stratospheric origin (specific humidity values < 0.15 g/kg) over the Atlantic in association with the upper level synoptic anomalies. These dry air masses extended deep into the troposphere, down to ~ 600 hPa. Nevertheless, specific humidity values gradually increased further down in the troposphere due to mixing with the surrounding -more humid- tropospheric air.

One of the most important aspects of STE is the transport of ozone between the stratosphere and troposphere. Examination of the distribution of modelled O3S mixing ratios in the upper troposphere during the period 22-25 March 1996, showed an isolated area of high O3S values (>160 ppbv) in the vicinity of the upper level trough/COL. Time-height cross sections of O3S mixing ratios provide useful information on the vertical extent of these O3S-rich air masses. *Figure 3* illustrates the time sequence of such O3S cross sections for the model grid-box that encompasses Izana. A tongue-like feature of elevated O3S concentrations is present, descending down to ~ 2 km

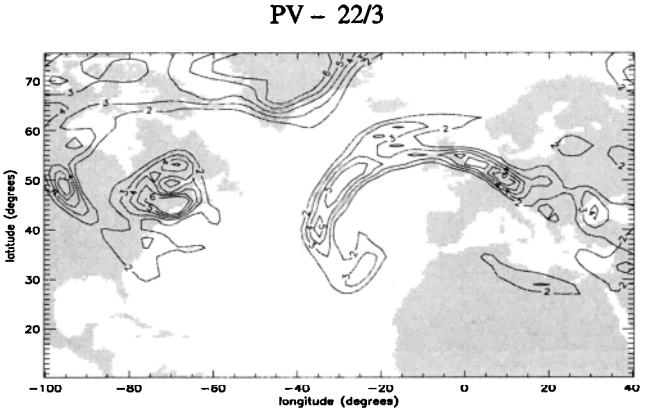

Figure 2. Simulated potential vorticity distribution at model level 10 (~320 hPa) for the 22nd of March 1996 (12:00 UTC). Values are in pvu units.

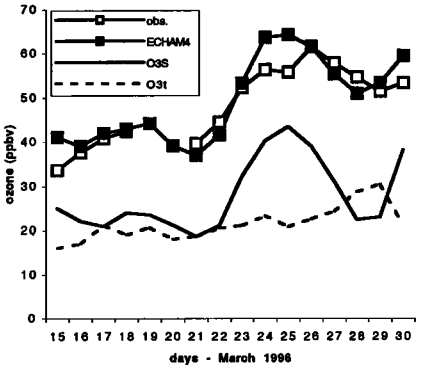

Figure 4. Observed and modelled surface ozone concentrations at Izana Observatory during the period 15-30 March 1996 (the O3S and O3t components of the modelled O3 concentrations are also shown for comparison).

between the 24th and 26th (in association with the passage of a cold front over Tenerife during that period). Although the stratospheric component of ozone decreases with altitude into the troposphere (due to mixing with the surrounding tropospheric air and - to a lesser extent - chemical destruction), the vertical extent of the 40 ppbv isoline is suggestive of the stratospheric influence at the altitude of Izana Observatory (~ 2.3 km). *Figure 4* shows the (daily averaged) surface ozone concentrations measured at Izana Observatory during the second half of March 1996. The observatory is located above the atmospheric boundary layer and is shielded from local anthropogenic emissions by the strong trade wind inversion (Cuevas 1995; Schmitt et al., 1997) and thus can be regarded as representative of free tropospheric conditions. ECHAM4-calculated O3, O3S and O3t mixing ratios (interpolated to the altitude of the Izana Observatory) are also shown for comparison. The model reproduces well the pronounced increase in surface ozone concentrations observed after the 22nd, although minor discrepancies in the actual magnitude and location of the O3 maximum remain. The temporal evolution of the stratospheric ozone tracer (O3S mixing ratios of ~ 40 ppbv occur during the period 23-26 March) is strongly suggestive of the stratospheric origin of the elevated surface ozone mixing ratios. On the other hand, the tropospheric ozone component shows no significant variation during that period and O3t values are relatively low (~ 20 ppbv). It is interesting to note that modelled CO concentrations for March 1996 showed a minimum (~ 40% lower than the average calculated monthly mean value) between the 21st and 25th, which is also indicative of the stratospheric origin of the high ozone episode. Unfortunately, there are no CO measurements available at Izana during that period to support the model results. Additional information about the origin of the air masses arriving at Izana Observatory can be obtained from 3-D back trajectories. In the present study we used the FLEXTRA trajectory model (Stohl et al. 1998), driven by the 3-D wind fields calculated by ECHAM4. We applied a linear temporal interpolation, while the spatial interpolation is bicubic in the horizontal and polynomial in the vertical. *Figure 5* shows a cluster of 3-day back-trajectories initialized on March 25 (12:00 UTC) at 750 hPa (~ 2.3 km altitude) from the longitude of Izana Observatory (16.3° W) and latitudes ranging from 27°N to 32°N at 1° intervals. The air masses arriving at Izana

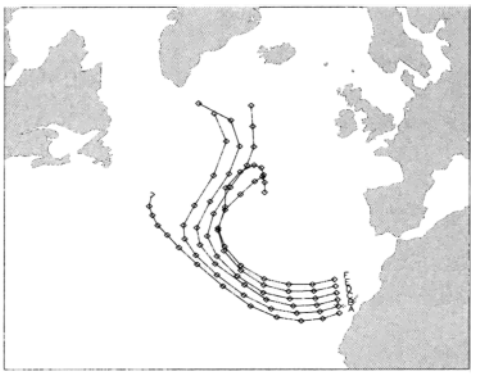

Figure 5. 3-day back trajectories (6-hour intervals) started on March 25 (12:00 UTC) at one degree latitude intervals from 27°N to 32°N and a fixed longitude of 16.3°W (longitude of Izana Observatory). The starting altitude is 750 hPa (approximate pressure level of Izana Observatory)

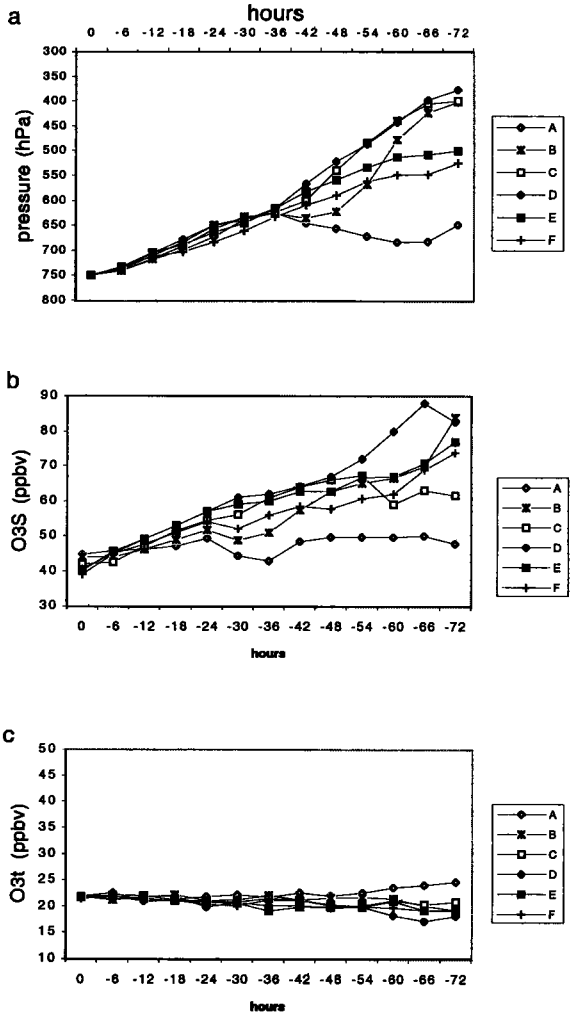

Figure 6. Time series of pressure *(a)*, O3S *(b)*, and O3t *(c)* for each one of the 3-day back trajectories shown in *figure 5*. The starting altitude is 750 hPa (approximate pressure level of Izana Observatory) and time 0 (starting point) corresponds to 25 March (12:00 UTC).

Observatory on the 25th, originated from the north-Atlantic upper troposphere (*Figure 6a*) near the region of the upper level trough/COL and then moved southwards, curving cyclonicaly as they descended into the lower troposphere. Time-series of the corresponding O3s and O3t values along these trajectories illustrate the stratospheric influence on ozone in these air masses (*Figure 6b,c*).

It should be mentioned that these results do not depreciate the role of photochemical ozone production due to natural and anthropogenic emissions. In fact, preliminary results from a 1-year ECHAM4 simulation ('nudged' version – manuscript in preparation) suggest that over the subtropical-Atlantic free troposphere (700-400 hPa) the stratospheric and tropospheric ozone components are of comparable magnitude during spring/early summer; however, the tropospheric ozone component increases considerably in the summer season (between July and September) as a result of intense photochemical activity. Nevertheless, as the present study indicates, STE events of episodic nature can inject stratospheric air into the troposphere which can dominate the tropospheric

ozone budget regionally and over short time-periods (few days). Furthermore, given the relatively high frequency of upper level trough/COL formation over the extratropical N-Atlantic (Kentarchos and Davies, 1998; Elbern et al, 1998) and the high frequency of N-Atlantic-originated air masses reaching Izana Observatory (Cuevas, 1995), the accumulated contribution of stratospheric ozone to the surface ozone budget at Izana Observatory due to STE processes may be of significant importance over longer time-scales.

Conclusions

We have investigated the origin of relatively high surface ozone concentrations measured at Izana Observatory during the period 23-27 March, 1996, by using a coupled chemistry-GCM (ECHAM4). The evidence given by calculated geopotential heights, potential vorticity and specific humidity fields, as well as a stratosphere-originated ozone tracer complemented by 3-D back trajectories, strongly implicate the upper troposphere and stratosphere as the source of these elevated ozone concentrations in association with an intense upper level trough/COL over the extratropical Atlantic Ocean. Furthermore, the passage of a surface cold front over Izana followed by a rapidly emerging high pressure ridge have caused further descent of the stratospheric ozone-rich air into the lower troposphere. The present case study illustrates the capability of the high-resolution chemistry-GCM (ECHAM4) to capture the origin and magnitude of short-term surface ozone variability in the lower free troposphere.

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