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Sudden increases in the NO_2 column caused by thunderstorms: a case study in the northern subtropical region

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Abstract

A long-term program for NO_2 column measurements started in 1993 at the subtropical Izaña Observatory (28° N, 16° W). Seasonal evolution shows a small day-to-day variability as compared with higher latitudes. Sharp increases in the column appear occasionally superimposed on the annual cycle. The origin of these spikes is explored by considering the possibility of tropospheric transport from polluted areas, stratospheric intrusions, meridional transport in the stratosphere and production by lightning, in a case study. From radiative transfer calculations and meteorological information available, it is shown that the NO_2 increase takes place in the upper troposphere with values of 300–400 pptv. Back-trajectories reveal that, for the case studied, the air masses came from an area of thunderstorms located upwind. After the analysis of the various possibilities, the NO_2 increase by lightning production appears to be the most feasible cause. Annual distribution of spikes displays a maximum in late winter and spring during the shift from midlatitude winter tropopause to summer tropopause.

1. Introduction

The partitioning and distribution of nitrogen oxides at the UTLS (Upper Troposphere Lower Stratosphere) in the tropical and sub-tropical regions has recently become a subject of interest. Chemical-Transport-Models underestimate the NO_x in this region (Jaeglé et al., 1998). The budget of NO_x caused by lightning remains as poorly quantified due to the intrinsic difficulties to accurately computing the production in each individual flash and to the irregular distribution in space and time of the thunderstorms. As a consequence, many uncertainties remain about the contribution of each source to the global budget of NO_x. In particular, some authors consider the contribution of lightning as the dominant source (Lamarque et al., 1996) while others (Ehhalt et al., 1992) consider that the main contribution comes from fossil fuel production and aircraft emissions, with lightning in third position. The latter conclusions were also obtained by

Jaeglé et al. (1998) for the USA region.

Significant uncertainties also exist in the magnitude of the overall density in the upper troposphere. Most of the recent studies on UTLS region rely on aircraft observations as the ground based stations in the tropics and subtropics are scarce. Denis et al. (1998) reported no signature of NO_2 contribution by thunderstorms on La Reunion, a tropical/equatorial station located under the ITCZ influence.

At Izaña Observatory (Tenerife, 28° N, 16° W) a long-term program for NO_2 column measurement was started in 1993 and continues to date. The station has recently been accepted as NDSC Complementary Station. Measurements are carried out by using the DOAS technique at zenith during twilight (Noxon, 1979). Typical measurements errors are 1–3%. Details of the instrument and technique have already been reported (Gil et al., 2000).

Very few papers have been published on the seasonal evolution of NO_2 in the tropics or subtropics (Yela et al., 1995; Denis et al., 1998; Yela et al., 1998). The NO_2 column displays a smaller amplitude than at mid and high latitudes and also less day-to-day variability. The summer is a very quiet period dominated almost completely by the easterlies. The day-to-day fluctuation on NO_2 column shows a small amplitude, as it is observed for total ozone as well.

Superimposed on the photochemically driven annual wave sudden abrupt changes in the NO_2 column are observed from time to time. These changes or spikes with periods covering a maximum of two twilights, are always positive (with increase in the column). Typical observed spikes range between 10% and 25% of increase of the mean of the contiguous days (Fig. 1). Frequency distribution is higher in spring for PM data but spikes can also be observed in AM data (see Fig. 2). To avoid vertical transport processes related to clouds, only cases with clear sky conditions are considered. Sky conditions were explored from the O_4 and H_2O records. Those spikes occurring in coincidence with high multiple scattering conditions were rejected.

The purpose of the paper is to discuss the origin of the observed sudden increases in the column by analyzing a case study of a well-defined spike in a clear sky low-aerosol twilight with steady meteorological conditions over the station.

2. A case study: day 132/2000

Day 132/2000 (11 May) has been selected as a first case study since in preceding and subsequent days the conditions of the sky were very steady and the error in the measurements low. The record of Aerosol Optical Depths (AOD), and slant columns of O_4 and H_2O between days 110 and 150 are depicted in Fig. 3. On the days between 128 and 139 AOD was 0.03–0.05. The O_4 column confirms this situation. The H_2O column shows a wet atmosphere on day 132 (Fig. 4).

Tropospheric humidity profiles from Santa Cruz de Tenerife (30 km from Izaña) radiosoundings show three wet well-defined layers peaking at a height of 3500, 7000 and 10 500 m, in addition to the absolute maximum in the boundary layer (MBL) (Fig. 5). The temperature profile shows that the MBL top inversion remains below the station. The height of the MBL inversion is still lower at twilight. Pressure on the station was high and steady.

3. Discussion

Sharp increases in the NO_2 column on clear days can be caused by a) upwelling of pollution from populated areas in the surroundings areas (Santa Cruz or Puerto de la Cruz), b) abrupt change in the meridional component of the stratospheric circulation, c) stratospheric intrusions to the troposphere, d) long range transport from remote polluted centers (namely the US), and e) thunderstorms.

The strong top MBL inversion below the station precludes an eventual upwelling of NO_2 from polluted sites.

Changes in the NO_2 column due to changes in the direction of the meridional component of the stratospheric circulation are a common feature at Izaña (Yela et al., 1998).

Those changes generally take place in time scales of few days and are restricted to the winter season. As already mentioned, in the studied case, North West circulation was found in the lower stratosphere and in the free troposphere. The situation was the same as the preceding and subsequent days. The fact that spikes are always positive (increasing), and their sudden character and short lifetime suggests that they are not the result of meridional transport. We exclude it as the cause of the spikes.

Stratospheric intrusion episodes are known to be a source of NO_x in the upper troposphere. Large distortion of the PV fields around the tropopause during the so-called “tropospheric folding events (TFE)” result in net transport to the troposphere of air masses of stratospheric origin containing large concentrations of O_3 and, eventually NO_2 (i.e. Langford et al., 1996). We explored this possibility as intrusions occur associated to the lower latitude side of the subtropical jet stream. In May the jet is located above the Canary Islands (Cuevas et al., 1999). Unusual high values of potential vorticity are observed north of Tenerife on day 132 at levels between 325 K and 360 K indicating a significant sinking of the tropopause in this area. Although PV-fields show the anomaly extending toward the Canary Islands, we found no sign of change in the ozone column between the spike day 132 and the previous day ruling-out this possibility as the cause of the observed NO_2 increase.

The altitude of the NO_2 enhancements was then explored by comparing the NO_2 slant column evolutions versus *sza* for the “event” and “not event” day. The ratio between two contiguous dusks (Fig. 6, lower panel, solid circles) can be formulated as follows:

$$r(sza) = \frac{SCD_{132}(sza)}{SCD_{131}(sza)} = \frac{VCD_{132}(sza)}{VCD_{131}(sza)} \cdot \frac{AMF_{132}(sza)}{AMF_{131}(sza)} \qquad (1)$$

Where $SCD132$ stands for slant column density plus the amount contained in the reference spectrum, VCD is the vertical column density and AMF is the air mass factor. All parameters are *sza* dependent.

If we assume that the AMF remains constant on both days (the increase in NO_2 does not disturb the shape of the profiles and there is no change in aerosols) and that VCD does not vary in the same day except for photochemical reasons then $VCD132=VCD131.f$, where f is the observed increase factor between the two days

$$r(sza) = \frac{SCD_{132}(sza)}{SCD_{131}(sza)} = \frac{VCD_{132}(sza)}{VCD_{131}(sza)} = f = constant \quad (2)$$

Therefore the ratio should be independent of sza. As this is not the case, there might be a number of possibilities: a) the NO_2 column is increasing dynamically during the evening b) the increase in r is an artifact due to AMF changes as results of the modification of the shape of the NO_2 vertical profile, and c) a combination of a) and b). Let us assume that option b) takes place, then;

$$r(sza) = \frac{SCD_{132}(sza)}{SCD_{131}(sza)} = f \cdot p(sza) \quad (3)$$

being

$$p(sza) = \frac{AMF_{132}(sza)}{AMF_{131}(sza)} \quad (4)$$

The evolution of r with the sza follows the AMF ratios. A radiative transfer code for zenith measurements (Sarkissian et al., 1995) has been run assuming the absorbent layer at different altitudes between 4 and 32 km. The resultant curves have been divided by the AMF-standard computed for an AFGL tropical profile centered at 28 km.

In Fig. 7 the $p(sza)$ factor has been plotted for different layer height assumptions versus the sza. By comparing the curves with the NO_2 ratio in Fig. 6 around twilight (sza 85° to 92°), it can be seen that the best correlation occurs for an NO_2 bulk height estimation of 12 km ($r2=0.85$). We have named this approach the SOTARC technique (from Slant Observations To Amf Ratio Correlation) for height estimation of shallow layers containing large concentrations of species.

Below sza 80° there is a discrepancy between the r-computed and the r-observed that may be attributed to real increase in the column during the measurements. In other words, the previously mentioned effects, *AMF*-changes and NO_2 amount changes take place simultaneously.

As for most of the cases considered in Fig. 7, *r-AMF* are smaller than the standard values used, the real vertical columns differ from those computed by a factor which is height dependent. If we assume that the extra NO_2 layer is located at approximately 10 km, then the underestimation would be as much as 20%. At 12 km the modeled ratio is close to one at 90°. As the vertical columns are obtained using values close to 90° and since the thickness of the layer might be of 2–3 km, the real change as compared to the previous day after *AMF* correction ranges between 18% and 38% increase.

Klemm et al. (1998) advise of the difficulty of establishing a clear separation between troposphere and stratosphere from a chemical point of view. They define a large transition zone of up to 2 km where mixing takes place. In May, the Subtropical Jet stream (STJ) is located at slightly higher latitudes than the Canary Islands and the spectrometer observed tropical air masses typical of the summer period. As the tropopause on those days was located at, or above, 14 km, the "dense" NO_2 layer clearly lie in the troposphere. Assuming that the depth of the layer extends from 10 to 12 km, the observed increase of the column (6.5E14 molec/cm^2) represents around 300–400 pptv.

It is worthwhile noting that for ozone the ratio *r* does not depart from the unity, indicating that the NO_2 enhancement is not related to O_3 photochemistry or the signal is too low to be identified.

The back-trajectory analysis in the upper troposphere (around 10 km altitude) shows that the air masses crossed North America. Computed isentropic backtrajectories at 324 K (~7 km), 332 K (~10.1 km), 334 K (~11 km) and 344 K (~12.5 km) for May 11 (19:40 GMT) show a very fast air mass transport form US to Canary Islands of about 3 days (not shown in the reduced domain of Fig. 8. Schultz et al. (1998) were able to reproduce the concentrations of several gases measured at Izaña using a photochemical trajectory model with trajectories initiating above the USA. The initial values required were in agreement with measurements carried out in the free troposphere over North America, suggesting that high concentrations of NO_x can be measured at

Izaña due to decomposition of transoceanic transported peroxyacetyl nitrate (PAN). While this mechanism could be considered in the lower free troposphere, it is unlikely in the upper troposphere. NO_x lifetime in the boundary layer is too short (about 0.5 days in spring) (Liang et al., 1998) to reach the higher levels at the measured concentrations. Jaeglé et al. (1998) found average concentrations of 370 pptv of NO_y in the upper troposphere of boundary layer origin transported upward by convection. NO_x concentration is of 70 pptv at the same altitudes above the most polluted areas of USA. Assuming that no NO_2 losses occur during transport above the Atlantic, concentrations measured at Tenerife are roughly ten times higher than those encountered above the USA. Amounts are, therefore, too large for long-range transport consideration.

We then investigated the occurrence of thunderstorms in the area during that period. ECMWF (European Centre for Medium-Range Weather Forecasts) analysis reveal a cyclonic system located on 9, 10 and 11 May NW of the Canary Islands with associated strong convection (Fig. 8). The system travels SE at 35–40 km/h passing the main center to the north of Tenerife on 11th May during the afternoon. High cloud tops are observed from false color Meteosat IR channel on 10 May (12 GMT) and on 11 May (00 GMT).

Data from the Lightning Imaging Sensor (LIS), aboard the TRMM Observatory display low lightning activity on 9 May over the area where the low pressure system is located at about 500 km NW of Tenerife.

The DOAS technique at zenith scans air masses located some 200 km in direction to the sun, westward of the station at dusk (Solomon et al., 1987); therefore the air mass measured by our instrument on the afternoon of the 11 May came from the thunderstorm area, as it is indicated in Fig. 8. The ending points of the trajectories correspond to the time of the Meteosat image. It is necessary to note that backtrajectories below (324 K) or above (344 K) the 10–12 km altitude layer also come from US. However only backtrajectories confined in the 10–12 km layer, where the NO_2 peak is observed, hit clearly the storm. This fact precludes a long-range pollution event and reinforces the hypothesis of the storm influence on the NO_2 record.

NO_x lifetime in the upper troposphere is around 5–10 days (Jaeglé et al., 1998) more than enough to reach the observatory. Pickering et al. (1998) found that most of the NO_x production for the tropical marine conditions at the end of the storm was located in the upper troposphere, in a layer centered at 10–11 km. The NO_x at this level is not only the result of lightning in the anvil but of updraft transportation. The height where most of the NO_x is stored is consistent with that retrieved from our observations and within the range of values measured by Chameides et al. (1987) and computed by modeling (Pickering et al., 1998) for a tropical marine regime in the Pacific.

4. Conclusions

Spikes in the NO_2 column record at the subtropical high altitude Izaña station are observed few times a year on am and pm data with amplitudes to up of 25% of the column in contiguous days. We have searched for the origin of these sudden increases in the column. Among the possibilities of local, regional and long-transport pollution, stratospheric intrusions and lightning, the latter appears as the only feasible mechanism in the case studied. By comparing the ratio of the NO_2 slant column versus *sza* between two days (the day of the spike and the previous day) with the ratios of *AMF* it is estimated that the NO_2 increases take place in the upper troposphere at 10–12 km. The concentration found of 300–400 pptv is compatible with NO_x production by lightning in an air mass coming from a thunderstorm that on those days was located Northwest, upwind of the Observatory.

The Slant observation to *AMF* ratio correlation technique constitutes a novel approach for shallow layer height estimation containing large concentration of species of atmospheric interest that can be successfully employed on days with clear skies.

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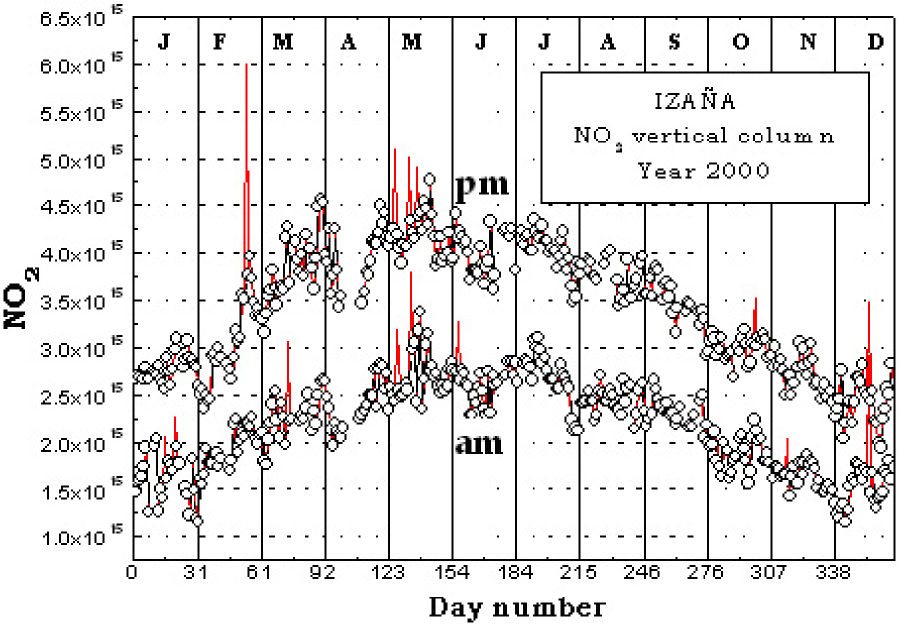

Fig. 1. Annual evolution of the NO_2 column for year 2000 measured at twilight. Spikes, as defined in the text, are shown in red.

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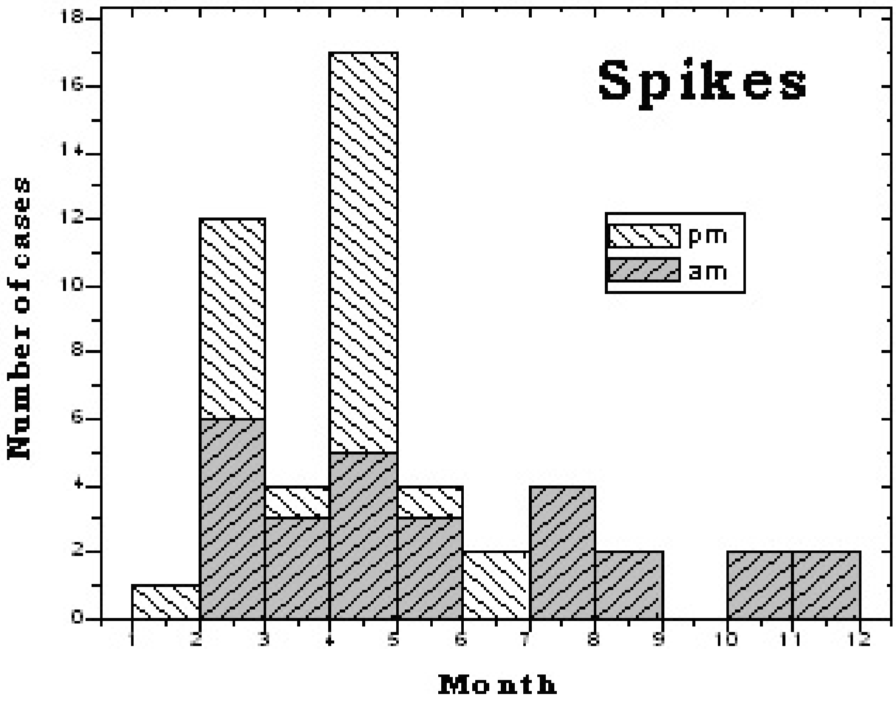

Fig. 2. Frequency distribution of spikes from 1993 to 2000 observed in the twilight records as defined in the text.

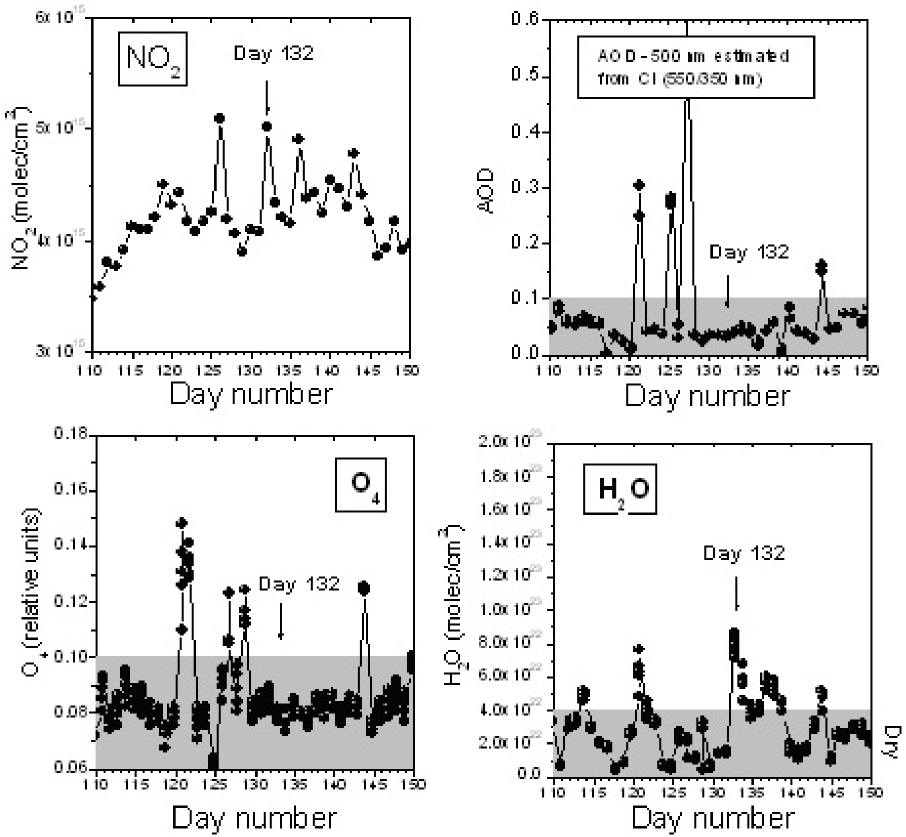

Fig. 3. The case study day 132 was clear and clean. The evolution of the Aerosol Optical Depth (AOD) (top right panel) estimated from the Color Index defined by the ratio of the intensities at zenith 550/350 nm, dimer of the molecular oxygen (lower left panel) and slant water vapor (lower right panel) are shown. Shaded in grey values representative of clear skies. Day 131 has been used as reference.

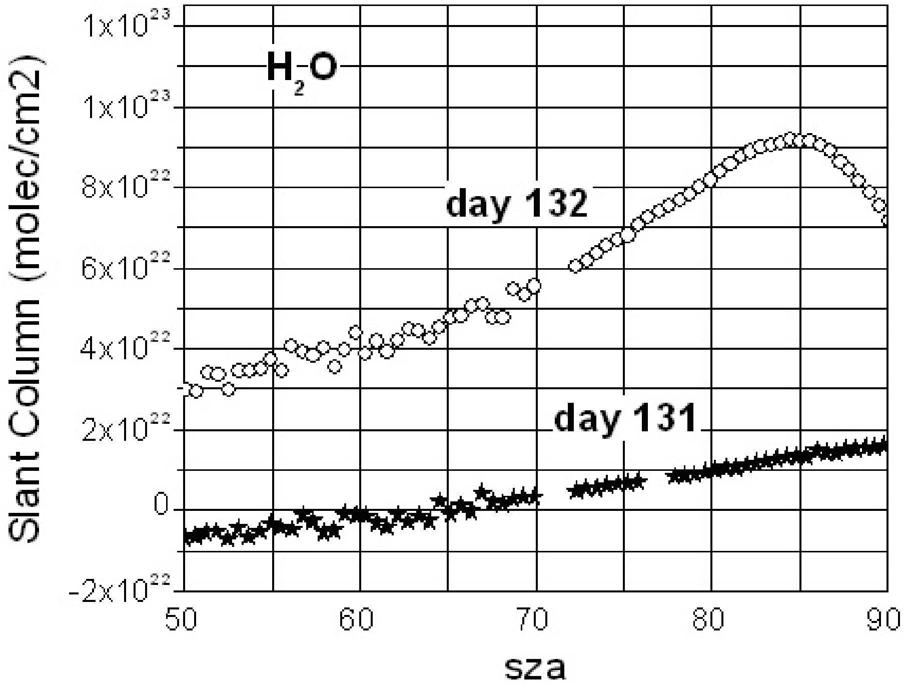

Fig. 4. Slant column of water vapor for days 131 and 132.

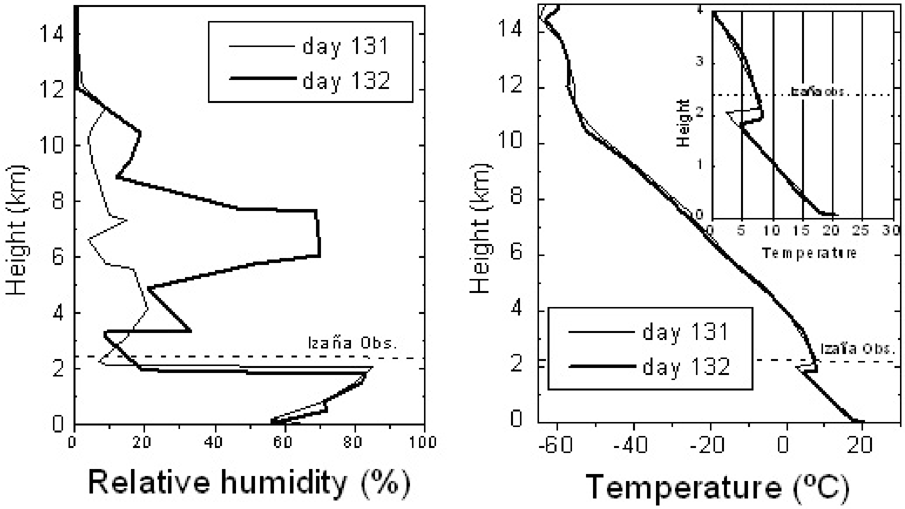

Fig. 5. Left: Relative humidity (%) from radiosounding profiles above Tenerife for clear sky days 131 (thin line) and 132 (thick line). Two wet layers at 5–9 km and 9–12 km are observed on day 132 when compared to the previous day. The sharp RH% reduction at 2 km defines the top of the marine boundary layer (MBL). The height of the station is also shown. Right: Temperature profile for the same day.

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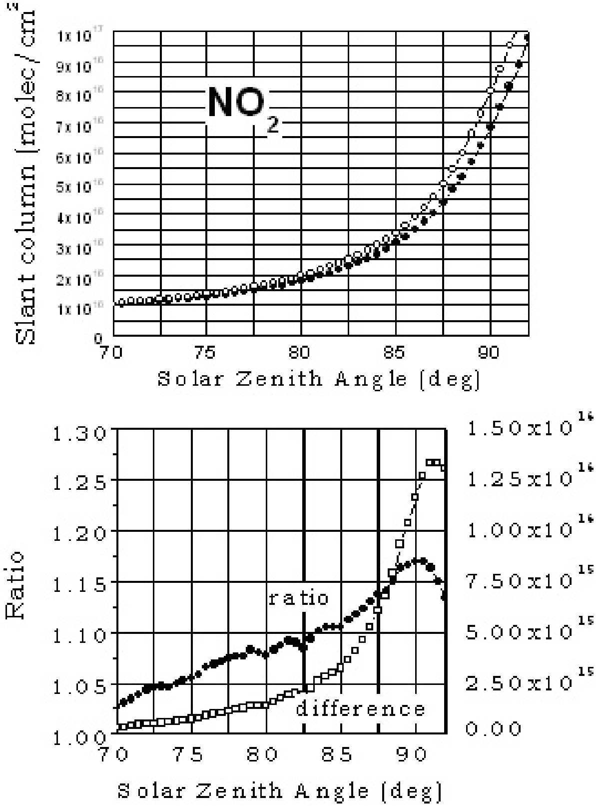

Fig. 6. Upper panel: NO$_2$ pm slant columns for the day of the spike (132, open circles) and the previous day (131, filled circles). Lower panel: ratio and differences between both curves.

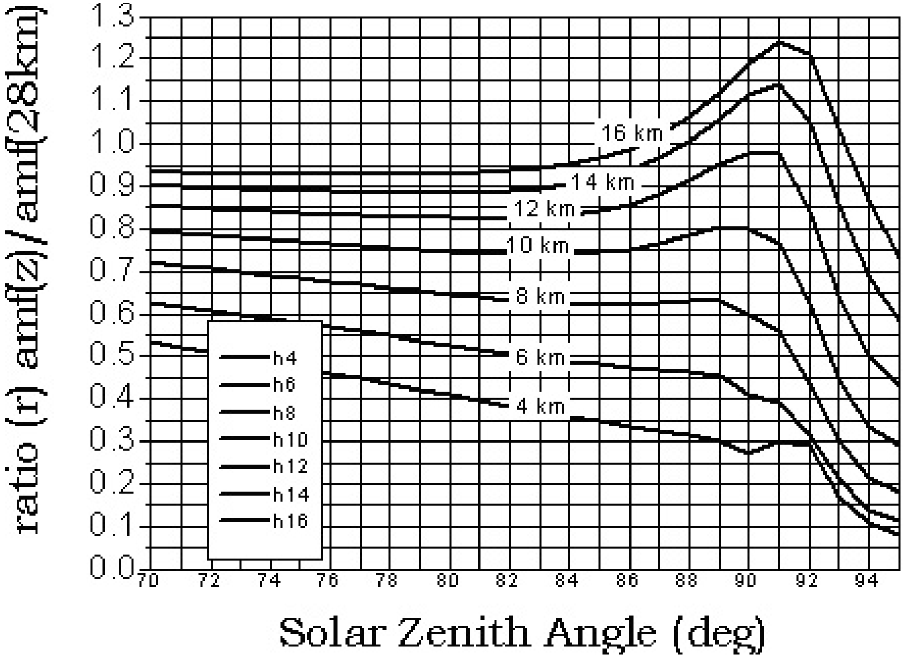

Fig. 7. Ratio obtained by dividing *AMF* computed assuming the layer at a given height by the standard *AMF* (height=28 km).

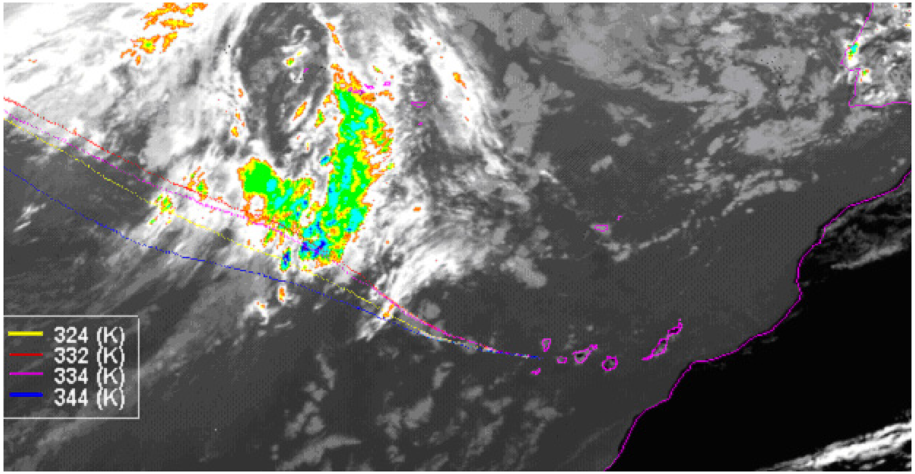

Fig. 8. Infrared channel of the Meteosat satellite for 10 May 2000 (12 GMT) showing in false colour (yellow and green) the active centers of the low pressure system. Trajectories at isentropic levels of 324 K (~7 km), 332 K (~10.1 km), 334 K (~11 km) and 344 K (~12.5 km) ending at the measurement point (200 km sunward of the observatory) on 11 May pm (19:40 GMT).

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