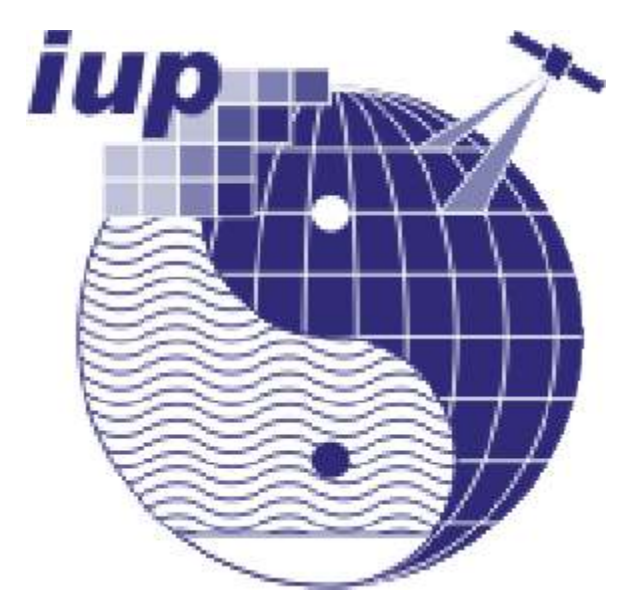
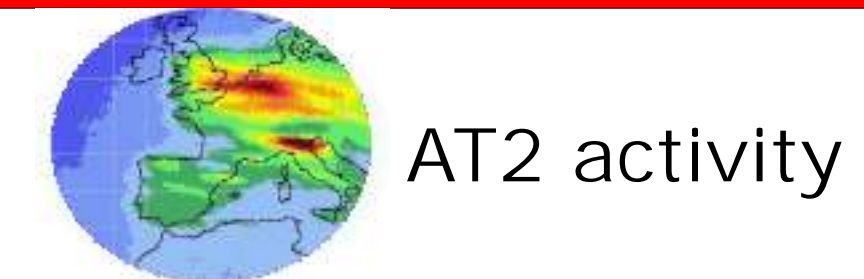


# Pollution events over the East Mediterranean : Synergistic use of GOME, ground based and sonde observations and models

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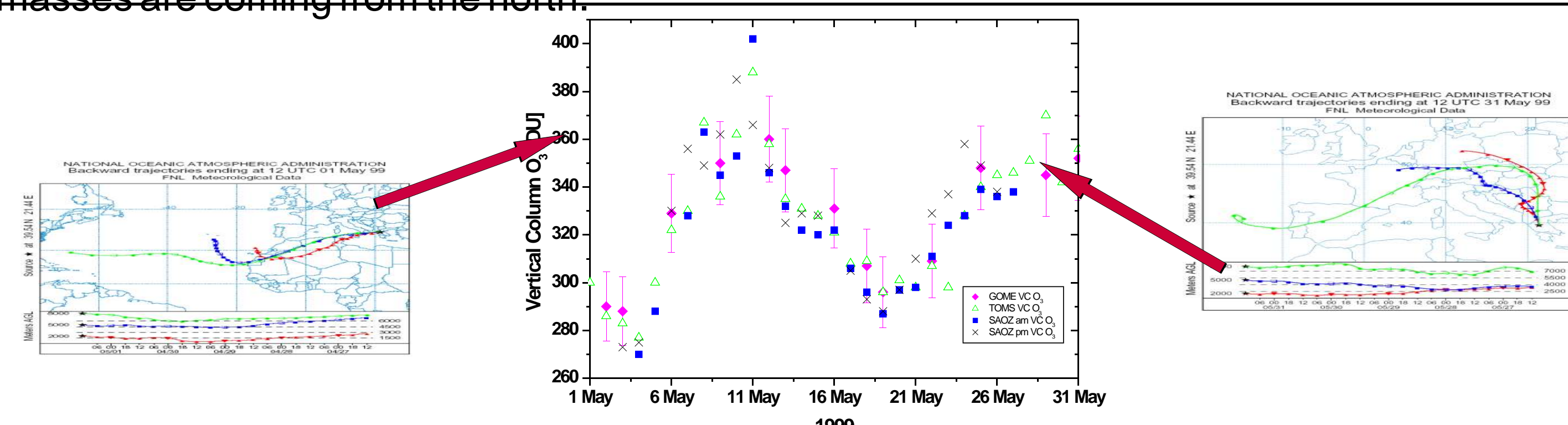
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## Introduction

Retrievals of trace gas columns of  $O_3$ ,  $NO_2$  and HCHO from the measurements of backscattered radiation by GOME (Global Ozone Monitoring Experiment) [Burrows et al. 1999 and 2000], interannual comparisons and comparisons with Systeme d'Analyse par Observation Zenithale (SAOZ) [Goutail et al. 1999], balloon [Thompson et al. 2003; Simeonov et al. 1998, Calpini et al. 1997] and LIDAR measurements show that the East Mediterranean in spring, and particularly in May 1999 during the international PAUR II campaign in Crete carried out at Finokalia (35°24' N; 25°60') by the University of Crete [Kouvarakis et al., 2001], is influenced normally by background conditions but also by biogenic emissions and urban pollution related to distinct changes in wind direction between north and south. When following air masses along the trajectory we derived that air masses transported from N-NW (North-North-West) Europe towards the Mediterranean region are associated with an increase of tropospheric amounts of  $NO_2$  by a factor of 1.4 to 1.5 and of HCHO by a factor of 1.6 to 1.8 as can be seen from GOME. Box model calculations show that such an increase of tropospheric amounts of  $NO_2$  and HCHO column enhanced the tropospheric  $O_3$  columns by only 1-2 DU per day locally. In addition to that backward trajectories emerging Crete were calculated to analyse the amount of 11-19 DU from the intrusion of stratospheric air masses into the troposphere during May 1999. As follows the long-range transport of  $O_3$  rich air masses and its precursors from industrial areas towards the Mediterranean region as well as stratospheric-tropospheric-exchange (STE) contribute mainly to enhanced tropospheric  $O_3$  levels both at the ground and in the free troposphere.

## Results

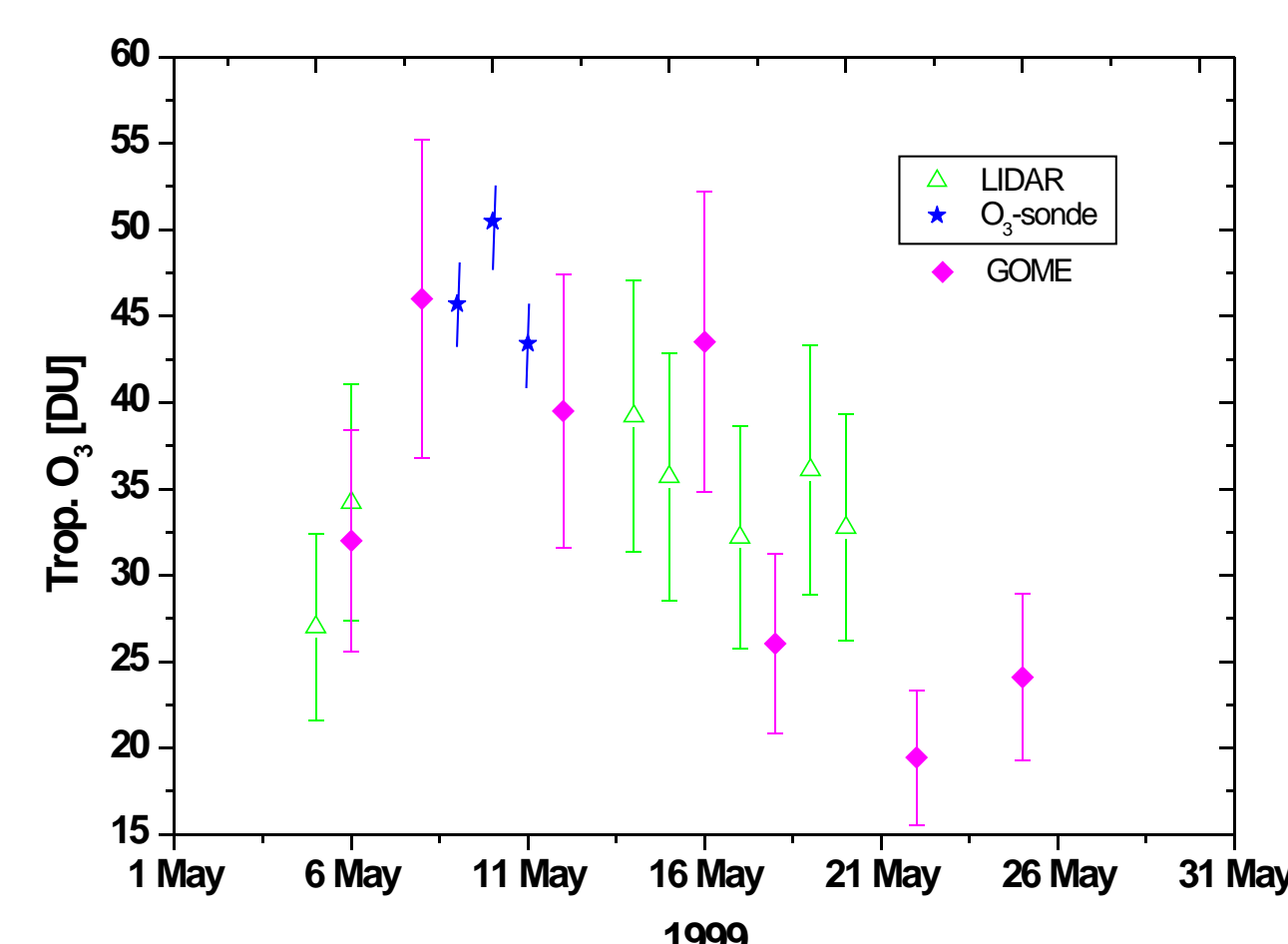
The comparison of the vertical columns of  $O_3$  from GOME [Burrows et al. 1998 and 1999] with SAOZ (Systeme d'Analyse par Observation Zenithale) and TOMS (Total Ozone Mapping Spectrometer) (a.m. and p.m. data [Goutail et al. 1999] data show a variation of 120 DU (Dobson Units) during May 1999 over Crete (see Fig 1a) situated in the sub-tropical region. From the calculation of 5-day back trajectories it can be seen that in the case that Crete is affected by south winds (influence by the transport of air masses from the tropics) the total columns of  $O_3$  are in the range of 270 DU (1<sup>st</sup> to the 4<sup>th</sup> of May 1999, see Fig. 1b) whereas high  $O_3$  columns up to 402 DU (5<sup>th</sup> to the 10<sup>th</sup> and 28<sup>th</sup> to 31<sup>st</sup> of May 1999, see Fig. 1c) are reached for the same trace gas when air masses are coming from the north.



**Figs. 1a/b/c.** Total columns of  $O_3$  measured by SAOZ and TOMS system and compared with GOME data for May 1999. 5-day back trajectories of air masses on May 1, 1999 (left) and on May 31, 1999 (right) showing strong influence from NW Europe and Balkans over Crete. The calculations were performed using the HYSPLIT 4 model (<http://www.arl.noaa.gov/ready/hysplit4.html>).

The comparison of the tropospheric column amounts of  $O_3$  based on ozonesonde [Thompson et al. 2003], LIDAR [Simeonov et al. 1998, Calpini et al. 1997] and GOME data show (see Fig. 2) an increase of 23 DU (27DU up to 50DU) e. g. between the 5<sup>th</sup> and the 10<sup>th</sup> of May 1999 (north-wind) followed by a reduction down to background conditions of 20 DU (a value that is close to the 24 ±5 DU estimates by Hudson and Thompson [1998] for the background tropospheric  $O_3$  column in the tropical regions) on the 22<sup>nd</sup> of May 1999 (south wind).

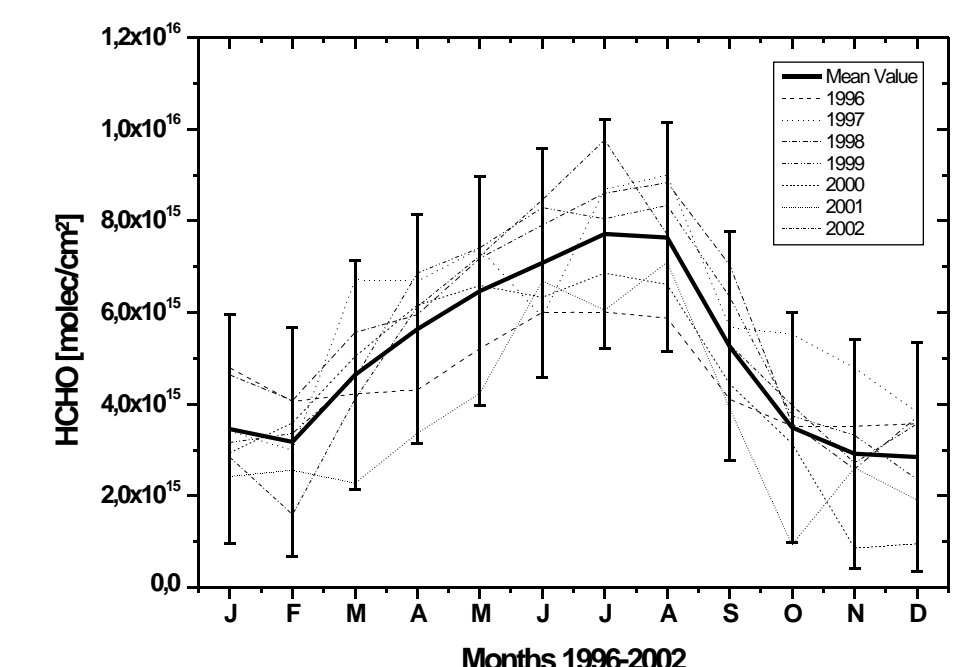
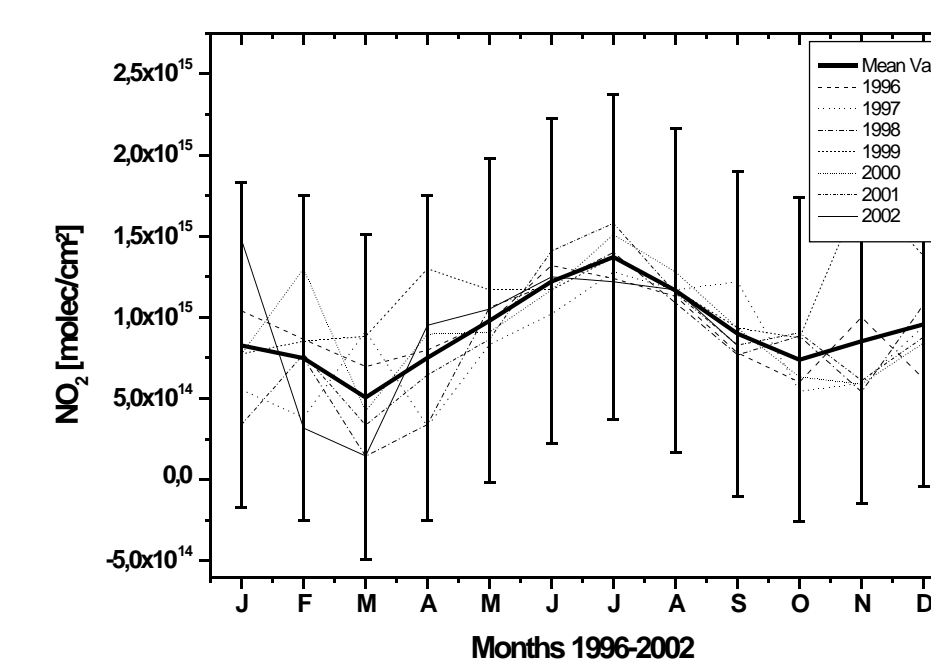
During the pollution events when air masses were transported from N-NW (North-North-West) Europe and the Balkans (more polluted compared to the background conditions over the Mediterranean, see Tab. 1) towards the Mediterranean region the precursors of tropospheric  $O_3$ ,  $NO_2$  and HCHO show an increase of a factor of 1.4-1.5 and of 1.6-1.8 respectively as can be seen from GOME when following air masses along the trajectory. Box model calculations show that such an increase of tropospheric amounts of  $NO_2$  and HCHO column enhanced the tropospheric  $O_3$  columns by only 1-2 DU per day. As no trend was observed for  $NO_2$  and HCHO tropospheric columns for the time period of 1996 to 2002 (see figs. 3a/b) no significant increase of locally produced tropospheric  $O_3$  is estimated.



**Fig. 2.** Variation of tropospheric vertical columns of  $O_3$  from satellite based observations by GOME, ozonesondes and ground based LIDAR observations as carried out at Nopigia, Crete for May 1999.

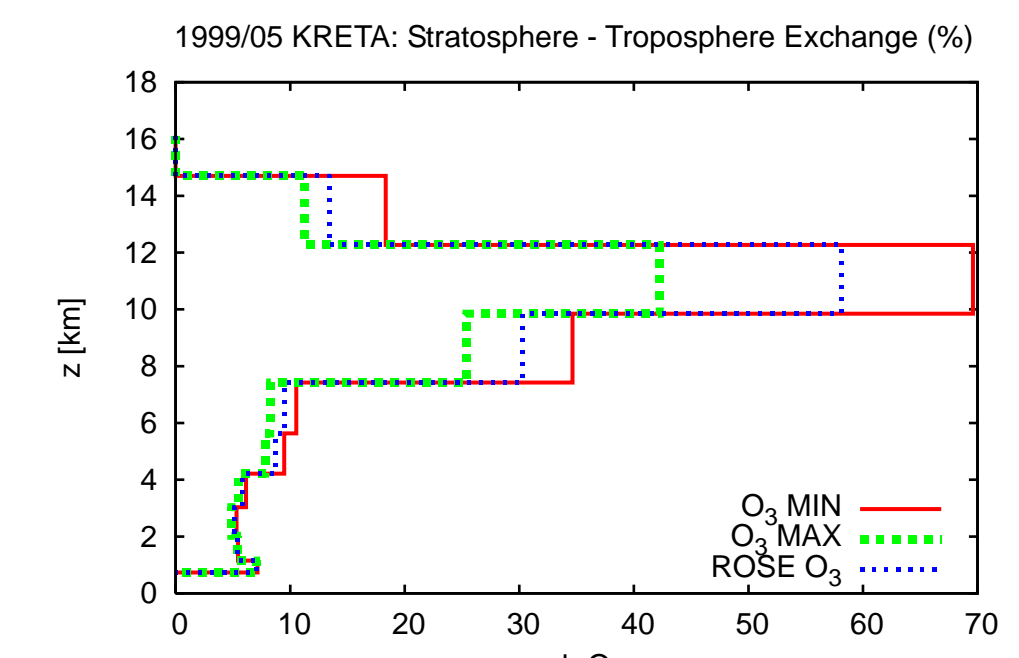
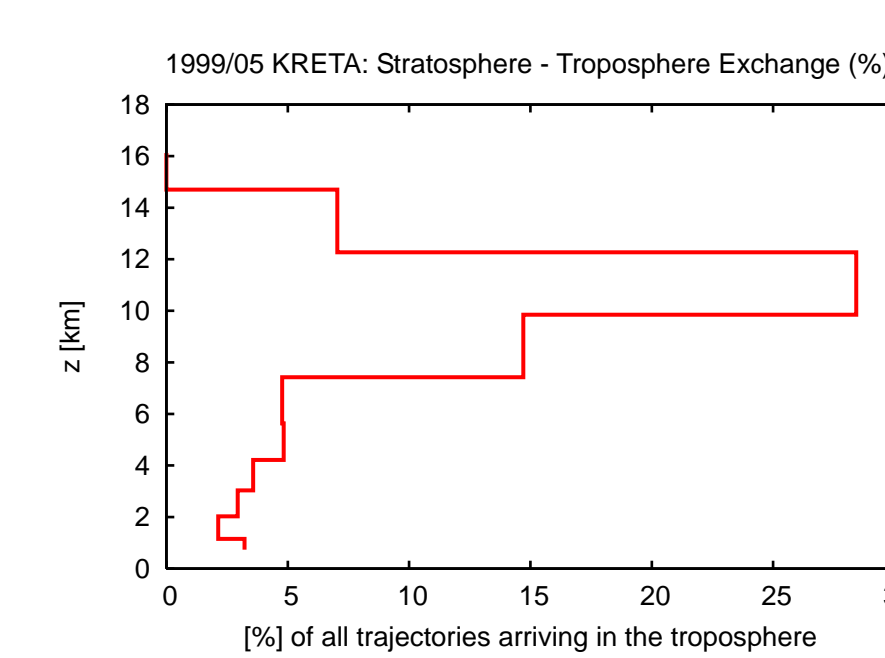
Trace Gas	Crete	Atlantic	Salonika	Po Valley	Istanbul
$NO_2$	$1.2 \times 10^{15}$	$6.0 \times 10^{14}$	$2.7 \times 10^{15}$	$4.4 \times 10^{15}$	$2.7 \times 10^{15}$
HCHO	$4.6 \times 10^{15}$	$3.1 \times 10^{15}$	$6.1 \times 10^{15}$	$6.6 \times 10^{15}$	$5.9 \times 10^{15}$

**Tab. 1.** Mean values of tropospheric  $NO_2$  and HCHO (given in molecules  $cm^{-2}$ ) for the month May based on GOME data for different regions.



**Figs. 3a/b.** The monthly values over Crete of total column of tropospheric (a)  $NO_2$  and (b) HCHO for the years 1996-2002 [all in molecules  $cm^{-2}$ ] as retrieved from GOME observations.

In addition to photochemical activity during this time period stratospheric tropospheric exchange (STE) is probably responsible for a significant fraction of the tropospheric  $O_3$  [Vaughn et al. 2000, Randriambelo et al. 1999]. As can be seen from figure 4a in a height of 10-12 km 30% of all trajectories arriving the troposphere are coming from the stratosphere. In the same altitude layer 70ppb  $O_3$  can be expected based on the intrusion of air masses from the stratosphere into the troposphere. The calculation of the tropospheric column caused by STE including the chemical conversion of  $O_3$  to OH shows values in a range of ~11.2 DU using the minimum, maximum data and the output of the 3D Chemistry-Transport Model ROSE [data are available from the World Data Center for Remote Sensing of the Atmosphere, WDC- RSAT (<http://wdc.dlr.de>)]. A similar increase can be received by the analysis of LIDAR  $O_3$  profile observations (an increase of ~11 DU per day).



**Figs. 4a/b.** Altitude dependent influence of stratospheric and tropospheric air masses arriving over Finokalia (a) in May 1999. The percentage scale shows the differences of the stratospheric influence in the mid and lower troposphere for this location on a base of 10-day back trajectory calculation. The amount of

## Conclusions

The Mediterranean region is mainly influenced by clean air conditions but from time to time an increase of the total columns as well as of the tropospheric column amounts of  $O_3$ , associated with the transport of air masses from the north direction towards the Mediterranean region, can be seen from GOME data compared with SAOZ, TOMS and sonde measurements. Model calculations (TM3 and ROSE) showed that only ~1.33 DU are photochemical produced whereas ~11.2 are caused by transport processes mainly from the stratosphere irreversible to the troposphere (STE, ~11.2 DU).

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