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# Influence of stratospheric airmasses on tropospheric vertical $O_3$ columns based on GOME (Global Ozone Monitoring Experiment) measurements and backtrajectory calculation over the Pacific

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**Abstract.** Satellite based GOME (Global Ozone Measuring experiment) data are used to characterize the amount of tropospheric ozone over the tropical Pacific. Tropospheric ozone was determined from GOME data using the Tropospheric Excess Method (TEM). In the tropical Pacific a significant seasonal variation is detected. Tropospheric excess ozone is enhanced during the biomass burning season from September to November due to outflow from the continents. In September 1999 GOME data reveal an episode of increased excess ozone columns over Tahiti (18.0° S; 149.0° W) (Eastern Pacific) compared to Am. Samoa (14.23° S; 170.56° W) and Fiji (18.13° S; 178.40° E), both situated in the Western Pacific. Backtrajectory calculations show that none of the airmasses arriving over the three locations experienced anthropogenic pollution (e. g. biomass burning). Consequently other sources of ozone have to be considered. One possible process leading to an increase of tropospheric ozone is stratosphere-troposphere-exchange. An analysis of the potential vorticity along trajectories arriving above each of the locations reveals that airmasses at Tahiti are subject to enhanced stratospheric influence, compared to Am. Samoa and Fiji. As a result this study shows clear incidents of transport of airmasses from the stratosphere into the troposphere.

## 1 Introduction

The tropical Pacific is often considered to be a region remote from major polluting influences because of its isolation from industrialized landmasses. However, the signature of pollution, particulary from biomass burning, makes a significant imprint on the air chemistry of the region (Singh et al., 2000), (Thompson et al., 2003). 7 years of GOME data were analysed with respect to tropical tropospheric excess $O_3$. The tropospheric excess ozone is the difference between the total amount of tropospheric ozone and its tropospheric background. The tropospheric excess ozone is globally derived from GOME data by applying the Tropospheric Excess Method (TEM) (Fishman et al., 1990). Another method to derive tropospheric $O_3$ columns from the measurements of a nadir-looking instrument is the Convective-Cloud-Differential (CCD) method which is discussed by (Valks et al., 2003) and (Ziemke et al., 1998).

The differences of monthly mean tropospheric excess $O_3$ between Tahiti and Fiji as well as Tahiti and Am. Samoa reveal frequently enhanced values at Tahiti for all years of GOME measurements (see Fig. 1). Similar observations of enhanced tropospheric $O_3$ values over Tahiti compared to Fiji and Am. Samoa can be seen by analysing $O_3$-sonde data (Oltmans et al., 2001). Figure 2 shows the timeseries of GOME tropospheric excess ozone for the three locations for one epsiode. At Tahiti (18.0° S; 149.0° W) the tropospheric excess ozone peaks at 12DU whereas at Am. Samoa (14.23° S; 170.56° W) and Fiji (18.13° S; 178.40° E) 3DU are not exceeded. The main objective of this case study is the analysis of one episode of enhanced tropospheric excess ozone at Tahiti which took place in September 1999. The question arises which processes may lead to such differences in tropospheric excess ozone between the Western (Am. Samoa and Fiji) and the Eastern Pacific (Tahiti).

Therefore in this case study an analysis of backtrajectories was carried out in order to analyse whether the probed airmasses were either influenced by processes taking place in the troposphere only (e. g. biomass burning) or by irreversible stratosphere-troposphere-exchange as for example discussed by Waugh et al. (2000) and Randriambelo et al. (1999). Backtrajectory analysis has become a widely used method for the interpretation of trace gas measurements in relation to large scale airmass transport (Moody et al., 1995).

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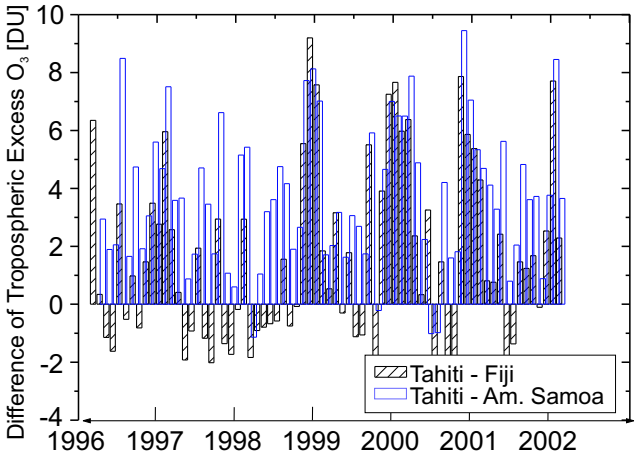


**Fig. 1.** Calculated differences of monthly mean values for tropospheric excess column amounts between Tahiti and Am. Samao as well as between Tahiti and Fiji for 1996–2002.

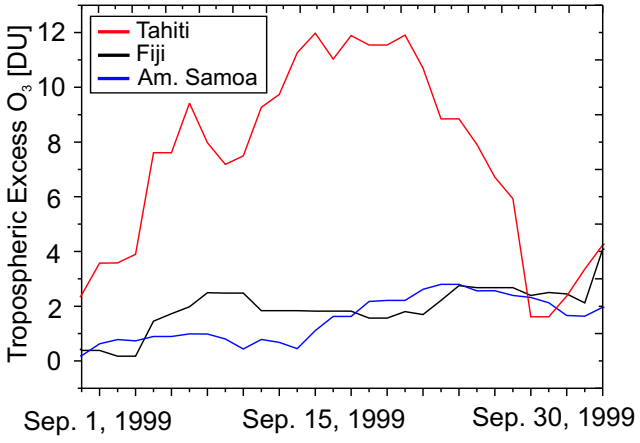


**Fig. 2.** Time series of the tropospheric excess column of ozone between 1 September to 30 September 1999 for the locations Am. Samoa, Fiji and Tahiti.

## 2 Methods and results

### 2.1 GOME

GOME was launched in April 1995 onboard the second European satellite ERS-2 in a sun-synchronous near polar orbit for measuring the sunlight scattered from Earth's atmosphere and/or reflected by the surface in nadir mode in a wavelength region of 240 to 790nm with a spectral resolution of 0.2–0.4nm. Once per day, the extraterrestrial solar irradiance is measured and can be used as an absorption free background in the data analysis carried out using the Differential Optical Absorption Spectroscopy (DOAS) technique (Burrows et al., 1999), (Platt, 1994). The main scientific objective is to measure the global distribution of $O_3$ and several trace gases which play an important role in the ozone chemistry of the earth's stratosphere and troposphere (Burrows et al., 1999). The local equator crossing time is 10:30 a.m. With 14 orbits per day global coverage at the equator is reached after three days for a 960km swath width (the size of one GOME ground pixel is $40\times320\text{km}^2$) (Burrows et al., 1999).

For this study GOME level-2-data (GOME products are computed from level-0 (raw) data using the GOME Data Processor (GDP) system which was designed and developed by the German Remote Sensing Data Center (DFD)) were extracted with a maximum distance of 200km away from the location of interest to derive vertical columns of $O_3$ in the 325.5 to 334.5nm wavelength region. The details on the analysis are given elsewhere (Ladstätter-Weißenmayer et al., 1998), (Richter et al., 2002) and are not repeated here.

As GOME is a nadir viewing instrument, both tropospheric and stratospheric absorptions contribute to the measured signal. The stratospheric $O_3$ column is dominated compared to the tropospheric amount. Therefore, the Tropospheric Excess Method (TEM) (Fishman et al., 1990), (Leue et al., 2001), (Richter et al., 2002) was used to derive the tropospheric excess columns of $O_3$. This method is based on the assumption that stratospheric $O_3$ does not vary zonally, and therefore the total columns measured on the same day at the same latitude over a clean air region can be used as an approximation of the stratopheric column over the region of interest. Here, data from Atlantic region (315–325° E) were used as reference section. The overall analysis then consists of three steps: (a) determination of the total slant column, (b) subtraction of the stratospheric contribution including the tropospheric background yielding the tropospheric excess slant column and (c) division by the airmass factor (AMF) to obtain the vertical tropospheric excess column (Rozanov et al., 1997).

There are general limitations using this method which introduce some uncertainty into the results. A number of assumptions have to be made for the calculation of the airmass factors which also contribute to the overall error of the GOME measurements. The main error sources are the inhomogeneities in the stratospheric $O_3$ field, uncertainties in cloud cover, the assumed vertical profile of $O_3$, the surface albedo and the aerosol loading which are required in calculating the AMF as described elsewhere (Richter et al., 2002). The overall error of the analysis is estimated to be in the order of 4DU. This is based on the comparison of a large number of Southern Hemisphere Additional Ozonesondes (SHADOZ) measurements with ozone columns derived from GOME. The uncertainty of 4DU influences the total column amount of $O_3$ for the GOME data over all three locations similarly. That means differences in the results can be considered as relative deviations.

For this study the excess vertical columns of $O_3$ based on GOME data for September 1999 with a cloud cover of less than 10% (due to GOME ICFA cloud algorithm) were analysed (see Figs. 1 and 2) for the Pacific region. The time series of the excess vertical columns of $O_3$ in DU (Dobson Units)

are shown in Fig. 2 for Tahiti, Am. Samoa and Fiji. Tropospheric excess vertical columns over Tahiti of up to 12DU were measured whereas over Fiji and American Samoa the values are close to zero (between 0 and 3DU). Similar results that means more variability of the $O_3$-profile can be seen over Tahiti compared to Am. Samoa and Fiji using the ozone sondes measurements, as described elsewhere (Oltmans et al., 2001). To determine whether anthropogenic emissions or stratospheric influence may explain differences of the ozone columns between the three locations, backtrajectory calculations using the trajectory model Traj.x (Meyer-Arnek, 2002), (Meyer-Arnek, 2003) were performed.

## 2.2 Backtrajectory analysis and discussion

To assess the differences in the excess tropospheric ozone over Am. Samoa, Fiji and Tahiti backtrajectories were calculated. Over each of the locations individual clusters of backtrajectories were started every six hours (at 0:00 UT, 6:00 UT, 12:00 UT and 18:00 UT) during September 1999 at ten different heights (900, 800, 700, 600, 500, 400, 300, 200, 150 and 100hPa) covering the troposphere and tropical tropopause region. All trajectories traveled backwards in time for 10 days. Altogether 145 000 trajectories were calculated by Traj.x for each site. Traj.x is a trajectory model developed at the Institute of Environmental Physics, University of Bremen. It is a kinematic trajectory model which calculates the transport of airparcels due to meteorological wind fields by applying a fourth-order Runge-Kutta scheme. Meteorological data were derived from European Centre for Medium-Range Weather Forecasts (ECMWF)'s 40-year's reanalysis data set (ERA-40). These data are available on 60 model levels at four times a day (at 0:00 UT, 6:00 UT, 12:00 UT, 18:00 UT). The analysis of all trajectories arriving over each of the three locations reveals that anthropogenic emissions can not be responsible for the differences in tropospheric ozone between the three Pacific locations. Any continental emission (for example from biomass burning) affects all locations nearly equally. This is proven by Fig. 3. It depicts the trajectory density of trajectories arriving over the three locations Tahiti, Fiji and Am. Samoa while residing in the planetary boundary layer (PBL).

The trajectory density is derived by projecting all trajectories residing in the lowest 2km (which is assumed to represent the PBL) onto a 1.125° latitude-longitude grid. Assuming that airmasses are only influenced by anthropogenic processes taking place on the earth's surface while residing within the PBL and assuming that no major anthropogenic emission takes place on the ocean there must be another source of tropospheric ozone. The advection of stratospheric airmasses with high ozone loading into the troposphere may increase the amount of tropospheric ozone (Galani et al., 2003).

In order to probe whether stratospheric airmasses enter the troposphere the potential vorticity (PV) criteria was applied.

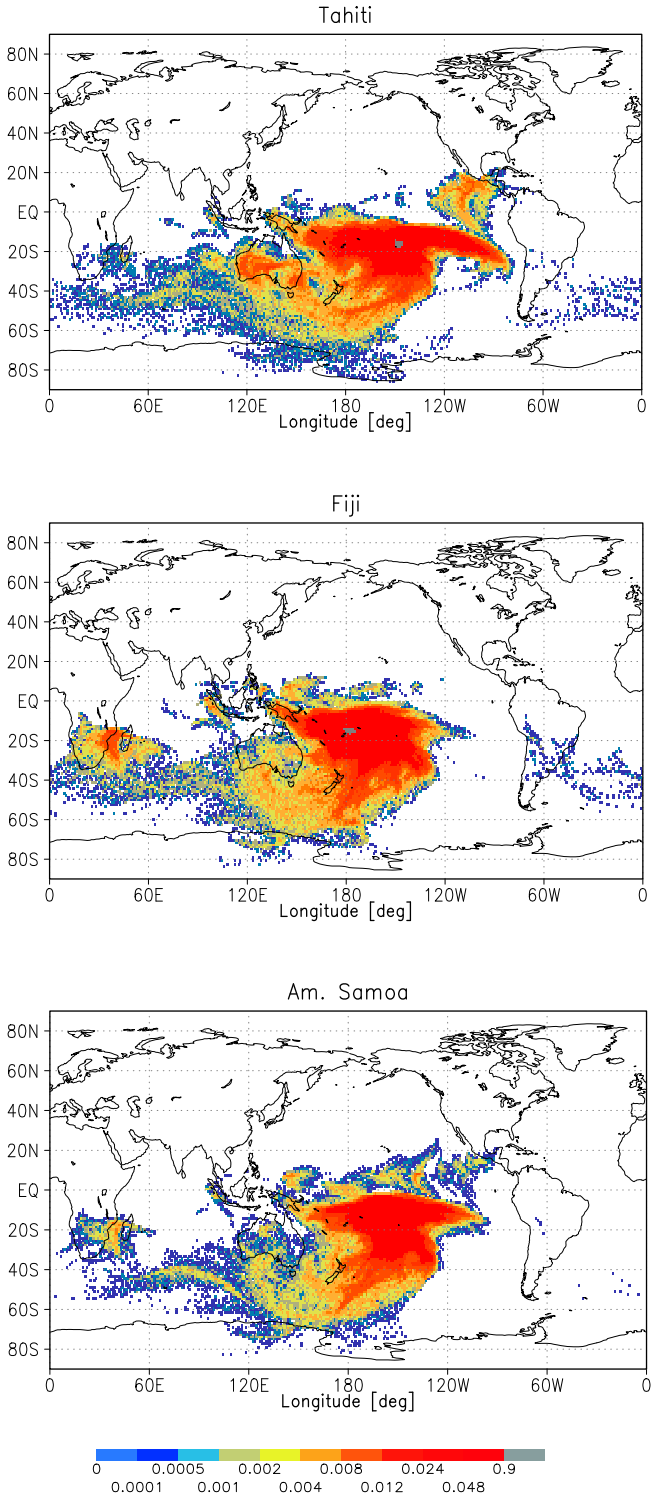


**Fig. 3.** Density of trajectories arriving above Tahiti, Fiji and Am. Samoa during September 1999 while residing in the planetary boundary layer. For each of the three locations, this quantity is normalised to one.

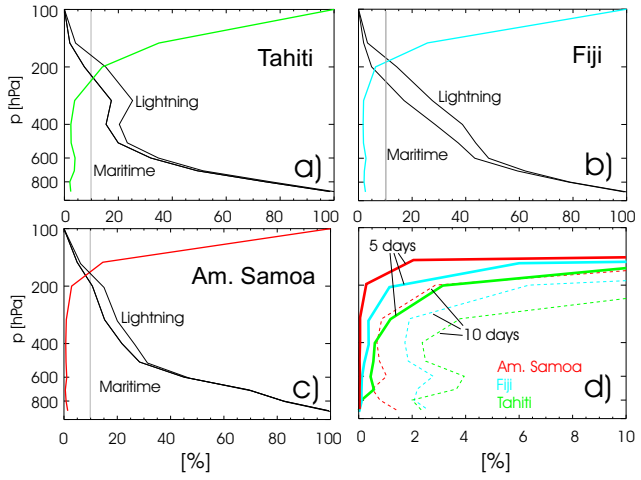


**Fig. 4.** Altitude dependent influence of stratospheric and tropospheric airmasses arriving over Tahiti, Am. Samoa and Fiji. For further information, see the text. In panel (d) the percentage scale is enlarged to underline the differences of the stratospheric influence in the mid and lower troposphere between the three locations. Additionally the stratospheric influence on 5- and on 10-days backtrajectories is shown.

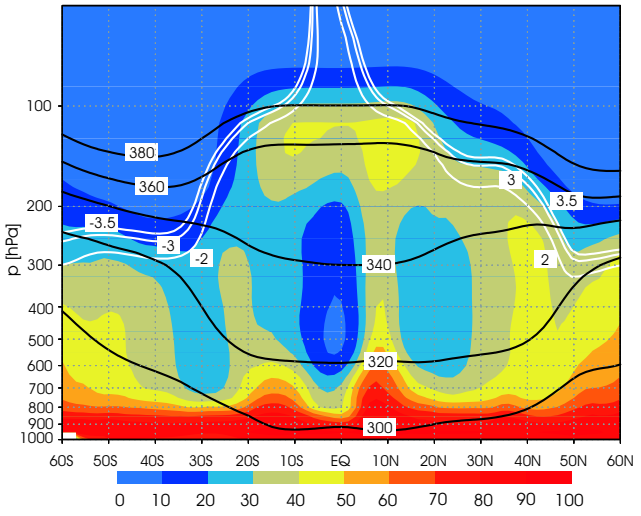


**Fig. 5.** Zonal average for the Pacific region from 160 to 220° E of relative humidity (color scale, given in percent), potential temperature (black lines, given in K) and potential vorticity (white lines, given in PVU) of September 1999. The relative humidity is denoted by the colorscale, the potential temperature by the black lines and the potential vorticity by the white lines. The latter is shown for absolute values of 2, 3 and 3.5PVU which represent different tropopause height definitions. The meteorological data is derived from the 40-years reanalysis (ERA-40) of the European Centre for Medium Range Weather Forecasts (ECMWF).

Whenever a trajectory at least once experienced a PV of more than 3.5 PV-units (PVU) this trajectory was assumed to be of stratospheric origin. The Figs. 4a–d show the altitude dependant fraction of stratospherically influenced trajectories arriving at each of the three locations. At each altitude a set of backtrajectories is released every 6 hours. Whenever the potential vorticity along a backtrajectory exceeds the threshold of 3.5PVU, the trajectory is assumed to be influenced by the stratosphere. In Figs. 4a–d the stratospheric influence of all trajectories arriving at each of the locations is expressed in percent. It is shown that 100% of all trajectories arriving at any of these locations at a height of 100hPa can be regarded as stratospherically influenced. The amount of stratospheric airmasses decays significantly at lower altitudes but at 600 hPa still 1 to 4% of all 10-day-trajectories got into contact with the stratosphere at least once.

In Fig. 4d the differences of the stratospheric influence on short term (5 day) and long term (10 days) backtrajectories arriving above the three locations are shown. Within the last 5 days no trajectories which were influenced by the stratosphere got to the lower troposphere (below 500hPa) above Am. Samoa. 0.25 respectively 0.5% of mid-tropospheric airmasses above Fiji and Tahiti were stratospheric within the recent 5 days. In general the stratospheric influence is highest above Tahiti and significantly lower for Fiji and Am. Samoa.

Additionally the contact of backtrajectories with the boundary layer is evaluated to account for processes emitting trace gases on the ground. Whenever a trajectory gets into the boundary layer it is assumed to be influenced by the ”regime” on the ground. A ”regime” may be the ocean (this is called ”maritime”) or biomass burning. The biomass burning locations for September 1999 are derived from the Along the Track Scanning Radiometer (ATSR) which is also onboard the ERS-2 satellite (Arino, 1997). In Figs. 4a–c no trajectories having experienced biomass burning are indicated. This underlines that biomass burning is of no significance for any of the three locations.

Lightning is a source of nitric oxide (NO) and therefore of nitrogen oxides ($NO_x$) in the troposphere. NO itself oxidizes to nitrogen dioxide ($NO_2$). Therefore, for tropospheric ozone, which is mainly produced by photolysis of $NO_2$, lightning should always be considered. Consequently backtrajectories getting into regions of high lightning activity are assumed to be influenced by lightning. The lightning dataset used for this evaluation is the Low Resolution Annual Climatology (LRAC) provided by the Global Hydrology Resource Center (GHRC) at the Global Hydrology and Climate Center, Huntsville, Alabama. It consists of 5 years of measurements derived from the Lightning Imaging Sensor (LIS) and of 5 years of data from the Optical Transient Detector (OTD). All measurements were merged together into an annual climatological dataset (Christian et al., 2003). For this study the September data of this dataset is applied. Figures 4a–c reveals that the importance of lightning is nearly equal for all three locations (Tahiti, Fiji and Am. Samoa). Consequently

lightning can not be accountable for differences in the ozone loading.

How do airparcels manage to cross the tropopause? Figure 5 denotes the zonally averaged (above the Pacific between 160 and 220° E) relative humidity (in percent), the potential temperature in Kelvin (solid black lines) and the absolute value of the potential vorticity in PVU (solid white lines) for September 1999. A dominant feature of this figure is created by the ascent of air at latitudes of 10° N carrying humidity to high altitudes and the descent of air in the subtropics. Potential vorticity values of 2 to 3.5 and potential temperature values in the range of 380 to 400K are denoted. The PV isolines shown in the graph represent different tropopause height definitions. As long as no diabatic process takes place airmasses are advected on layers of constant potential temperature. Lines of constant potential temperature (denoting the isentropic transport) and constant potential vorticity (denoting the tropopause) intersect at latitudes of around 30° north and south. This implies that stratosphere-troposphere-exchange will mainly take place at those latitudes which is impressively underlined by Fig. 5.

To account for the importance of stratospheric airmasses on the lower or mid troposphere only those trajectories arriving at heights between 800 and 500hPa over Tahiti, Fiji or Am. Samoa are considered. For this trajectory subset a quantity which is referred to as stratospheric trajectory density is derived by projecting all trajectories which currently experience a potential vorticity of more than 3.5PVU independent of their current height onto a 1.125° latitude-longitude-grid on the ground. To better account for the importance of the stratospheric influence the resulting trajectory density in Figs. 6a–c is multiplied with the total percentage of trajectories arriving at 800 to 500hPa (lower troposphere) above the specific location. Figure 6 reveals that trajectories leave the stratosphere at latitudes between 40 and 20 degrees south and dive irreversibly into the troposphere. Concerning the latitude range this is in good agreement with theories of stratosphere-troposphere-exchange. The stratospheric trajectory density is highest for the trajectories arriving in the Tahitian region.

The analysis of trajectories arriving above the three Pacific locations Am. Samoa, Fiji and Tahiti in September 1999 reveals that all these locations were hardly (but nearly equally) influenced by continental outflow (see Fig. 3). Consequently no anthropogenic pollution event can account for the differences of the tropospheric ozone loading over the three locations derived from GOME measurements. For this reason stratosphere-troposphere-exchange is examined as a potential ozone source of this particular episode. Trajectories arriving above each of the three locations are differently influenced by the stratosphere. Airmasses above Tahiti in the Eastern Pacific where GOME data reveal enhanced tropospheric ozone columns are stronger influenced by the stratosphere than those over Fiji and Am. Samoa in the Western Pacific (see Figs. 6a–c). Especially in the lower and mid

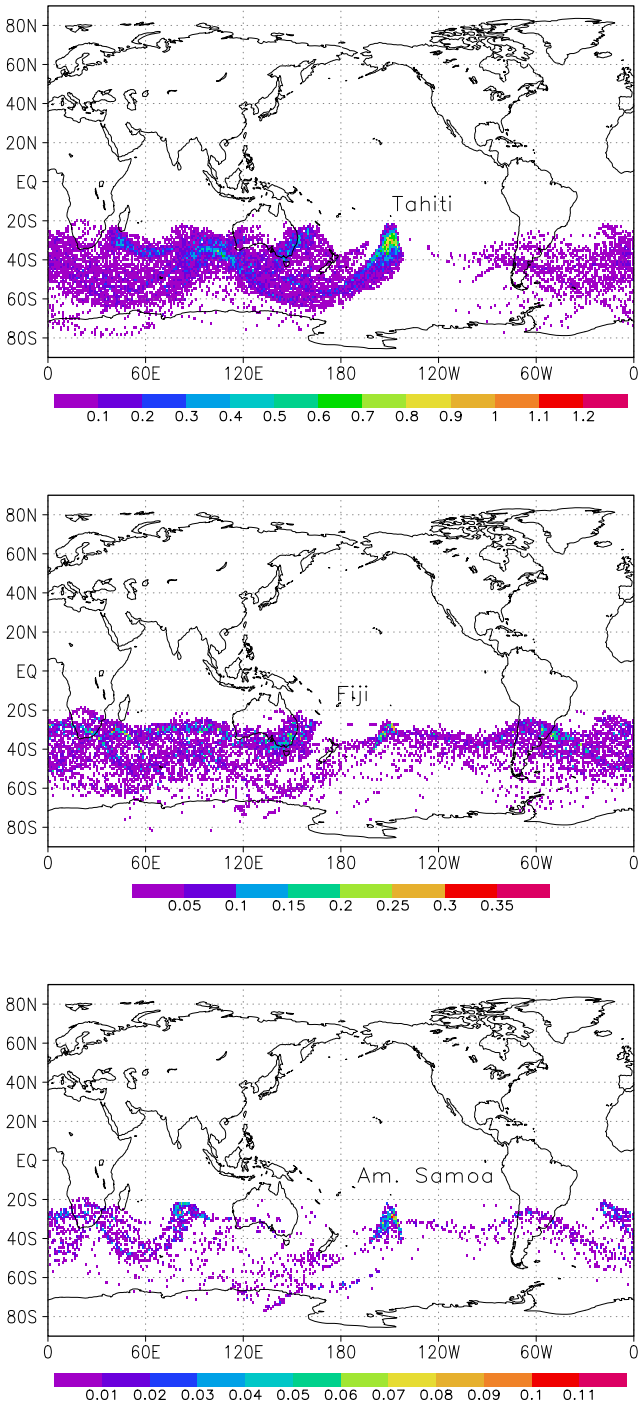


**Fig. 6.** Trajectory density (see its definition in the text) of trajectories arriving at Tahiti, Fiji and Am. Samoa at altitudes of 800 to 500 hPa in September 1999 while being in the stratosphere. It can clearly be seen that airmasses arriving at Tahiti are more subject to stratospheric influence that trajectories arriving at Fiji or Am. Samoa.

troposphere differences of the stratospheric influence (depicted in Figs. 4a–d) between the three locations become obvious. As a result the stratospheric influence on airmasses over a certain location may be used as a qualitative indicator of such episodes. In the remote Pacific such events of stratosphere-troposphere-exchange transporting air with enhanced ozone loading into the lower to mid troposphere can clearly be seen since no local or regional emissions contribute to the tropospheric ozone loading. Nevertheless a trajectory based analysis can only account for qualitative effects. A quantitative analysis of episodes and their quantitative influence on troposperic ozone columns with this approach is not possible.

## 3 Conclusions

Episodes of enhanced tropospheric ozone above Tahiti (Eastern Pacific) compared to Am. Samoa or Fiji (both Western Pacific) occur relatively often (see Fig. 1). This case study shows the influence of stratospheric airmasses on the tropospheric amounts of $O_3$ over the Eastern and Western Pacific region. For this study tropospheric excess ozone was derived from GOME measurements. An increase of up to 12DU over Tahiti (Eastern Pacific) compared to Am. Samoa and Fiji (Western Pacific) was observed during an episode taking place in September 1999 (see Fig. 2). An analysis of backtrajectories for the whole episode reveals that all three locations would have been impacted nearly equally by anthropogenically induced continental outflow. For this reason the influence of stratospheric airmasses on these locations was examined. This analysis reveals that the lower and mid troposphere (LT and MT) over Tahiti is much stronger influenced by stratospheric airmasses than the LT and MT above Fiji or Am. Samoa. The deduction of the stratospheric trajectory density considering backtrajectories arriving in the LT and MT over the three locations underlies that the stratospheric influence is most enhanced in the Tahitian region compared to Fiji or Am. Samoa. Since no major anthropogenic emission of ozone precursors takes place in the remote Pacific in this case study the stratosphere-to-troposphere-exchange of airmasses can be identified as the significant source of enhanced tropospheric ozone above the Eastern Pacific.

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