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Study of Stratosphere-troposphere exchange events of ozone in India and Greece using ozonesonde ascents

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ABSTRACT: Stratosphere-troposphere Exchange (STE) is an important factor controlling the budget of ozone in the upper troposphere and lower stratosphere. Studies of STE events in India are so far restricted to co-ordinated campaigns, and measurements over longer periods are relatively scarce. In the light of this observation, this paper aims to identify the Indian latitudes most likely to be affected by STE, the frequency of occurrence of shallow and deep STE events, the depth up to which the ozone from the stratosphere descends into the troposphere during STE events and the resultant trend of ozone in the troposphere under the possible influence of STE over the 24 years from 1982 to 2006. In addition, a case study of the STE of ozone, which occurred during a cut-off low event at Athens, Greece, is presented in order to understand the parameters that may contribute to the evolution of these events. It is concluded that STE plays a minor role in the Indian tropospheric ozone budget. On the whole, the occurrence of STE events in India is found to increase with increase in latitude and occur more frequently during winter followed by summer. The occurrence of deep STE is higher at high latitudes while the occurrence of shallow STE is higher at low latitudes.

KEY WORDS stratospheric intrusion; ozone; latitudinal effect; seasonal effect

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1. Introduction

Stratosphere–troposphere exchange (STE) is a part of the general circulation of the atmosphere that transports air and atmospheric constituents across the tropopause (Mohanakumar, 2008). Diffusive and convective transports cause exchange of air between the boundary layer and the free troposphere, carrying ozone and its precursors to higher altitudes (Roelofs and Lelieveld, 2000). An important and significant natural source of ozone in the troposphere is downward transport of stratospheric ozone (Holton *et al.*, 1995; Hocking *et al.*, 2007), called STE of ozone and is related to the stratospheric circulation called the Brewer–Dobson circulation. The mechanisms leading to STE from the lowermost stratosphere to the troposphere are tropopause folds, cut-off lows and quasi adiabatic transport along isentropic surfaces (Vaughan, 1988; Holton *et al.*, 1995). The budgets of the annual and global cross-tropopause transport of ozone are highly uncertain (Roelofs and Lelieveld, 2000). Changes in ozone have their largest impact on climate when they occur in the upper troposphere (UT)/lower stratosphere (LS) regions (Forster and Shine, 1997). UT/LS ozone is determined by both transport and chemistry depending on the region and season of the year. The relative contribution of different processes varies strongly across the tropopause. Any significant long-term change in the downward flux of ozone from the stratosphere may thus contribute to long-term LS/UT ozone changes due to STE (Poberaj *et al.*, 2009). Shallow exchange events (events restricted only to the UTLS region) are partially reversible in nature and produce compositional changes in the tropopause region. However, deep STE events (events which influence not only the UTLS region, but also the Earth's surface) are largely irreversible and have a highly significant impact on atmospheric chemistry.

In general, the quantification of the strato-tropospheric ozone exchange is very important for the following reasons. It is well established that tropospheric ozone is normally only 10% of the total ozone content. An increase in tropospheric ozone will have various impacts, such as: (1) adverse effects in human health and plants due to excess in the oxidizing capacity of the troposphere, (2) the enhancement of the greenhouse effect which might be compensated by the cooling stemming from the tropospheric aerosol content (Tzanis and Varotsos, 2008). Conversely, decrease in stratospheric ozone content will be accompanied by: (1) cooling of the stratosphere, and, (2) amplification of the solar ultraviolet radiation reaching the ground (Varotsos *et al.*, 1995; Kondratyev and Varotsos, 1996; Katsambas *et al.*, 1997; Alexandris *et al.*, 1999; Feretis *et al.*, 2002). Thus, a transport of ozone from the stratosphere to the troposphere may result

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in the appearance of some or all of the above mentioned impacts.

It has been observed that background tropospheric ozone levels have increased significantly in India (Mandal *et al*., 2004; Jain *et al*., 2005). Several studies suggest that the ozone decrease in the lower stratosphere driven by changes in stratospheric circulation have contributed to this observed increase in tropospheric ozone. Jenkins *et al*. (2008) provided observational evidence for enhanced LT/UT ozone through convective processes over the tropical Atlantic. Thompson *et al*. (1997) provided observational evidence for the changes in ozone level in the UT by ozone transport from the LS. Thus, determining where and when stratospheric intrusions of ozone significantly affect near-surface concentrations has become important. Roelofs and Lelieveld (2000) have observed that after ozone descends from the stratosphere it mostly resides in the troposphere immediately below the tropopause. Although STE of ozone has been well studied in India, the measurements are mostly restricted to co-ordinated campaigns (Mandal *et al*., 1998) and study over longer periods is relatively scarce. In the light of these observations, the present paper aims to identify the Indian latitudes most likely to be affected by STE, the frequency of occurrence of shallow and deep STE events, the depth up to which the ozone from the stratosphere descends into the troposphere during shallow and deep STE events and the resultant trend of ozone in the troposphere under the possible influence of STE over the period 1982–2006. This will help understand the contribution of tropospheric transport patterns and stratospheric circulation leading to the observed upward trend in tropospheric ozone, which will in turn improve understanding of changes in tropospheric ozone, and is therefore relevant for regional air quality and climate change. Additionally, due to the fact that 'cut-off lows' are among the mechanisms leading to STE from the lowermost stratosphere to the troposphere as mentioned above, a case study of STE of ozone during a cut-off low event over Athens, Greece is presented, in order to understand the parameters that may contribute to the evolution of STE events.

2. Measurement sites, data and analysis

The Indian subcontinent lies between 8°N and 36°N latitude. It is difficult to generalize the climate of India, which comprises a wide range of weather conditions across varied topography and a large geographical area. The India Meteorological Department (IMD) has recognized four official seasons: winter (December, January and February), summer or pre-monsoon (March, April and May), monsoon (June, July and August) and post-monsoon (September, October and November) (Riley and Spolton, 1974; Pant and Rupakumar, 1997). New Delhi (28.4 °N, 77.13 °E) located in northern India in the extra tropics, Pune (18.5 °N, 73.5 °E) in the tropics and Thiruvananthapuram (8.28 °N, 76.56 °E) located in southern India close to the sea in the tropics have entirely different geographical morphology and hence different local climatic conditions (Figure 1).

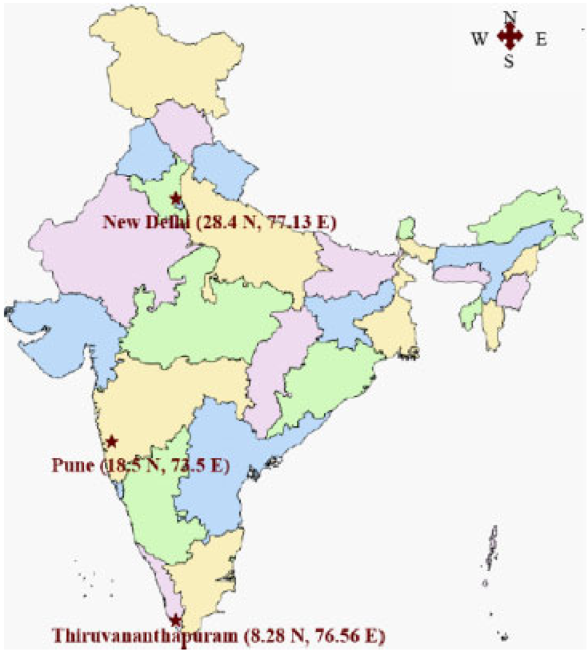

Figure 1. Map of India depicting the measurement sites. This figure is available in colour online at wileyonlinelibrary.com/journal/met

Around 410 vertical ozone and temperature profiles spanning over the period from 1982 to 2006 from ozonesonde and total ozone (Dobson Unit; DU) from the Dobson Spectrophotometer at New Delhi, Pune and Thiruvananthapuram were obtained from the World Ozone and Ultraviolet Radiation Data Centre (WOUDC). These profiles were measured by IMD at all standard pressure levels (1–1000 hPa with accuracy of 0.5 hPa) at intervals of 15 days. Five-day back trajectories at different pressure levels were retrieved from the European Centre for Medium-Range Weather Forecasts (ECMWF). Vertical pressure velocity at the surface and tropopause were retrieved from NCEP/NCAR Reanalysis.

Additionally, a case study of the STE of ozone over Athens, Greece (38°N, 24 °E), during 1997 intensive sounding campaign is presented. The ozonesondes used for the vertical soundings were Electrochemical Concentration Cells (ECC, Komhyr, 1969). The ECC ozonesonde is an ozone measuring device designed to be flown with and on the same balloon as a standard National Weather Service meteorological radiosonde (RS-80, Vaisala). Data were taken during the ascent of the balloon (Totex, 1200 gr). The ascent velocity was about 5 m s^{-1}. Soundings were performed at around 1000 UTC.

An examination of the Indian ozonesonde profiles from 1982 to 2006 and NCEP Reanalysis data from 1948 to 2009 indicated that the tropopause height at New Delhi, Pune and Thiruvananthapuram varied between the 100 and 105 hPa pressure level (around 16.5 km from the Earth's surface). Each ozone profile was grouped into 16 standard pressure levels: 1000, 925, 850, 700, 600, 500, 400, 300, 250, 200, 150, 100, 90, 80, 70 and 50 hPa, which was in turn equated to its equivalent altitude in kilometres. Thereafter, the vertical ozone profiles were divided into three sections. The regions extending approximately 5 km below and above the tropopause were considered as the UT and LS region respectively. The region lying below the UT and extending from the surface to 11 km was termed as middle and lower troposphere (MLT). The columnar ozone partial pressure (mPa) for each of these three regions was determined and the ozone anomalies were identified. The days for which a decrease in stratospheric ozone and a corresponding increase in tropospheric ozone was observed, but the total ozone remained roughly constant (±10 DU) with respect to other days (±5 days) was used for preliminary identification of the STE events.

Thereafter, all the vertical ozone profiles from 1982 to 2006 for each station were grouped into four seasons: summer, monsoon, post monsoon and winter. These profiles were averaged to obtain a normal ozone profile for each season, which was in turn compared with the ozone profiles for STE events in each season, to determine the amount and depth up to which the ozone from the stratosphere descended into the troposphere during STE events (Figure 2). These events were further analysed in detail using back trajectories from the ECMWF at altitudes where enhanced ozone concentration was observed compared to the normal profile to check the possibility of horizontal transport of ozone from the areas surrounding these stations in increasing tropospheric ozone. Since ozone in the troposphere is influenced by the transport of ozone-rich air from high latitudes, the profiles for which the wind direction was from the north, were omitted from this study. Since high insolation and humidity destroys ozone (Roelofs and Lelieveld, 2000), air parcels from high latitudes passing over sea/ocean are not expected to contribute to the observed enhanced ozone levels. The vertical pressure velocity (Pa s^{-1}) at the surface and tropopause from NCEP reanalysis were examined to rule out the possibility of upward transport of surface ozone and its precursors to higher altitudes and simultaneously to confirm the downward transport of ozone from the stratosphere. The STE events, which were restricted only to the UTLS region, were termed as shallow events, while those which influenced not only the UTLS region but also the MLT were termed as deep events. This analysis was further used to identify the season in which maximum STE events occurred, the stations which were most likely to be affected by STE and the frequency of occurrence of shallow and deep STE. To determine the possible long term influence of STE events on the tropospheric ozone variability, the linear trend of total ozone in the MLT region was examined.

3. Discussion of results

It was observed that the frequency of occurrence of STE events was higher during winter, followed by summer (Table I). This may be because during winter, as the air begins to cool, ozone rich air grows denser and sinks to lower altitudes, resulting in a decrease in stratospheric ozone and simultaneous increase in tropospheric ozone. Between October and May, the Intertropical Convergence Zone (ITCZ) is located to the south of Trivandrum (Roelofs and Lelieveld, 2000). Peixoto and Oort (1992) observed that the vertical velocity profiles during winter exhibited a strong ascending motion over the mean position of the ITCZ, and a strong subsidence between 10°N and 30°N latitudes. Annes *et al*. (2001) studied the zonal, meridional and vertical velocity during summer and winter at Gadanki (13.5 °N) in India and observed strong subsidence at Gadanki. They reported that the lower stratospheric vertical motion was downwards during both winter and summer thus affecting the STE processes over India. Moreover, the higher occurrence of STE events during winter is also associated with the seasonality of the global meridional circulation, which peaks in winter, followed by a second maximum during summer associated with the seasonality of the tropopause height (Appenzeller *et al*., 1996). Rosenlof (1995) reported that the mass flux across stratosphere was largest between December and February and smallest between June and August.

Multiple regression analysis was used to determine the nature of the linear relationship between the dependent variable (occurrence of deep and shallow STE of ozone) and independent variable (order of latitude). It was observed that the occurrence of deep STE is higher at high latitudes and the occurrence of shallow STE is higher at low latitudes (Table I). The multiple correlation coefficient 'R' for deep and shallow STE events were 0.897 and 0.964 respectively, indicating a strong relationship between the occurrence of deep and shallow STE of ozone and order of latitude (°N). The coefficient of determination 'R^2' was found to be 0.8043 for deep STE events and 0.9291 for shallow STE events, indicating that 80.43% of the occurrence of deep STE and 92.91% of the occurrence of shallow STE events of ozone were explained by the independent variables i.e. order of the latitude (°N). It is concluded from the regression analysis that the order of latitude is an important factor affecting the occurrence of deep and shallow STE events.

Higher ozone levels as compared to the average of all the profiles for different seasons was observed in the height ranges of 0–11.5 km for deep STE events and 13–16 km for shallow STE events. The amount of ozone from the stratosphere which descended into the troposphere, and the depth up to which it descended, during shallow and deep STE events are mentioned in Table II. The conversion relation is indicated below (Guide to the WMO/GAW Ozone Data, 2007):

$$\text{Ozone partial pressure (mPa)} \times 7.892 = \text{total ozone (DU)}$$

To determine the possible long term influence of STE events on the MLT ozone variability in India, the linear

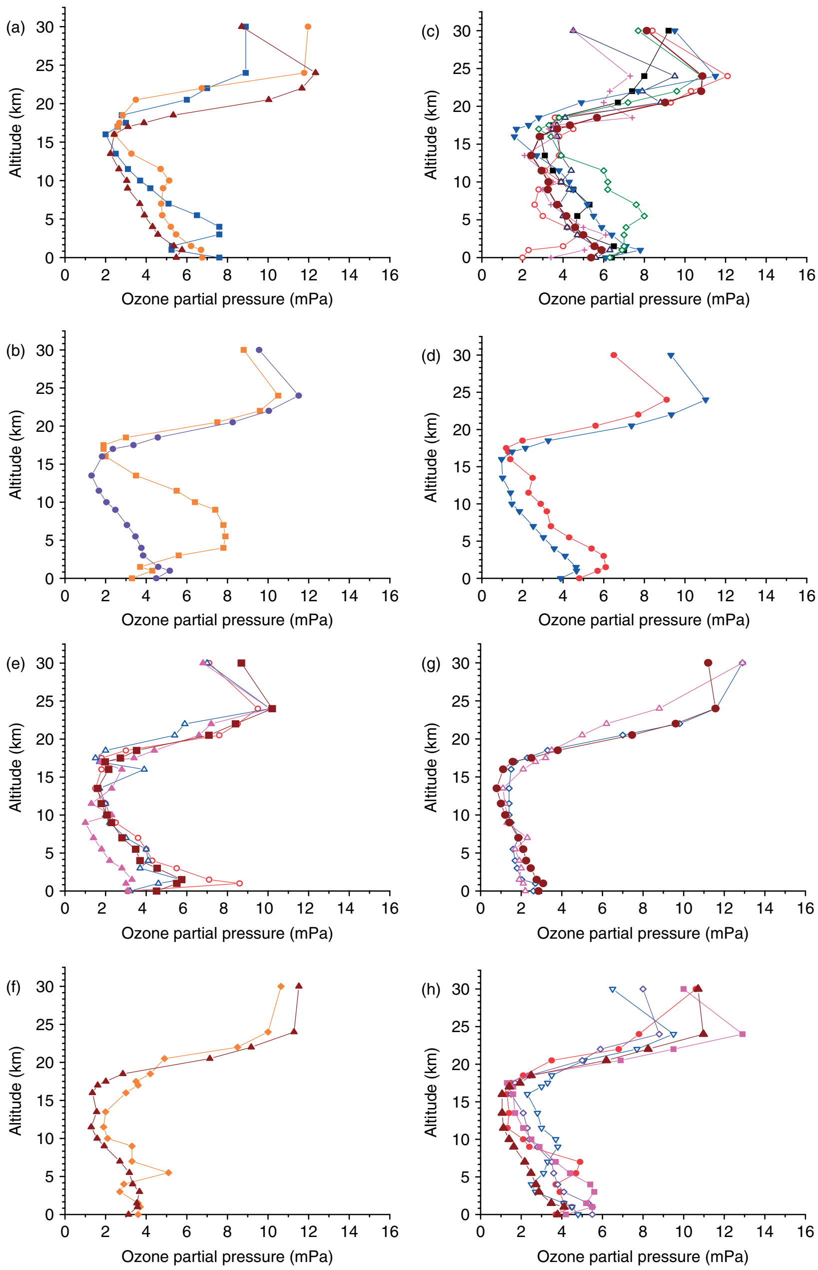

Figure 2. Comparison between stratosphere-troposphere exchange (STE) events of ozone with normal ozone profiles during (a) summer at New Delhi (profile 1 – solid square; profile 2 – solid diamond and normal profile – solid triangle), (b) monsoon at New Delhi (profile 3 – solid square and normal profile – solid diamond), (c) winter at New Delhi (profile 4 – solid square; profile 5 – open circle; profile 6 – open triangle; profile 7 – solid triangle; profile 8 - open diamond; profile 9 – cross and normal profile – solid diamond), (d) summer at Pune (profile 1 – solid diamond and normal profile – solid triangle), (e) winter at Pune (profile 2-open diamond; profile 3 – open triangle; profile 4 – closed triangle and normal profile – solid square), (f) summer at Thiruvananthapuram (profile 1 – solid diamond and normal profile – solid triangle), (g) post monsoon at Thiruvananthapuram (profile 2 – open rectangle; profile 3- open triangle and normal profile – solid circle) and (h) winter at Thiruvananthapuram (profile 4 – solid circle; profile 5 – open triangle; profile 6 – open diamond; profile 7 – solid square and normal profile – solid triangle). This figure is available in colour online at wileyonlinelibrary.com/journal/met

Table I. General features of the STE events covered in this study.

Place	Total no of profiles	Occurrence of shallow STE events (%)	Occurrence of deep STE events (%)	Total no of STE events during summer	Total no of STE events during monsoon	Total no of STE events during post monsoon	Total no of STE events during winter
New Delhi	148	0.68	5.41	2	1	0	6
Pune	94	1.06	3.19	1	0	0	3
Thiruvananthapuram	167	1.2	2.99	1	0	2	4

Table II. Amount of ozone (DU) that descends into the troposphere at the corresponding altitude during STE episodes.

Station	Profile number	Altitude (km) up to which the ozone from the LS descends into the troposphere	Amount of ozone that descends into the troposphere (DU)
New Delhi	1	5	19.7
New Delhi	2	11.5	11.8
New Delhi	3	4	15.8
New Delhi	4	0	15.8
New Delhi	5	13	35.5
New Delhi	6	0	11.83
New Delhi	7	1	11.8
New Delhi	8	1	31.6
New Delhi	9	4	7.9
Pune	1	4	15.8
Pune	2	1	23.7
Pune	3	11.5	15.8
Pune	4	13	7.9
Thiruvananthapuram	1	5	15.8
Thiruvananthapuram	2	13	7.9
Thiruvananthapuram	3	13	7.9
Thiruvananthapuram	4	1	23.7
Thiruvananthapuram	5	0	19.7
Thiruvananthapuram	6	0	15.8
Thiruvananthapuram	7	0	19.7

Table III. Comparison between the trend of total ozone (DU $year^{-1}$) in the MLT at New Delhi, Pune and Thiruvananthapuram from 1982 to 2006.

	MLT (DU $year^{-1}$)
New Delhi	+4.11
Pune	+3.93
Thiruvananthapuram	+1.602

trend of total ozone in the MLT region were examined for the years 1982–2006. The results are summarized in Table III. The observed increasing trend in MLT ozone at all the three stations is in agreement with the observations of Mandal *et al*. (2004) and Jain *et al*. (2005) that the background tropospheric ozone levels have increased significantly in India in the recent years. The standard deviation was observed to be higher at New Delhi compared to Pune and Thiruvananthapuram.

Further analysis of the synoptic situation and history of the relevant air parcels are essential (for instance plots of potential vorticity and cross-sections of potential temperature will have to be examined) to indicate strongly the exchange that occurred. This is intended to be the subject of future work.

4. A case study of STE of ozone over Athens, Greece (38°N, 24°E)

In the following, a case study of the stratosphere-troposphere ozone exchange over Athens, performed at the Athens Ozone Station of the University of Athens during the 1997 intensive sounding campaign, by using ozonesonde ascents is presented. As mentioned earlier, intrusion of stratospheric air into the troposphere occurs in tropopause folding events associated with rapid cyclogenesis. Major tropopause folding events occur at the flanks of large-scale northern flow (Shapiro *et al*., 1987). A three-fold tropopause structure model has been proposed, according to which the sounding station in Athens is situated at such a latitudinal band (36–38°N), where stratospheric air intrusions should occur by means of tropopause folding between the polar and the mid-latitude troposphere. This fact makes the Athens region subject to the influence of the polar front and sometimes to the impact from the subtropical jet stream. Influence of the meteorological situation has been revealed by observations as the laminated structure of the vertical profile of ozone over Athens (Varotsos *et al*., 1994, 2008).

Observations made near a cut-off low in the upper troposphere suggest that a cut-off low may also contribute to the removal of ozone from the stratosphere (Bamber *et al*., 1984). A case study of the stratosphere-troposphere ozone exchange over Athens, Greece, in relation to the presence of a cut-off low in the upper troposphere, which refers to the period from 28 February to 2 March 1997, is presented. The conclusions of this study are in close agreement with previously published results (Varotsos *et al*., 1994, 2008) and are summarized below.

It should be noted here that the potential temperature is used as the vertical coordinate, since it acknowledges the likely motion of air parcels. The potential temperature is the temperature that an unsaturated parcel of dry air would have if brought adiabatically and reversibly from its initial state to a standard pressure (typically

100 kPa). By definition, the potential temperature is conserved under the adiabatic process. Since no heat is added to or subtracted from the air parcel during a dry-adiabatic process, the entropy is, by definition, constant. Additionally, the entropy S is proportional to the logarithm of potential temperature θ ($S = C_p \ln\theta +$ constant, where C_p is specific heat of dry air at constant pressure). Thus, a surface of constant potential temperature in the free atmosphere is also a surface of constant entropy, or an 'isentropic surface'. Under adiabatic conditions air parcels move along isentropic surfaces and this fact is used in isentropic analysis, a form of synoptic analysis which allows visualization of air motions and in particular analysis of large-scale vertical motion. In other words, the use of conserved quantities under adiabatic motion on synoptic timescales, such as that of Ertel's potential vorticity (PV), could provide a very useful tool for the study of the large scale dynamical processes (e.g. McIntyre and Palmer, 1983, 1984; Hoskins *et al.*, 1985; Hoskins, 1991).

The definition of potential vorticity, as introduced by Ertel (1942) for the adiabatic atmosphere, and when expressed in isentropic coordinates with the quasi-static approximation (Lait, 1994), is:

$$\mathrm{PV} = -g(\zeta_\theta + f)\frac{\partial\theta}{\partial P} \quad (1)$$

where g is the acceleration due to gravity, ζ_θ is the vertical component of the relative vorticity on the isentropic surface, f is the Coriolis parameter and θ is the potential temperature.

It has long been recognized that PV can be employed as an approximate material tracer (Hoskins *et al.*, 1985). Furthermore it can be used as the horizontal spatial coordinate replacing thus the conventional geographic coordinates (Norton, 1994; Lary *et al.*, 1995), and reducing the tracer field from three dimensions to two.

Exploiting the characteristic property of PV to obtain absolute values that depend strongly upon height and the meteorological conditions, it is usually normalized by using the PV equivalent latitude (φ_e), as the horizontal co-ordinate, instead of PV itself. Along this line φ_e is calculated by considering the area into a given PV contour on a given θ surface. This calculation is performed by considering that φ_e is the latitude of a latitude circle which encloses equal area as that PV contour. The PV equivalent latitude (φ_e) may be defined as:

$$\varphi_e = \sin^{-1}\left(1 - \frac{A(q)}{2\pi r^2}\right) \quad (2)$$

where $A(q)$ is the area enclosed by the contour of potential vorticity, q, and r is the Earth's radius.

The 'equivalent PV latitude-potential-temperature coordinates' analysis is described in detail in Lary *et al.* (1995).

A cut-off low system (i.e. isolated region of closed geopotential contours in the upper-level flow) developed over Athens, Greece region during the period 28 February to 2 March 1997. As can be clearly seen from Figure 3(a), there exists an increase in ozone partial pressure in the layer between the isentropic surfaces of 300 and 470 K, which is accompanied with an abrupt increase in total ozone content (from 332 DU on 28 February to 405 DU on 1 March and 400 DU on 2 March).

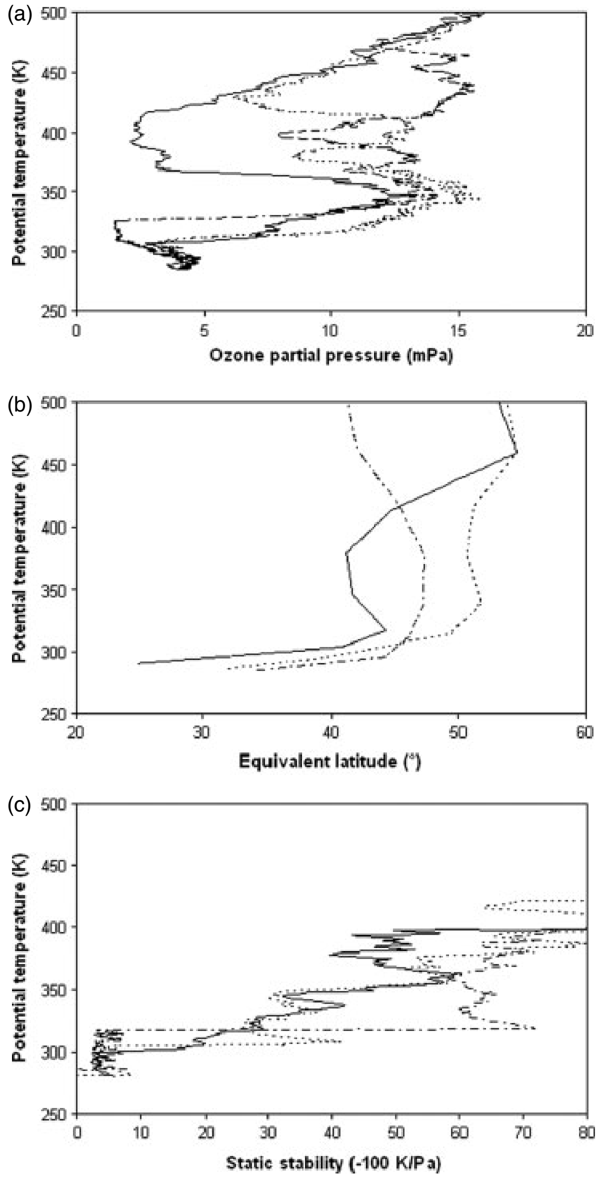

Figure 3. (a) Ozone profiles measured by ECC-type ozonesondes, (b) equivalent latitude analysis and (c) static stability profiles, *versus* potential temperature, at Athens, Greece for 28 February1997 (———), 1 March 1997 (- - - -) and 2 March 1997 (– · –).

The 'equivalent PV latitude-potential-temperature coordinates' analysis shows that the potential vorticity on 1 March is higher than that of 28 February (Figure 3(b)), which leads to the conclusion that the origin of the air masses is from higher latitudes. This is in close agreement with the higher ozone values observed at the layer between the isentropic surfaces of 300 and 450 K. On 2 March the PV equivalent latitude decreases due to the weakening of the cut-of-low event.

The static stability profiles in the layer between the isentropic surfaces of 360 and 400 K (Figure 3(c)), show that the second day it has been increased, while the third day this increase covers the entire layer between the isentropic surfaces of 320 and 400 K. This fact is in close agreement with the observation that cold air masses are transported from northern latitudes.

5. Conclusions

STE plays a minor role in the tropical tropospheric ozone budget. It is observed that the occurrence of deep STE is higher at high latitudes and the occurrence of shallow STE is higher at low latitudes. STE events in India were found to occur more frequently during winter followed by summer. However, since only two vertical ozone profiles could be obtained *per* month for each station in India, it may be possible that the STE events which had occurred on other days could not be identified in this study. Additionally, strong indications of cross-tropopause flow were observed when a cut-off low system was installed over Athens, Greece region, indicating that such features may also contribute to the removal of ozone from the stratosphere.

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