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Influence of Stratospheric Intrusion on Upper Tropospheric Ozone over the Tropical North Atlantic

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Abstract: This study observed the upper tropospheric ozone enhancement in the northern Atlantic for the Aerosols99 campaign in January-February 1999. To find the origin of this air, we have analyzed the horizontal and vertical fields of Isentropic Potential Vorticity (IPV) and Relative Humidity (RH). The arch-shaped IPV is greater than 1.5 pvus indicating stratospheric air stretches equatorward. These arch-shaped regions are connected with regions of RH less than 20%. The vertical fields of IPV and RH show the folding layer penetrating into the upper troposphere. These features support the idea that the upper tropospheric ozone enhancement originated from the stratosphere. Additionally, we have investigated the climatological frequency of stratospheric intrusion over the tropical north Atlantic using IPV and RH. The total frequency between the equator and 30°N over the tropical north Atlantic exhibits a maximum in northern winter. It suggests that the stratospheric intrusion plays an important role in enhancing ozone in the upper troposphere over the tropical north Atlantic in winter and early spring. Although the tropospheric ozone residual method assumed zonally invariant stratospheric ozone, stratospheric zonal ozone variance could be caused by stratospheric intrusions. This implies that stratospheric intrusion influences ozone variance over the Atlantic in boreal winter and spring, and the intrusion is a possible source for the tropical north Atlantic paradox.

Keywords: tropospheric ozone, stratospheric intrusion, isentropic potential vorticity, relative humidity, the tropical north Atlantic

Introduction

Tropospheric ozone plays a key role in atmospheric oxidation, global warming, and air quality change (Brasseur et al., 2003; McKee, 1993). There has been great demand for satellite measurement of tropospheric ozone because of its spatial and temporal coverage. However, as stratospheric ozone accounts for about 90% of the total ozone column located above tropospheric ozone, it makes difficult to detect tropospheric ozone from a satellite (http://www.ozonelayer.noaa.gov).

The first indirect method widely used for tropospheric ozone determination from satellites is the tropospheric ozone residual method (TOR) (Fishman and Larsen, 1997). This method determines stratospheric ozone column from Stratospheric Aerosol and Gas Experiment (SAGE) measurements in conjunction with the thermal tropopause height from weather data. It is then subtracted from Total Ozone Mapping Spectrometer (TOMS). There is a disagreement between tropospheric ozone derived from the residual methods and the precursor CO measured from the "Measurements Of Pollution In The Troposphere" (MOPITT) over the tropical north Atlantic in boreal winter. This has been called the tropical Atlantic paradox (Thompson et al., 2000).

The residual methods allowed the computation of tropical tropospheric ozone under the assumption that tropical stratospheric ozone is invariant with longitude. Therefore, these methods have not considered stratospheric influence on enhanced tropospheric ozone. However, Kim et al. (1996) and Newchurch et al. (2001) suggested that tropical stratospheric ozone varies with longitude. Although Thompson et al. (2003) suggested that the Southern Hemisphere Additional Ozonesondes (SHADOZ) data and the Stratospheric Aerosol and Gas Experiment (SAGE)

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satellite observations do not support longitudinal variance in tropical stratospheric ozone, the spatial and temporal sampling of those observations is inadequate to resolve the issue; therefore, the longitudinal structure of tropical stratospheric ozone is still open to debate. In order to investigate the stratospheric influence on tropospheric ozone enhancement over the northern tropical Atlantic in boreal winter, this study used ozonesonde measurement during the Aerosols99 campaign.

The Aerosols99 ship-board campaign was conducted over the Atlantic in January-February 1999 to understand the distribution of tropical tropospheric ozone. Two interesting features of ozone enhancement occurred in the middle troposphere over the southern Atlantic and in the upper troposphere over the northern Atlantic during the campaign. Many studies focused on elevated ozone in the middle troposphere of the southern Atlantic and suggested that the enhanced amounts are due to tropospheric transport from the biomass burning regions (e.g., South American and African continents) and ozone supplied by lightning NO_x (Thompson et al., 2000; Martin et al., 2002; Edwards et al., 2003; Jenkins et al., 2003). However, none of those studies investigated the upper tropospheric ozone enhancement over the northern Atlantic observed during the Aerosol99 campaign in detail.

A few campaigns (e.g., MOZAIC, TROPOZ II, AEROCE) were conducted to understand the distribution of tropospheric ozone over the Atlantic Ocean and found upper tropospheric ozone enhancement during the occasional campaign in terms of stratosphere-troposphere exchange by using model, tropical cyclone, subtropical jet front system, and wave-breaking events (Gouget et al., 1996; Baray et al., 1999; McCaffery et al., 2004). However, the answers for the relationship between upper tropospheric ozone enhancement and stratospheric intrusion over the Atlantic remain incomplete due to the limited data over the Atlantic.

The main goal of this study is to analyze the role of the stratospheric influence as the cause of upper tropospheric ozone increases over the northern Atlantic during the Aerosols99 campaign and to estimate the climatological frequency of stratospheric intrusion events between the equator and the subtropical region of 30°N. In addition, this study will cover the possible influence of stratospheric dynamics as the cause of the northern Atlantic paradox.

Data

The Aerosols99 trans-Atlantic cruise campaign was initiated to understand tropospheric ozone over the tropical Atlantic during the northern winter. The ship-board measurements were conducted from Norfolk, Virginia (37°N, 76°W) via Cape Town, South Africa (34°S, 22°E) to Port-Louis, Mauritius (22°S, 51°E) around 1200 UTC in January-February 1999 (http://croc.gsfc.nasa.gov/shadoz/). Among a set of 25 ozonesondes during the campaign, this study has found a distinct layer of elevated ozone (>90 ppbv) in the upper troposphere on January 19. In order to analyze the meteorological fields of this event, this study used the European Centre for Medium-Range Weather Forecasts (ECMWF) data with a horizontal resolution of $2.5^\circ \times 2.5^\circ$. The connection between the air mass of stratospheric origin characterized by isentropic potential vorticity (IPV) on the 350 K from ECMWF data based on the following equation (Ertel, 1942);

$$PV = -g\eta_\theta \frac{\partial \theta}{\partial p}$$

where, η_θ is the absolute vorticity on an isentropic surface, θ is the potential temperature, and p is the pressure. The tropopause height is defined as the altitude with the 1.5 PV unit (pvu) as recommended by Hoskins et al. (1985). Additional data to analyze the Stratosphere-Troposphere Exchange (STE) is relative humidity (RH) from ECMWF data.

Analysis

Meteorological fields

During the course of the ship track between 30°N and the equator, there were 11 ozonesonde measurements. One of those Aerosols99 soundings showed a significant ozone enhancement in the upper troposphere on January 19 (Fig. 1). Pollution-related source was not evident from the analysis of CO and aerosol optical thickness reported by Thompson et al. (2000). Figure 1 shows ozone mixing ratios up to 100 ppbv with RH less than 20% and a rapid increase of ozone amounts with extremely dry air above the 100 mb, where the typical tropical tropopause height is located.

In order to track the origin of the enhanced ozone near 200 mb, this study has used the IPV on the 350 K isentropic surface according to Chen's work (1995), which suggested that this value could be used to probe stratospheric intrusions. Various values of IPV (1 pvu $=10^{-6}$ m^2 s^{-1} K kg^{-1}) are used to indicate the separation between stratosphere and troposphere with 2 pvu being a common value (Stohl et al., 2003). This study has used an air mass greater than 1.5 pvu as stratospheric origin. The intersection of the ship track and the A-B cross-section line in Fig. 2 is the location where the measurement occurred on January 19. This Figure shows that the crest of high IPV with 3 pvu, well above 1.5 pvu stretches equatorward and the arch-shaped IPV greater than 1.5 indicating stratospheric air extends toward the northern coastal area of South America.

The connection of the ozone enhancement shown in Fig. 1 to STE has been further analyzed from the vertical structure of IPV. Figure 3 shows the vertical cross section of IPV from A (12.5°N, 67.5°W) across the sounding location (25°N, 55°W) to B (37.5°N, 42.5°W) in Fig. 2. The vertical field of IPV exhibits a tongue like feature extending from the lower stratosphere (~90 mb) to the upper troposphere (250 mb). This feature in the vertical structure of IPV agrees well with the high arch-shaped IPV in Fig. 2. This pattern follows the description of ozone rich-air during STE event by Danielsen (1968).

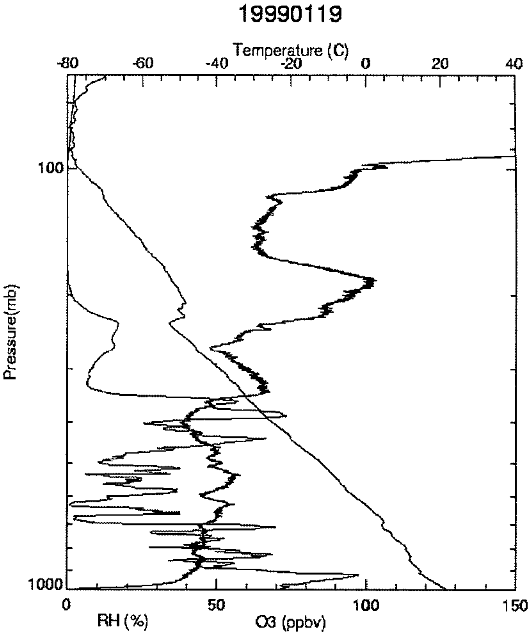

Fig. 1. Profiles of ozone mixing ratio (ppbv), relative humidity (%), and temperature (°C) on the event day (12UTC, January 19, 1999).

In addition, this study has investigated the relationship between IPV and RH fields over the arch-shaped region. Figure 4 shows the relative humidity on the 350 K isentropic surface where IPV was calculated. It shows that the regions with relative humidity less than 20% is strongly connected with the arch-shaped regions of IPV fields shown in Figs. 2 and 3.

This dryness is also found in the vertical cross section of ECMWF relative humidity in Fig. 5. The dry layer with relative humidity less than 20% is part of a meteorological structure that slopes downward toward the upper troposphere. This dry layer is similar to the boundary of stratospheric origin (1.5 pvu) shown in Fig. 3. This evidence supports the conclusion that the upper tropospheric ozone enhancement on January 19, 1999 is associated with a stratospheric intrusion event over the northern Atlantic.

Frequency in 1999

This study shows strong evidence indicating an

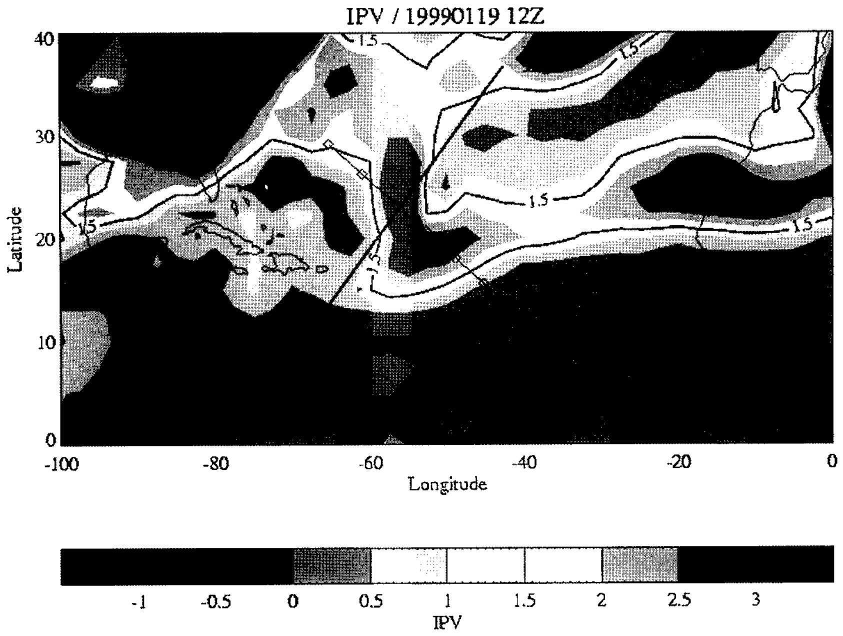

Fig. 2. IPV on the 350 °K isentropic surface at 1200 UTC on January 19, 1999. The shipboard measurement trajectories are indicated by the diamonds. The measurement on January 19 is located at 25°N and 57°W. The contour interval for the IPV is 0.5 pvu (1 pvu=10^{-6} $m^2 s^{-1}$ K kg^{-1}). The thick line represents +1.5 pvu indicating the tropopause. The cross line between A via a measurement position and B is used for the vertical cross sections shown in Fig. 3 and 5.

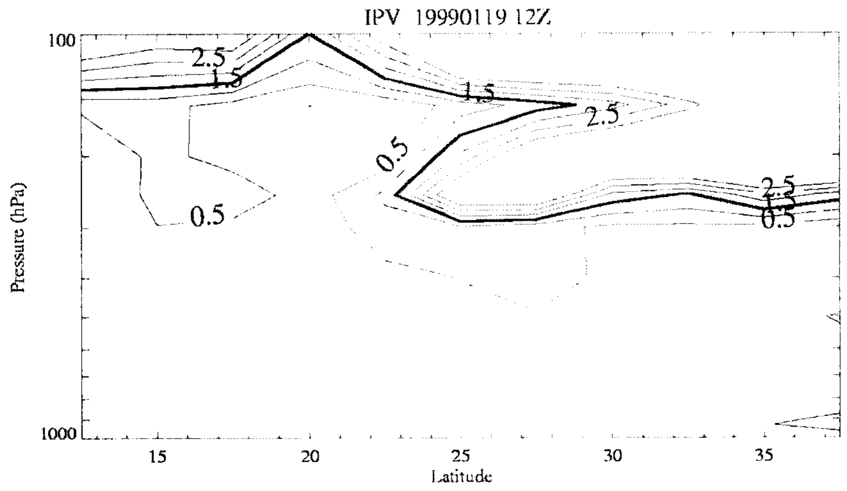

Fig. 3. Vertical cross section of IPV at the cross line from 12.5°N, 67.5°W to 37.5°N, 42.5°W centered at measurement location (25°N, 55°W) (as displayed by the cross line in Fig. 2) on January 19, 1999 at 12 UTC. Thick line represents an indicator of stratospheric air with IPV value 1.5 pvu. The contour interval for the IPV is 0.5 pvu (1 pvu=10^{-6} $m^2 s^{-1}$ K kg^{-1}).

intrusion of stratospheric air into the troposphere over the northern Atlantic. In order to examine the seasonal and latitudinal intensity of this stratospheric intrusion, this study investigated the frequency of stratospheric intrusion over the northern Atlantic with two indicators, IPV and RH. Based on the previous discussion, this study has defined the stratospheric intrusion occurs when the IPV is greater than 1.5 pvu and relative humidity less than 20%. To analyze the frequency, this study selected the region between 40°W and 60°W

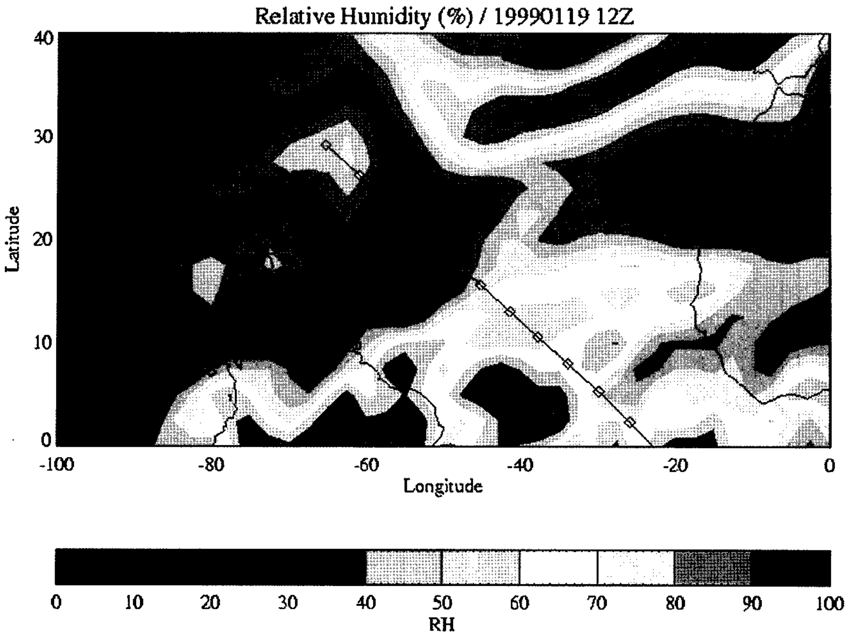

Fig. 4. Relative humidity (%) on the 350 K isentropic surface at 1200 UTC on January 19, 1999. The shipboard measurement trajectories are indicated by the diamonds. The measurement on January 19 is located at 25°N and 57°W. The contour interval for relative humidity is 10%.

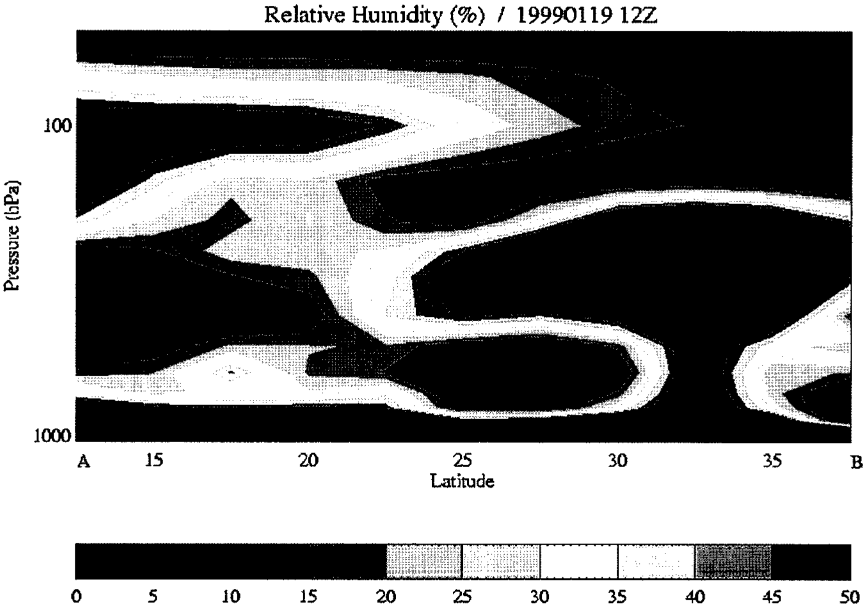

Fig. 5. Vertical cross section of relative humidity (%) at the cross line from 12.5°N, 67.5°W to 37.5°N, 42.5°W centered at measurement location (25°N, 55°W) (as displayed by the cross line in Fig. 2) on January 19, 1999 at 12 UTC. The contour interval for humidity is 5%.

south of 30°N. The frequency is calculated by the number of the events divided by the total number of days, over the analysis region with 5° latitude interval between the equator and 30°N for the year of 1999.

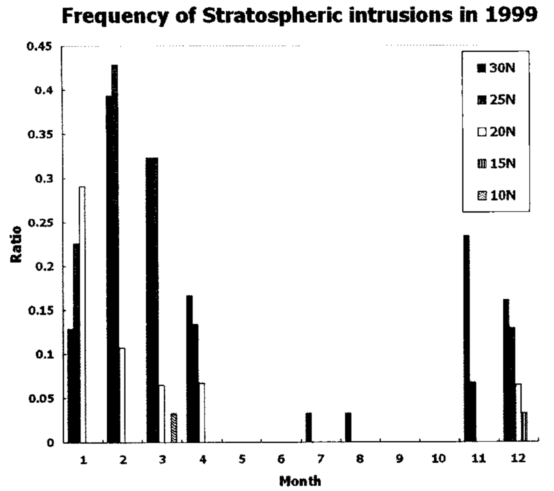

Fig. 6. Frequency of event day that stratospheric air intrusion defined by IPV greater than 1.5 pvu and relative humidity less than 20%. An event satisfying the criteria is counted and divided by the total number of days for each month in 1999. The ratio is calculated in the zonal direction between 40°W and 60°W at 30°N (black box), 25°N (gray box), 20°N (white box), 15°N (striped box), and 10°N (zigzag box).

The histogram in Fig. 6 clearly illustrates that the frequency at 30°N is higher for northern winter and spring and lower for northern summer and autumn. The maximum frequency between 10°N and 30°N is observed from January through March. This seasonality is in agreement with the climatology of intrusion studied by Waugh and Polvani (2000), although they identified intrusion events within 10°S and 10°N. It means that intrusion activities indicating stratospheric influence can propagate in the tropical regions. The maximum occurrence in northern winter appears to be strongly connected to the activities of planetary waves described in Kodera (1995), Perlwitz and Graf (1995), and Kuroda and Kodera (1999). However, a marginal frequency of stratospheric intrusions is observed in December at 15°N and March at 10°N. This study has not observed any noticeable intrusion event south of 10°N. This evidence suggests that stratospheric intrusion over the subtropical northern Atlantic is responsible for ozone variations in the upper troposphere during the northern winter.

Relevance to Atlantic Paradox

The residual methods have subtracted stratospheric ozone measured by various satellites from total ozone measured by TOMS for retrieving tropical tropospheric ozone on the assumption of zonally invariant stratospheric ozone in the tropics (Fishman et al., 1990; 1996; 2003; Hudson and Thompson, 1998; Ziemke et al., 1998; Thompson and Hudson, 1999). An important issue in residual techniques is to understand the cause of the disagreement between the derived ozone and the precursor distributions seen in the fire counts and MOPITT CO over the northern Atlantic in boreal winter and spring. This discrepancy has been called the tropical northern Atlantic Paradox (Thompson et al., 2000). The residual method assumes zonal stratospheric ozone is invariant in the tropics. However, Kim et al. (2005) suggested upper stratospheric disturbance in winter and spring could affect this assumption and result in causing the Northern Atlantic Paradox.

The seasonality of the stratospheric intrusion frequency (Fig. 6) is remarkably similar to the climatological seasonality over the tropical Atlantic observed from Waugh and Polvani (2000). They reported that the stratospheric intrusion intensity over the northern Pacific appears to be stronger than over the northern Atlantic for boreal winter, which is associated with the planetary wave activities. The stratospheric intrusion events influence the ozone distribution by the cross-tropopause transport (Allen et al., 2003). Wang et al. (1998) using SAGE II measurements, reported the ozone column amounts of $4\text{-}5\times10^{17}$ cm^{-2} transported between stratosphere and troposphere over the northern Atlantic.

This evidence suggests the stratospheric intrusion can cause the deviation in the zonal distribution of tropical stratospheric ozone. Therefore, the longitudinal distribution of stratospheric ozone can be affected by stratospheric dynamics due to STE events and possibly result in causing deviation from zonally invariant stratospheric ozone during the dynamically active season of winter and spring.

Conclusions

This study has observed a distinctive ozone enhancement in the upper troposphere of the northern Atlantic from a sounding measurement for the Aerosols99 ship-board campaign conducted for January-February 1999. To find the origin of this air, this study analyzed the horizontal distribution of IPV and relative humidity, as well as the vertical distribution of IPV and relative humidity. The horizontal field in IPV represents the equatorward displacement of air, and the boundary of stratospheric air (1.5 pvu) that penetrates into the subtropical northern Atlantic with dry air (RH<20%). The vertical fields of IPV and RH show the folding layer penetrating into the upper troposphere. These features strongly support the conclusion that the elevated ozone in the upper troposphere originated in the stratosphere.

In order to examine the seasonal and latitudinal intensity of stratospheric intrusions, this study investigated the frequency of stratospheric intrusion over the northern Atlantic with two indicators, IPV and RH. Defining stratospheric intrusion as events with IPV greater than 1.5 pvu and RH less than 20%, we calculated the frequency by the number of the events divided by the total number of days over the analysis region with 5° latitude interval between equator and 30°N for the year of 1999. The highest frequency of the stratospheric intrusion occured in boreal winter and spring, which is appears to be strongly connected to the strongest activity of planetary waves for these seasons. This analysis suggests that stratospheric intrusion plays an important role in enhancing ozone in the upper troposphere over the northern Atlantic in boreal winter and spring.

Although the tropospheric ozone residual methods have assumed zonally invariant stratospheric ozone, stratospheric zonal ozone variance can be caused by stratospheric intrusion events. This study strongly supports the conclusion that stratospheric intrusions influence ozone variance over the Atlantic in boreal winter and spring.

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