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Transport of tropospheric and stratospheric ozone over India: Balloon-borne observations and modeling analysis

P.R. Sinha [a,*,1], L.K. Sahu [b], R.K. Manchanda [a,2], V. Sheel [b], M. Deushi [c], M. Kajino [c], M.G. Schultz [d], N. Nagendra [a], P. Kumar [e], D.B. Trivedi [a], S.K. Koli [a], S.K. Peshin [f], Y.V. Swamy [g], C.G. Tzanis [h], S. Sreenivasan [a]

[a] *Balloon Facility, Tata Institute of Fundamental Research, ECIL Post 5, Hyderabad 500 062, India*
[b] *Physical Research Laboratory (PRL), Navrangpura, Ahmedabad 380 009, India*
[c] *Meteorological Research Institute, Japan Meteorological Agency, Ibaraki 305-0052, Japan*
[d] *Institute for Energy and Climate Research (IEK-8), Forschungszentrum Jülich, Wilhelm-Johnen-Str., Jülich 52425, Germany*
[e] *Space Applications Centre, Indian Space Research Organization, Ahmedabad 380015, India*
[f] *Indian Meteorological Department (IMD), New Delhi 110003, India*
[g] *Indian Institute of Chemical Technology, Hyderabad 500 007, India*
[h] *Faculty of Physics, University of Athens, University Campus Bldg. Phys. V, Athens 15784, Greece*

HIGHLIGHTS

- Balloon-borne in-situ measurement of tropospheric and stratospheric O_3 over India.
- Role of central African and Southeast Asian air masses in tropospheric enhancements of O_3 over India.
- Inter-comparison of vertical O_3 profiles with MACC-II and CCM2 simulation.
- Large contribution of the FT-O_3 to the tropospheric column O_3 (TCO) over both sites of HYD and TVM.

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ABSTRACT

This study describes the spatio-temporal variation of vertical profiles of ozone (O_3) measured by balloon-borne ozonesondes over two tropical sites of Trivandrum (TVM) and Hyderabad (HYD) in India from January 2009 to December 2010. In the lower troposphere, the mixing ratios of O_3 over HYD (18–66 ppbv) were similar to TVM (18–65 ppbv). In the free troposphere, the O_3 mixing ratios over HYD were higher than those over TVM throughout the year. In the tropical tropopause layer (TTL) region (above 15 km), the mixing ratios of O_3 over TVM were higher (83–358 ppbv) compared to those measured over HYD (89–216 ppbv). Prevailing of O_3 laminae between about 14 and 17 km is seen for both sites for most profiles. A strong seasonal variation of O_3 is observed in the lower stratosphere between 18 and 24 km over TVM, however, it is not pronounced for HYD. Transport of air masses from the biomass burning region of the central Africa, Southeast Asia and the Indo Gangetic plains (IGP) influenced and led to enhancements of lower and mid-tropospheric O_3 over HYD and TVM while, the isentropic (325 K) potential vorticity (PV) at 100 hPa showed transport of O_3–rich air from the lower stratosphere to the upper troposphere during winter and spring months over both sites. The free tropospheric O_3 mixing ratios (FT-O_3; 0–4 km) contribute substantially to the tropospheric column O_3 (TCO) with an annual average fraction of 30% and reveal the similar seasonal variations over HYD and TVM. The vertical profiles of O_3 obtained from the Monitoring Atmospheric Composition and Climate - Interim Implementation (MACC-II) reanalysis and the Meteorological Research Institute-Chemistry Climate Model version 2 (MRI-CCM2) are compared with the ozonesonde data over both sites. The

* Corresponding author.
E-mail address: prs@eps.s.u-tokyo.ac.jp (P.R. Sinha).
[1] Now at Department of Earth and Planetary Science, Graduate School of Science, the University of Tokyo, Tokyo 1130033, Japan. Also at National Institute of Polar Research, Tachikawa-shi, Tokyo 190-8518, Japan.
[2] Now at Department of Physics, University of Mumbai, 400 098, India.

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simulated magnitude, phase and vertical gradient of O_3 from both MRI-CCM2 and MACC-II are in good agreement with measurements in the stratosphere while there are significant differences in the tropospheric columns.

1. Introduction

Tropospheric ozone (O_3) is a greenhouse gas and plays an important role in the radiative balance of the Earth atmosphere (Stevenson et al., 2013). It controls the oxidation capacity of the atmosphere through the formation of OH radicals (Brasseur et al., 1998). O_3 and its photochemical derivative OH are the major oxidants as they react with reduced trace gases in the troposphere. On the other hand, O_3 at the surface is a major pollutant because of its detrimental effects on human health and plants (Finlayson-Pitts and Pitts, 2000). Several important environmental problems such as impact on air quality and agricultural crop production have been attributed to high levels of O_3.

Atmospheric dynamics in combination with photochemical processes are the major factors that controls the variability of stratospheric O_3 and former also contributes in the coupling between stratosphere and troposphere (Varotsos et al., 1994, 1999). Therefore, the dynamical processes in the stratosphere do have significant impacts on the O_3 distribution over the tropics. Using long term observations from the Stratospheric Aerosol and Gas Experiment (SAGE) II experiment and ozonesonde data, Randel and Thompson (2011) have shown that inter annual changes in O_3 and temperature are highly correlated throughout the tropical lower stratosphere (16–27 km).

The seasonal and inter annual variations of tropospheric O_3 have also been investigated using the Measurement of Ozone and Water Vapor by Airbus In-Service Aircraft (MOZAIC) data over Chennai (12.99 °N, 80.15 °E), Delhi (28.56 °N, 73.56 °E), and Hyderabad (Sahu et al., 2010a, 2010b; 2014) and highlighted the role of different transport pathways of air masses along with the local meteorological variations. The vertical profiles below 30 km of altitude were also obtained in some studies using balloon soundings over the Indian subcontinent and surrounding oceanic regions (de Laat, 2002; Gupta et al., 2007; Lal et al., 2013, 2014; Mandal et al., 1999; Peshin et al., 2001; Zachariasse et al., 2000). These studies have investigated the layered structure of O_3 in the free troposphere and emphasized the role of long-range transport of air masses and few studies also highlighted the intrusion of stratospheric air into the troposphere. Several recent studies have shown the strong annual and seasonal variation of stratospheric O_3 over the tropics and attributed it to the tropical upwelling (mean circulation in tropical lower stratosphere) (Randel et al., 2007) and quasi-horizontal mixing with the extra-tropics (Abalos et al., 2013; Stolarski et al., 2014). In the present work, we have analyzed two one year records of O_3 mixing ratios up to 30–35 km in the stratosphere for the first time over HYD and TVM. The key objectives of this study is to characterize the temporal and latitudinal variations of O_3 in the troposphere and stratosphere and identify the underlying processes that govern these variations over HYD and TVM. We also compared the ozonesonde profiles with the ones that obtained from the MACC-II reanalysis and MRI-CCM2 simulation for the assessment of consistency between model results with the ozonesode measurements.

2. Study location and measurements

2.1. Site description

The megapolis of Hyderabad (HYD) is the 5th largest city of India with a population of about 7.75 million as per census 2011 (http://censusindia.gov.in/). It is located in southern India (17.47 °N; 78.58 °E; altitude: 545 m; Fig. 1). In recent years, air quality in the city has declined due to rapid population growth, industrial developments and a large increase in traffic. The other site of Trivandrum (TVM) with a population of about 3.31 million as per census 2011 is situated along the coast of Arabian Sea (AS) at the southern tip of peninsular India (8.53 °N, 76.87 °E, altitude: 60 m; Fig. 1). In general, levels of surface O_3 over HYD and TVM are significantly influenced by regional emissions and local meteorological conditions (David and Nair, 2011; Swamy et al., 2012).

The climatic conditions of both sites are controlled by the Indian monsoon system with a dry season during winter (December-February) and spring (March-May) and a wet season during summer (June-September). The fall (October-November) is a transition season with sparse rainfall. Long-range transport carries various precursor gases and aerosol particles over study sites. In general, during winter and spring, the air masses are advected mainly from westerly direction between 5 and 10 km and traversed across intense biomass region of central Africa at 5 and 10 km, while at lower altitude (below 1 km) they are mainly originated from northeast and traveled across Indo-Gangetic plains (Sahu et al., 2014; Sheel et al., 2014). During summer, the

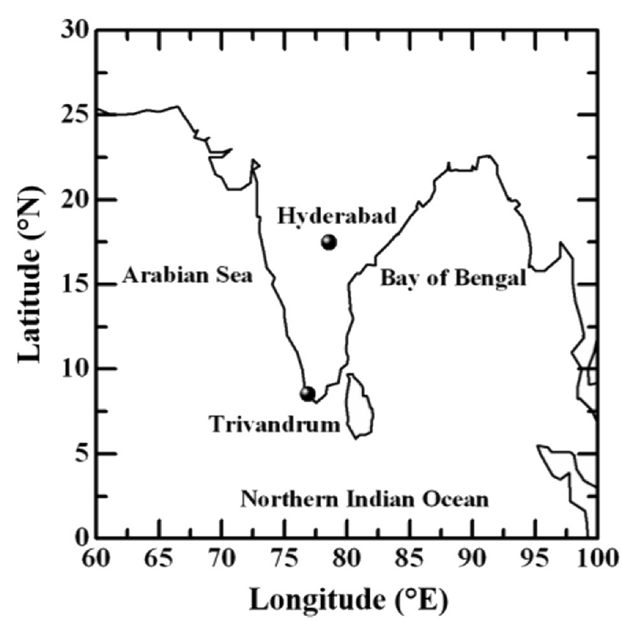

Fig. 1. Terrain of Peninsular India and measurement sites of Hyderabad and Trivandrum.