Vertical distribution of ozone over the Indian Ocean (15°N–20°S) during First Field Phase INDOEX-1998

T. K. Mandal*, D. Kley[†], H. G. J. Smit[†], S. K. Srivastava[‡], S. K. Peshin[‡] and A. P. Mitra^{*§}

National Physical Laboratory, Dr K. S. Krishnan Road, New Delhi 110 012, India

The vertical distribution of ozone over the Indian Ocean has been measured during the First Field Phase (FFP) of Indian Ocean Experiment (INDOEX) from 15°N to 20°S in February and March 1998. Using various ozonesondes during onward (18 February through 13 March) and return (18-28 March) journeys. A latitudinal gradient in ozone mixing ratio from southern (pristine air) to northern (pollutant air) hemisphere is observed in the troposphere. In the northern hemisphere, ozone mixing ratio of the order of 50 ppbv is observed except within the region of 5-8°N, where ozone mixing ratio shoots to 120 ppbv. These large values are likely attributable to continental flow with the advection of photochemically produced ozone. In addition, a layered structure was observed during both onward and return journeys. A pocket of low ozone values (<10 ppbv) is observed simultaneously in the upper troposphere and near the surface. Total ozone maps obtained from total ozone mapping spectrometer-Earth Probe (TOMS-EP) over this region (30°N-30°S, 30°E-150°E) remained around 250 ± 5 DU, except during 26-28 March 1998 when total ozone reached 270 DU. Marine and continental ozone profiles of Indian, East Asian, and South African stations are compared.

TROPOSPHERIC ozone is an important trace gas both because of its role in gas and aqueous-phase chemical reaction as well as its ability to act as a greenhouse gas. Tropospheric concentrations of ozone are influenced by photochemical production/destruction^{1,2}, multiphase processes³, downward transport from the stratosphere⁴ and deposition to surfaces⁵. Ozone is also intimately involved in the cycles of NO_x , OH, and non-methane hydrocarbons. Photodissociation of ozone molecules by solar UV radiation (< 320 nm) produces O¹D radicals which in turn, through its reaction with water vapour, give rise to OH radicals which are the main oxidizing agents. These radi-

cals react with CO, CH₄, light non-methane hydrocarbons, and NO₂ to produce ozone in the free troposphere. Since many of these radicals have both anthropogenic and natural sources, the transport and production/destruction of tropospheric ozone is influenced by anthropogenic emissions⁶. Long-term continental observations both in the northern and southern hemispheres confirmed an increase in tropospheric ozone with a simultaneous decrease of stratospheric ozone.

There is a vast body of information available on vertical ozone profiles measured from ground stations, which are primarily localized in the continental region. However, data concerning the spatial (i.e. vertical and horizontal) distribution in the remote marine atmosphere are still rare. Several reports regarding field observations either by aircraft or balloon-borne ozone soundings over the Atlantic and Pacific Ocean⁷⁻¹² are available. However, no such report regarding measurements of ozone over the Indian Ocean exists.

We report the meridional cross-section of vertical distribution of ozone and humidity measured during onward and return journeys over the Indian Ocean. In addition, we will address the comparative study of marine and continental ozone profiles with an emphasis on the objectives of INDOEX while assessing the role of movement of ITCZ in the formation and transport of trace species over the Indian Ocean and its surroundings during winter dry season.

Field experiments

The ozone soundings were performed on-board the ORV Sagar Kanya from 15°N to 20°S during First Field Phase experiment (INDOEX) in February/March 1998. During onward and return journeys a total of 26 ozonesodes, out of which 15 electrochemical concentration cells (ECC ozonesondes) and 11 modified Brewer Mast (MBM) ozonesondes, were flown on board alternately. We used ECC ozonesondes together with modified RS 80 radiosondes (Vaisala) and prepared according to the detailed

¹Institute for Chemistry of the Polluted Atmosphere, Forschungszentrum Jülich, Germany

[‡]India Meteorological Department, Lodi Road, New Delhi 110 012, India

⁴For correspondence

instrumentation given by Komhyr¹³. With the equipment, we measured simultaneously the ECC current, which corresponds to O₃ partial pressure, the temperature of the inlet air, and with the meteorological RS 80 sonde, atmospheric temperature and relative humidity. However, MBM ozonesondes, which were also flown following the ground preparation¹⁴, gave only O₃ partial pressure and atmospheric temperature. In case of MBM ozonesondes, the humidity sensor was detached due to a technical problem. However, atmospheric temperature as well as

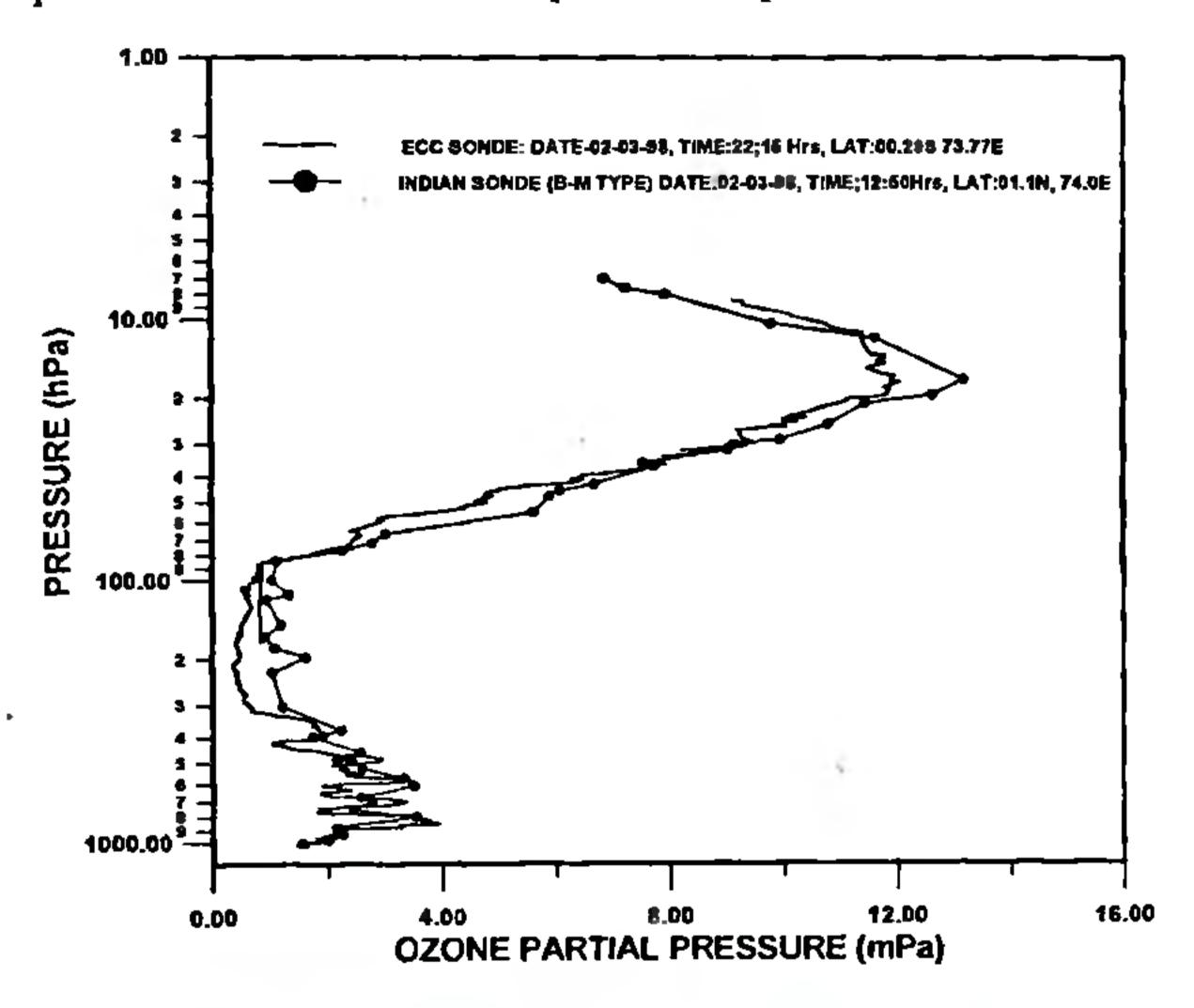


Figure 1. Comparison of two type ozonesondes: electro-chemical cell (ECC) and modified brewer mast (MBM) on 2 March 1998.

humidity data obtained from radiosondes, which were flown at 00 and 12 GMT everyday, appear in the present analysis.

Before launching, the ECC types ozonesondes were intercompared with a ground-based Dasibi ozonemeter. Generally, the normalization factor of MBM, taken from three Indian stations with respect to ground-based Dobson spectrometers remains within 0.8–1.2. The normalization factor of MBM soundings with respect to the column densities (measured by total ozone mapping spectrometer-Earth probe or TOMS-EP typically falls within 0.8-1.2, which gives a margin for the accuracy of our data. However, these data have not been corrected in such way in the present analysis. The quality of ECC and MBM had been assessed through several international intercomparisons¹⁵⁻¹⁹ and found reasonably reliable. Recent JOSIE intercomparison¹⁹ has established a complete picture of the quality of ECC and MBM ozonesondes, and First Field Phase of INDOEX allowed us also to verify the quality of the ECC and MBM ozonesondes. Figure 1 represents the vertical distribution of ECC and MBM ozonesondes flown on 2 March 1998. In spite of the time as well as position differences between the two flights, a coherent pattern is observed between two data sets at all levels.

Results and discussion

Vertical distribution of ozone and relative humidity

Figures 2 a, b and 3 a, b represent the meridional crosssection of the vertical distribution of ozone and relative

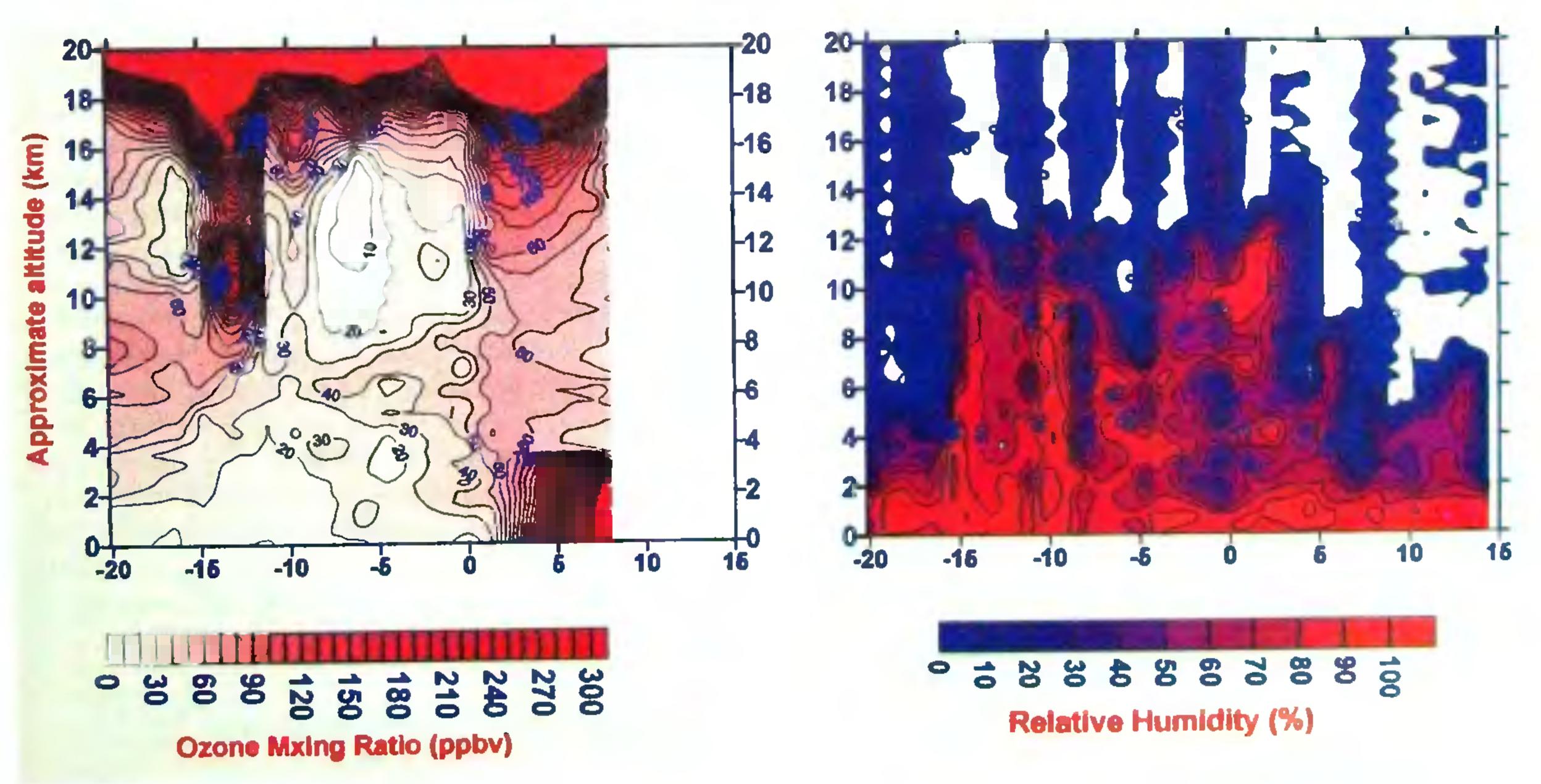


Figure 2. Contour diagram of the ozone mixing ratio (ppbv) and relative humidity (%) in the troposphere, evaluated from the individual soundings performed during the onward journey of first field phase INDOEX-98. The white patch in the relative humidity contour during the onward journey indicates the absence of data.

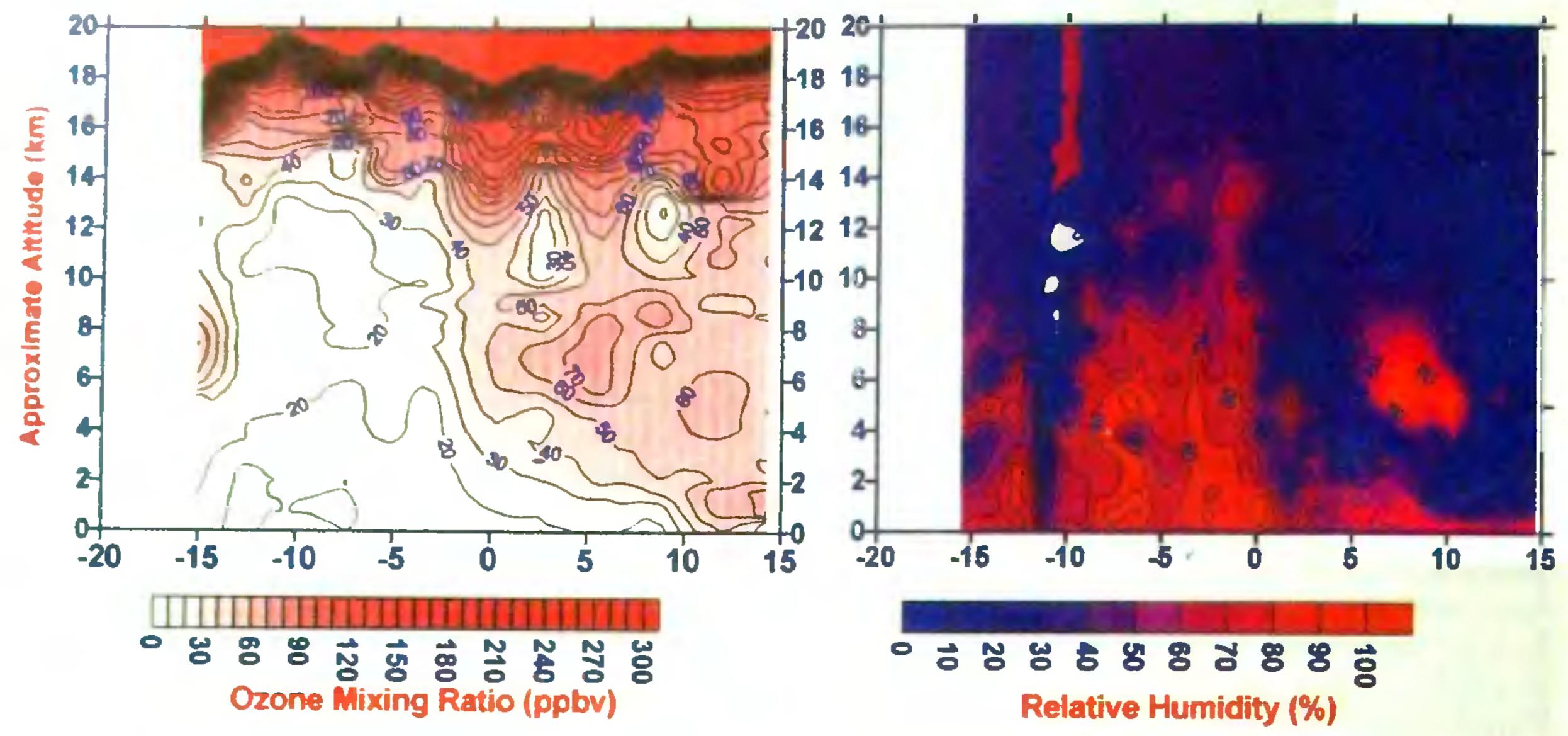


Figure 3. Contour diagram of the ozone mixing ratio (ppbv) and relative humidity (%) in the troposphere, evaluated from the individual soundings performed during the onward journey of first field phase INDOEX-98.

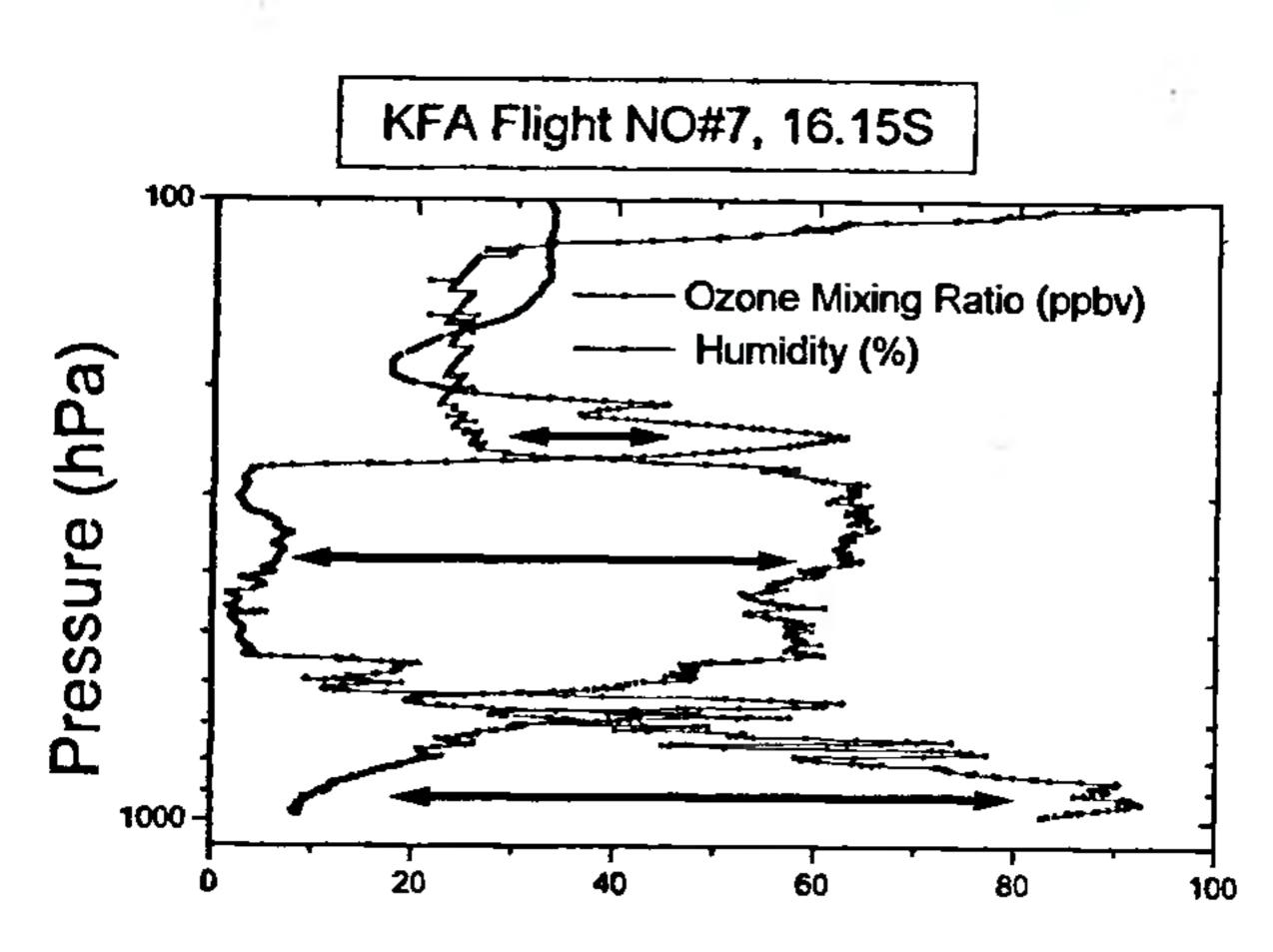


Figure 4. Ozone mixing ratio (ppbv) and relative humidity (%) profile at 16S, launched during the onward journey. The arrows show the anticorrelation between ozone relative humidity.

humidity over the Indian Ocean during onward (18 February to 13 March 1998) and return (18 to 28 March 1998) journeys of First Field Phase of INDOEX respectively. During both parts of the journey the broad feature of ozone meridional cross-section of ozone is quite similar to the structure as observed over the Atlantic and Pacific Oceans⁷⁻¹². These are (i) positive latitudinal gradient from south to north in ozone concentration from near surface to below tropopause, (ii) low ozone value (<10 ppbv) around equator near surface as well as in the upper troposphere, and (iii) several finer structures observed in few profiles. A layered structure of ozone mixing ratio of the order of 20-40 ppbv is observed between 5

and 8 km. Extremely low ozone concentration (1-5 ppbv) near the surface is observed in the boundary layer of the Pacific Ocean^{20,21}. During the onward journey, a low value within the range 4-8°S is observed at the upper troposphere, which is similar to the result of CEPEX. This low ozone concentration is caused by transport of ozone toward the ITCZ in conjunction with photolytic destruction of ozone in the presence of water vapour²². In addition, a large value of the ozone concentration of the order of 120 ppbv is observed near surface in the northern side of equator during the onward journey. This bears the signature of biomass burning²³. However, during this period no large scale biomass burning was noticed during southern side of India (Badrinath, pers. commun.). This may be correlated to the ozone precursors observed near the surface as well with the low humidity (~ 20%) as observed in the relative humidity structure. In addition, during the onward journey, a patch of high ozone value of the order of 60-80 ppbv is observed within the region of 16-20°S. This high value of ozone concentration may signal stratosphere-troposphere exchange. The low relative humidity observed with the particular flight at 16°S (Figure 4), may support this hypothesis. During the return journey, in the southern side of equator less ozone concentration is observed.

Ozone stratification in the troposphere

The presence of ozone laminae has recently attracted considerable interest²⁴⁻²⁷. Reid and Vaughan²⁴, for example, found the laminar structure at lower stratosphere near the polar vortex break during winter and spring. However,

Grant et al.²⁸ discovered that these layers also exist at the tropical and subtropical troposphere.

In addition to these land observations, the same types of layered structure are observed over the Pacific Ocean¹¹. Several finer structures of the order of 50 m are observed within the ozone profiles; however, the layered structure of ozone mixing ratio of 30-60 ppbv at 5-8 km is observed during the onward as well as the return journey during the First Field Phase of INDOEX. Figures 5 a, b represent the latitudinal distribution of ozone mixing ratio and relative humidity (%) averaged between 5 and 8 km. The layer defined by ozone mixing ratio shows a close chemical relationship between ozone and water vapour except within 4-14°S during onward journey. However, Newell et al. 11 have shown that these layers are closely related to other chemical species at that particular levels. Due to absence of observation of other chemical species (CO, CH₄, NO_x) at those particular levels, the chemical and physical relation of these layers observed during INDOEX-FFP-98 is not yet clear.

Reid and Vaughan²⁴ have related the lower stratospheric laminae structure near polar vortex and midlatitude to the exported air from the vortex. Teitelbaum et al.26 found a close correspondence between a stationary class of laminae and lee waves. Pierce and Fairlie²⁹ argued that a large-scale quasi-isentropic motion may lead to the generation of thin filaments of material through chaotic advection. The resulting interweaving of air masses with different trace gas concentrations, coupled with vertical shear, can lead to the formation of laminae in trace gas concentration²⁵. Internal gravity waves³⁰ and Rossby waves²⁸ are correlated to the potential temperature may form laminae structure in ozone profiles. Stoller et al. 30 focused on the chemical signature with conventional ozone chemistry in laminae structure of tropospheric ozone profiles, but due to a nonlinear relation between

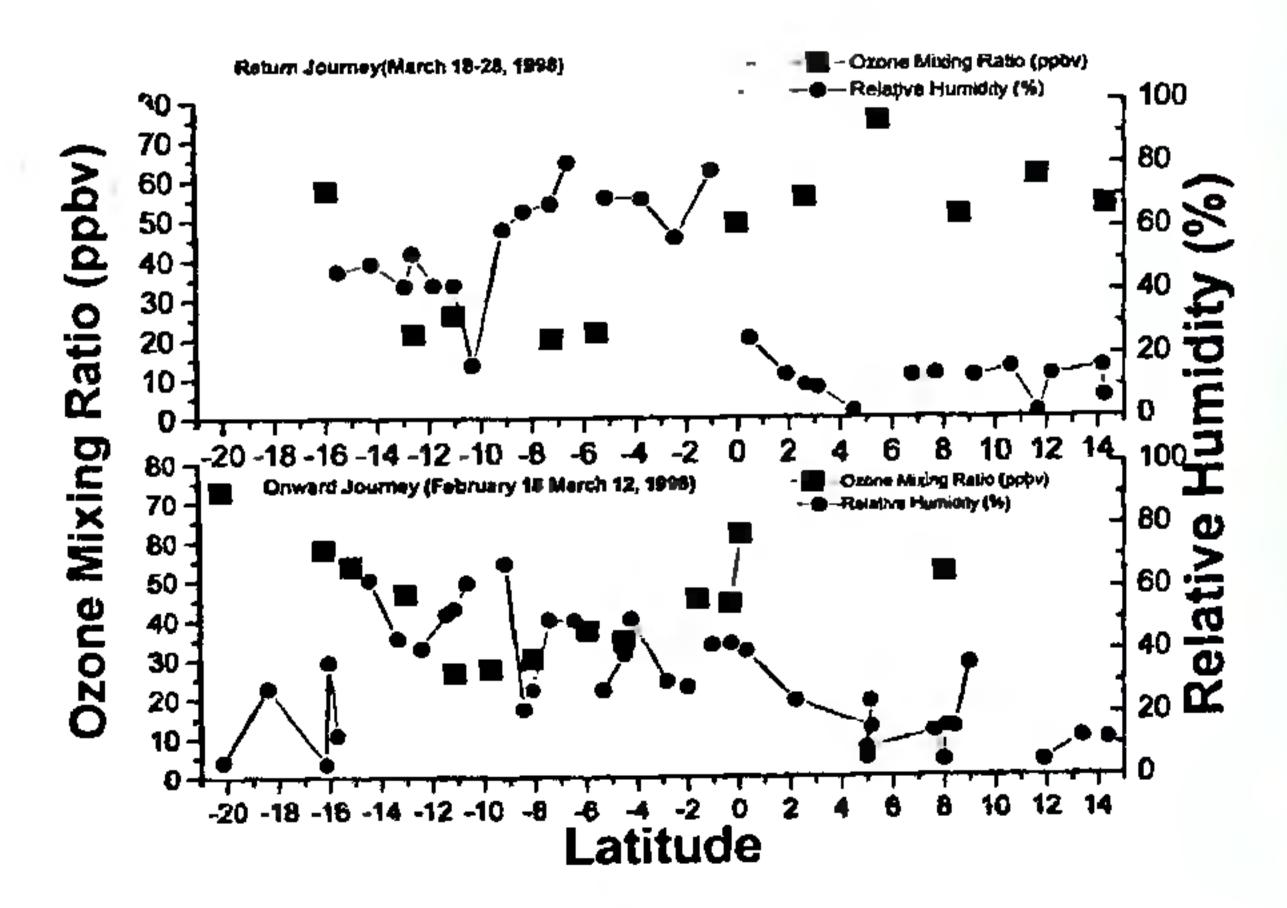


Figure 5 a-b. Latitudinal distribution of ozone mixing ratio (ppbv) and relative humidity (%) averaged between 5 and 8 km during onward (a) and return (b) journey.

the dynamics and chemical processes, analysis is not conclusive.

Ozone column densities

The latitudinal dependence of ozone column densities evaluated from the individual soundings is consistent with the corresponding TOMS-EP data. Total ozone during the cruise period remained around 250 ± 5 DU during the whole journey except during 24-28 March 1998 (Figure 6), when a tongue of high ozone of the order 270 DU is observed over the northern part of equator. It has enhanced total ozone by 20 DU over normal value. During PEM-West A Campaigns, observations indicate the continental influence over north-western Pacific due to easterly motion of wind flow^{31,32}. The enhanced ozone observed here may either be transported from East Asia or may be of photochemical production.

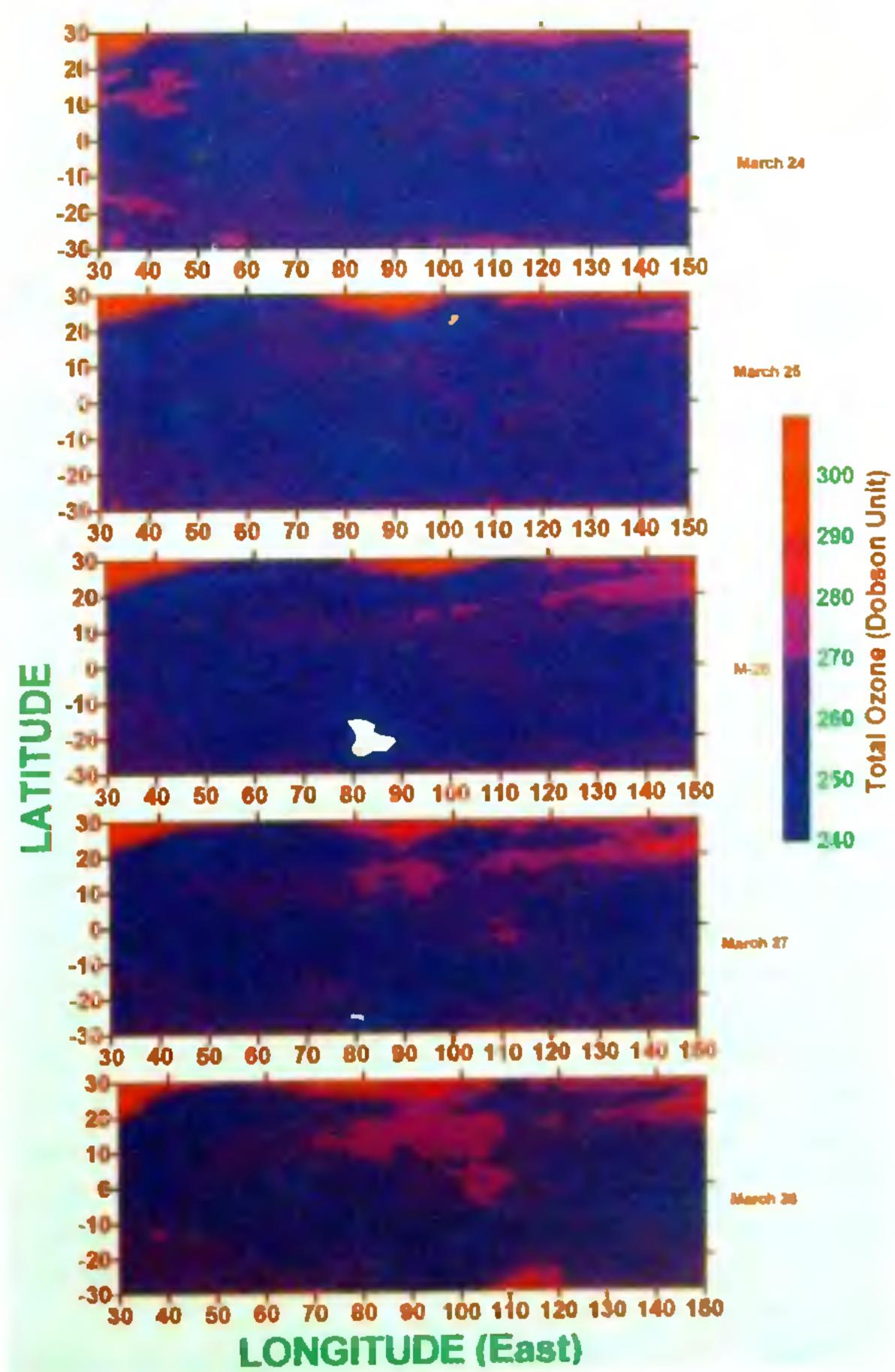


Figure 6. Contour diagram of total ozone (Dobson Units) obtained from Earth-Probe Total ozone mapping spectrometer (TOMS) for the period of 24-28 March 1998.

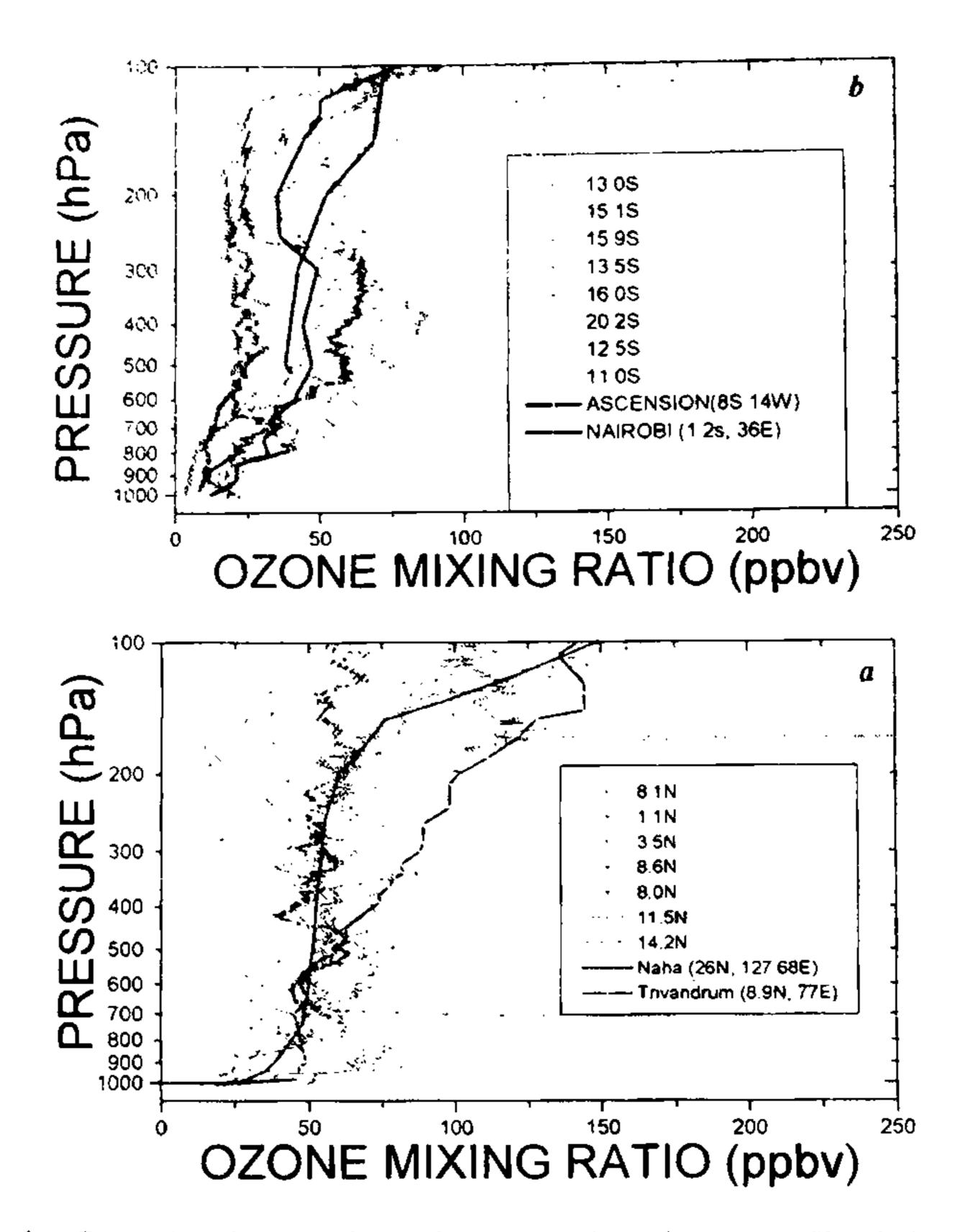


Figure 7 a, b. Comparison between the marine and continental ozone profiles during the onward and return journey. a, Comparison between the marine profiles at the northern side of ITCZ, and continental profiles for the two stations: Naha (26°N, 136°E) and Thiruvananthapuram (8.9°N, 77°E). The continental profiles have been averaged for the period of FFP-INDOEX-98; b, Comparison between the marine profiles at the southern side of ITCZ, and continental profiles for the two stations: Nairobi (1.2°S, 36°W) and Ascension (8°S, 36°E). The continental profiles have been averaged for the period of FFP-INDOEX-98.

Comparison between marine and continental ozone profiles

Figure 7 a represents the comparison between the marine profiles, taken northern side of the ITCZ during the onward and ozone profiles over Naha (26°S, 127.68°E) and Thiruvananthapuram (9°N, 77°E) averaged during the cruise period. The averaged profile over Naha resembles marine ozone profiles. However, ozone profiles over Thiruvananthapuram show higher concentration in the upper troposphere. In addition, the position of ITCZ has varied between 10 and 15°S during onward journey and between 5 and 10°S during the return journey. The high tropospheric ozone observed in the marine ozone profiles at the northern side of ITCZ might have been affected by the continental outflow.

Figure 7 b represents the comparison among the ozone profiles at the southern side of ITCZ and averaged profiles at Ascension (85°S, 14°W) and Nairobi (1.2°S, 36°E). Except for the two profiles, other profiles show

higher concentration than the two continental profiles by 50 ppbv in the middle troposphere. This high ozone may be attributed to the stratosphere-troposphere exchange as low relative humidity represents the stratospheric air.

Conclusion

This paper provides vertical distribution of ozone and relative humidity during the onward and return journey of First Field Phase of INDOEX. Several features emerge:

(1) The positive latitudinal gradient of tropospheric ozone from southern to northern hemisphere is clear during both journeys. A pocket of low ozone value (< 10 ppbv) is observed near the surface as well as at the upper troposphere. In addition, the high tropospheric ozone at the northern side of equator is accompanied by low humidity as well as the continental outflow, as observed from the two continental stations, Naha and Thiruvananthapuram.

- (2) The tongue shape of total ozone observed during 24–28 March has enhanced total ozone by 20 DU, which may be due to either local production of pollutants carried out from East Asian region or sudden drop of humidity within this region.
- (3) Layered structures are observed at 5-8 km during both onward and return journey. These resemble observation over the Pacific and Atlantic Oceans. The layers defined by ozone mixing ratio of the order of 30-60 ppbv show a close relationship between ozone and water vapour. However, other chemical species (CO, CH₄, NO_x) at those particular levels may have contribution in formation of layered structure. Since upward transport of lower tropospheric ozone (< 10 ppbv) may not enrich this high value, these layers may represent the transfer of stratospheric air into the troposphere.
- (4) The comparison between marine and continental ozone profiles suggests that the northern side of ITCZ resembles more the continental profiles as observed over Naha and Thiruvananthapuram, whereas the tropospheric ozone of marine profiles at the southern side of ITCZ has a higher value than the averaged ozone profiles observed at Ascension and Nairobi. The high tropospheric ozone at the middle troposphere may be the signature of stratospheric intrusion as well as of continental flow.

Finally, better meteorological analysis is required to know the source of layered structure at 5-8 km and low ozone value (< 10 ppbv) and the contribution of continental ozone to tropospheric ozone budget over Indian Ocean.

- Chameides, W. L. and Walker, J. C. G., J. Geophys. Res., 1973, 78, 8751-8760.
- Ayers, G. P., Penkett, S. A., Gillet, R. W., Bandy, B., Glabally, I. E., Meyer, C. P., Elsworth, C. M., Bentley, S. T. and Forgan, B. W., Nature, 1992, 360, 446-449.
- 3. Leliveld, J. and Crutzen, P. J., Science, 1990, 264, 1759-1761.
- 4. Danielsen, E. F., J. Atmos. Sci., 1968, 25, 502-518.
- 5. Glabally, I. E. and Roy, C. R., Q. J. R. Meteorol Soc., 1980, 106, 599-620.
- 6. Jacob, D., Logan, J. A., Gardener, G. M., Yevich, R. M., Spivakovsky, C. M. and Wofsy, S. C., J. Geophys. Res., 1993, 98, 14817-14826.
- 7. Smit, H. G. J., Kley, D., McKeen, S., Voltz, A. and Gilge, S., Ozone in the Atmosphere (eds Bojkov, R. D. and Fabian, P.), A. Deepak, Hampton, Va., 1989, pp. 419-422.

- 8. Smit, H. G. J., Gilge, S. and Kley, D., Physico-Chemical Behaviour of Atmosphere Pollutants, CEC, Air Pollution Report 23 (eds Restelli, G. and Angeletti, G.), Kluwer, 1990, pp. 633-637.
- 9. Weller, R. et al., J. Geophys. Res., 1996, 101, 1387-1399.
- 10. Kley, D., Q. J. R. Meteorol. Soc., 1996, 123, 2009-2040.
- 11. Newell, R. E. et al., J. Geophys. Res., 1996, 101, 1943-1960.
- 12. Browell, E. V. et al., J. Geophys. Res., 1996, 101, 1691-1712.
- 13. Komhyr, W. D., NOAA Tech. Memor, ERL, ARL-149, 1986.
- 14. Sreedharan, C. R., J. Phys. E. Sci. Instrum. Sr. 2, 1968, 995-997.
- 15. Attmannspacher, W. and Dutch, H., Berich. Deutsch. Wetter-dienst., 1970, 120.
- 16. Attmannspacher, W. and Dutch, H., Berich. Deutsch. Wetter-dienst., 1981, 157.
- 17. Chatterjee, K., Roy, N. B., Balwali, R. R., Seth, M. M., Chand, N. and Bhattacharyya, P., Indian J. Radio Space Phys., 1986, 15, 147-158.
- 18. World Meteorological Association, Global Atmosphere Watch, Report-27, Geneva, Switzerland, 1994.
- 19. Smit, H. G. J. et al., JOSIE: Proceedings of XVIII Quadrennial Ozone Symposium (eds Bojkov, R. and Visconti, G.), 1998 (in press).
- 20. Routher, F., Dennett, R., Davis, D. D., Wartburg, A., Haagenson, P. and Delnay, A. C., J. Geophys. Res., 1980, 85, 7307-7321.
- Singh, H. B., Gregory, G. L., Anderson, B., Browell, E., Sache, G. W., Davis, D. D., Crafford, J., Bradshaw, J. D., Talbot, R., Blake, D. R., Thornton, D., Newell, R. and Merrill, J., J. Geophys. Res., 101, 1907-1918.
- 22. Kley, D. et al., Science, 1996, 274, 230-233.
- 23. Delany, A. C. et al., J. Geophys. Res., 1985, 90, 2455-2429.
- 24. Reid, S. J. and Vaughan, G., Q. J. R. Meteorol. Soc., 1991, 117, 825-844.
- 25. Orsilini, Y. J., Hansen, G., Hoppe, U. P., Manney, G. and Fricke, K., Q. J. R. Meteorol. Soc., 1995, 121, 1923-1941.
- 26. Teitelbaum, H., Moustaoui, M., Ovarlez, J. and Kelder, H., Tellus, 1996, A48, 83, 442-435.
- 27. Newman, P. A., J. Geophys. Res., 1996, 101, 12879-12892.
- 28. Grant, W. B., Pierce, R. B., Oltmans, S. J. and Browell, E. V., Geophys. Res. Lett., 1998, 25, 1863-1866.
- 29. Pierce, R. B. and Fairlie, T. D., J. Geophys. Res., 1993, 98, 18589-18595.
- 30. Stoller, P. et al., J. Geophys. Res., 1998 (in press).
- 31. Talbot, R. W. et al., J. Geophys. Res., 1996, 101, 1713-1726.
- 32. Gregory, G. L., Anderson, B. E. and Browell, E. V., J. Geophys. Res., 1996, 101, 1919-1930.

ACKNOWLEDGEMENTS. We thank the crew of ORV Sagar Kanya for their help aboard the ship. We also thank Dr U. C. Kulshrestha, Dr K. S. Zalpuri, Dr Prabhat K. Gupta, Mr D. J. Russell and Mr K. Shaikkoya for help during preparation and launching of ozonesondes. We thank the Earth-Probe TOMS data set team for data and the Director, National Physical Laboratory and Director General, India Meteorological Department for the infrastructures for this analysis. ECC ozonesondes were provided by Institute for Chemistry of the Polluted Atmosphere, Forschungszentrum Jülich, Germany and MBM ozonesondes were provided by India Meteorological Department, India.