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SOUTHERN AFRICA: A GIANT NATURAL PHOTOCHEMICAL REACTOR

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1. INTRODUCTION

The notion of a 'large natural photochemical reactor' has been applied to the Mediterranean Basin (Millan *et al.*, 1997), where dynamic and photochemical processes have combined to produce elevated summertime tropospheric ozone levels. Further, Lelieveld *et al.* (2002) have referred to the region as an air pollution crossroads, where pollution pathways from Europe, North America and East Asia intersect, leading to the regional enhancement of ozone.

The analogy of a 'giant natural photochemical reactor' is extended in this paper to the central and southern African tropics, where tropospheric ozone enhancement occurs over a vast geographical area from the Congo to South Africa, and over a long period of time, from June to October. Maximum regional ozone enhancement in September occurs between 10º - 20ºS over Zambia and Zimbabwe, which is designated as the core of the 'photochemical reactor'. Features that contribute towards the 'giant natural photochemical reactor' are abundant sources of ozone precursors (biomass burning, lightning, biogenic and urban-industrial sources), and meteorological conditions that promote anticyclonic recirculation on a sub-hemispheric scale.

2. DATA

Tropospheric ozone data from 10 stations over central and southern Africa that are part of either the SHADOZ (Thompson *et al.,* 2003a,b) network or the MOZAIC (Marenco *et al.,* 1998) data base were used in this study. In addition, limited data (9 profiles in September 2000) from Lusaka, Zambia that were obtained during SAFARI-2000 were used to supplement the main data sets. The location of stations is indicated in Figure 1.

Mean monthly total tropospheric ozone (TTO) values were computed for all stations by integrating ozone amounts between the surface and 12 km. Five-day back and forward trajectory modelling with trajectory origins of 2, 5 and 10 km, was undertaken using the Hysplit model, version 4.7 for each of the stations. Modelled ozone values were generated using the Comprehensive Air Quality Model with extensions (CAMx), version 4.00 (ENVIRON, 2003). Only contributions from urban-industrial activities and biogenic emissions were included in the emissions inventory for the modelling study. Full details of data sources are given by Zunckel *et al.* (2006). It is acknowledged that the omission of biomass burning sources is a significant shortcoming of the modelling study and future studies are planned to address this issue.

3. RESULTS

3.1 Atmospheric Circulation over the African Subcontinent

One of the requirements of the 'giant natural photochemical reactor' notion is large-scale re-circulation of air masses to permit build-up of ozone. This was investigated through back and forward trajectory modelling at each of the stations in the study area (not shown here). The northern boundary of re-circulation occurs at ~10°S. Equatorwards of this latitude, flow is predominantly east-west in accordance with the mean climatology of this equatorial zone where easterly winds prevail for most of the year. The core region of anticyclonic circulation is located between 15-25°S over Zambia, Zimbabwe and the northern parts of South Africa. Back trajectories from Lusaka and Harare indicate a broad easterly sector origin in the lower troposphere (origin at 2 km), but a more westerly and northerly sector origin, suggestive of anticyclonic recirculation, at higher altitudes (origin at 10 km). Further south, for example at Irene, Johannesburg and Maputo, anticyclonic circulation is evident in the lower troposphere (origin at 2 km), but flow is predominantly westerly in the middle to upper troposphere. These results suggest that the anticyclonic recirculation core in the lower troposphere is located at ~25ºS, but that throughout most of the free troposphere the core region of anticyclonic recirculation is located further north (15° - 20°S).

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3.2 Spatial Distribution of Observed Tropospheric Ozone

Spatial plots of mean monthly TTO (Fig. 1) highlight the marked latitudinal gradient and the strong seasonal signal over the African subcontinent. Between January and May, TTO values are below 25 DU over the whole African subcontinent (the month of January is shown in Fig. 1a as an example). In the first part of the year, there is a NW/SE trending tongue of relatively higher TTO values (>20 DU) that extends from Brazzaville to Irene and sometimes includes Maputo. Particularly low values occur over East Africa (Entebbe, Kigali and Nairobi) due to the dominance of maritime south-easterly flow. The strong zonal gradient between Nairobi and Kigali is also noted. The latter station experiences more continental air masses and consequently higher ozone than Nairobi, as noted by Sauvage *et al*. (2004).

An abrupt change in the spatial distribution pattern occurs in June when highest TTO values are located in the west of the region, between 5° and 10°S, peaking over Luanda (> 35 DU) (Fig. 1b). The maximum persists over Luanda for the next two months but tropospheric ozone amounts generally increase over the whole of the study domain with the exception of the north-east quadrant. By September the maximum has strengthened and shifted southwards and eastwards to maximise over Lusaka, where TTO values exceed 40 DU (Fig. 1c). A broad regional enhancement, where TTO values are above 35 DU occurs between latitudes 10° and 20°S. By October the maximum has shifted further south and east, with peak TTO values over Irene and Maputo (Fig. 1d). This south- and east-wards migration of the TTO maximum corresponds closely with the well known latitudinal and seasonal progression of biomass burning over Africa (Cahoon *et al.,*1992; Swap *et al.,* 2003) and is significant endorsement for the dominant role played by biomass burning over the continent.

In November and December, the pattern of the first part of the year is replicated, with one important exception, namely that the Irene TTO values remain relatively high. The possible influence of urban-industrial effects as an explanation for the relatively elevated tropospheric ozone values in summer and

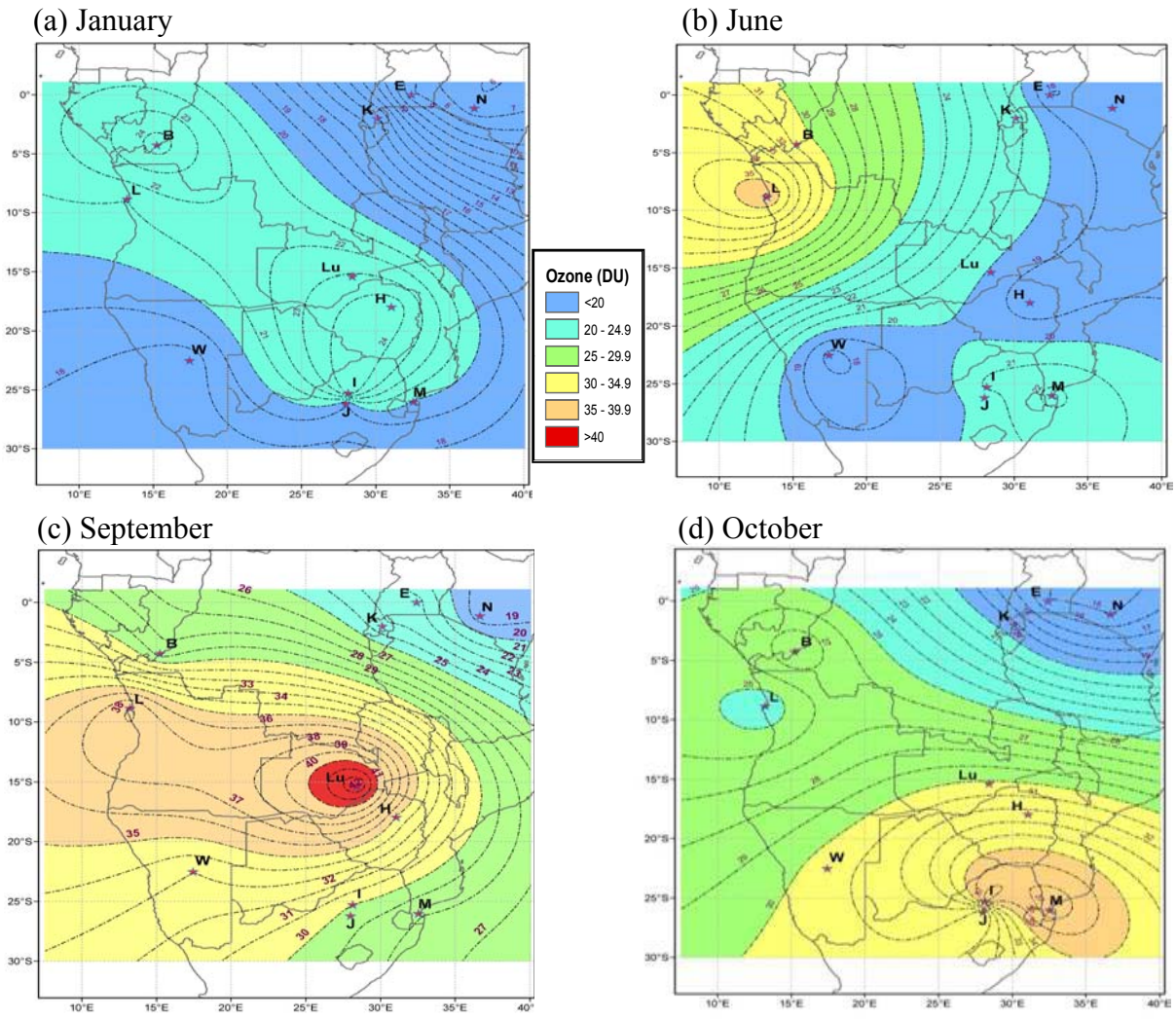

Figure 1: Spatial plots of observed monthly total tropospheric ozone (TTO) in Dobson Units (DU) (surface to 12 km) for January, June, September and October. Stations are designated as follows: E-Entebbe; N-Nairobi; K-Kigali; B-Brazzaville; L-Luanda; Lu-Lusaka; H-Harare; W-Windhoek; I-Irene; M-Maputo; J-Johannesburg

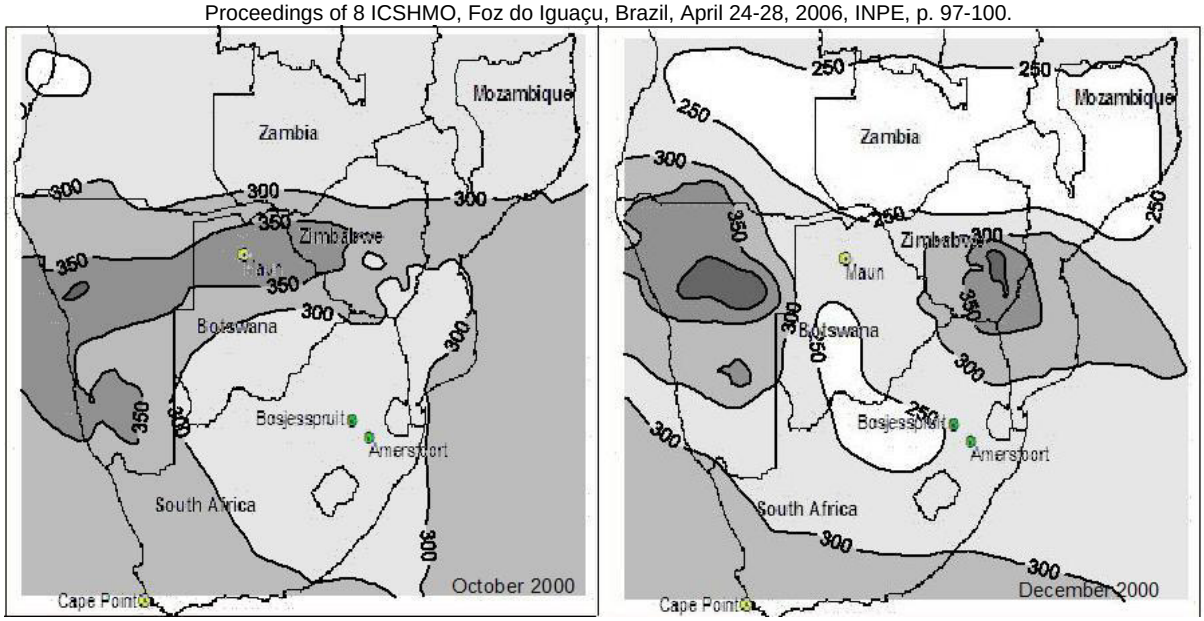

Figure 2: Spatial plots of modelled monthly ozone (ppb) integrated over the surface to 4 km layer for October and December 2000 using the CAM_x model

consequently the lower seasonal range at Irene has been noted previously (Diab *et al.,* 2004).

3.3 Spatial Distribution of Modelled Tropospheric Ozone

Average modelled total ozone concentrations integrated over the lowest 4 km of the troposphere range between 250 and 400 ppb over southern Africa in October and December 2000 (Fig. 2). The maximum lower tropospheric concentrations occur in both months in a zonal band extending from west of Namibia, over Botswana, Zimbabwe and Mozambique. The band of maximum concentration is well defined in October (spring) and appears to be associated with the northern limb of the semi-permanent anticyclonic gyre (Garstang *et al,* 1996). In summer the band of maximum concentration is broken over Botswana with the weakening of the anticyclone and the establishment of a summer surface trough. Average modelled surface ozone concentration (integrated up to 70 m) in October and December 2000 ranges between 30 and 40 ppb (Zunckel *et al*, 2005), with maxima corresponding roughly with those seen for the lower troposphere over Namibia and Zimbabwe. As may be expected, the bulk of the ozone in this lower 4 km of the troposphere resides above the immediate surface layer.

4. CONCLUSION

Observed and modelled tropospheric ozone amounts are shown to maximise in spring over a broad region to the north of South Africa, predominantly Zimbabwe and Zambia. This region (15-25°S) coincides with the core of the anticyclonic recirculation and is furthermore known to be an area with a high incidence of biomass burning and biofuel use. Future work will focus on the inclusion of these sources into the model simulations.

REFERENCES

Cahoon, D.R., *et al.,* 1992: Seasonal distribution of African savanna fires. *Nature,* 359, 812 – 815.

Diab *et al*., 2004: Tropospheric ozone climatology over Irene, South Africa from1990-1994 and 1998-2001. *J. Geophys. Res.,* 109, D20301, doi: 10-1029/2004JD004793.

ENVIRON, 2003: User's Guide to the Comprehensive Air Quality Model with Extensions (CAM_x), ENVIRON International Corporation, 101 Rowland Way, Novato, CA 94945.

Garstang, M., *et al*., 1996: Horizontal and vertical transport of air over Southern Africa, *J. Geophys. Res.,* 101 (D19), 23721-23736.

Lelieveld, J., *et al.,* 2002: Global air pollution crossroads over the Mediterranean. *Science*, 298, 794-799.

Marenco, A., *et al.,* 1998: Measurement of ozone and water vapor by Airbus in-service aircraft: The MOZAIC airborne program, an overview. *J. Geophys. Res*., 103 (D19), 25631 – 25642.

Millan, M.M., *et al.,* 1997, Photooxidant dynamics in the Mediterranean basin during summer:

Results from European research projects. *J. Geophys. Res.*, 102 (D7), 8811 – 8823.

Sauvage, B., *et al.*, 2004: Tropospheric ozone over equatorial Africa: regional aspects from the MOZAIC data. *Atmos. Chem. Phys.*, 4, 3285-3332.

Swap. R.J., *et al.,* 2003: Africa burning: a thematic analysis of the Southern African regional Science Initiative (SAFARI 2000). *J. Geophys. Res., 108* (D13), 8465, doi:10.1029/2003JD003747.

Thompson, A.M., *et al.,* 2003a: Southern Hemisphere Additional Ozonesondes (SHADOZ) 1998-2000 tropical ozone climatology, 1, Comparison with Total Ozone Mapping Spectrometer (TOMS) and ground-based measurements, *J. Geophys. Res*., *108 (D2)*, 8238:doi:10.1029/2001JD000967.

Thompson, A.M., *et al.,* 2003b: Southern Hemisphere Additional Ozonesondes (SHADOZ) 1998-2000 tropical ozone climatology, 2, Tropospheric variability and the zonal wave-one, *J. Geophys. Res*. *108 (D2)*, 8241, doi:10.1029/2002JD002241.

Zunckel, M., *et al.,* 2006, Modelled surface ozone over southern Africa during the Cross Border Air Pollution Impact Assessment Project, J. Env. Modelling and Software, in press.