Available online at www.sciencedirect.com
SCIENCE DIRECT
Environmental Modelling & Software 21 (2006) 911–924
Environmental Modelling & Software
www.elsevier.com/locate/envsoft

Modelled surface ozone over southern Africa during the Cross Border Air Pollution Impact Assessment Project

M. Zunckel [a,*], A. Koosailee [a,f], G. Yarwood [b], G. Maure [c], K. Venjonoka [d], A.M. van Tienhoven [a], L. Otter [e]

[a] CSIR Environmentek, P.O. Box 17001, Congella 4013, Durban, South Africa
[b] ENVIRON International Corporation, 101 Rowland Way, Novato, CA 94945, USA
[c] Department of Physics, Eduardo Mondlane University, Box 257, Maputo, Mozambique
[d] Ministry of Environment, Wildlife and Tourism, Private Bag BR132, Gaborone, Botswana
[e] Climatology Research Group, University of the Witwatersrand, Private Bag 3, Wits 2050, South Africa
[f] Sasol Synfuels, Private Bag X1000, Secunda 2302, South Africa

Received 2 June 2004; received in revised form 31 March 2005; accepted 11 April 2005
Available online 24 June 2005

Abstract

Monitoring of surface ozone over southern Africa has shown that ambient concentrations often exceed a threshold of 40 ppb at which damage to vegetation by ozone could be expected. The Cross Border Air Pollution Assessment Project (CAPIA) was therefore established to assess the potential impacts of ozone on maize, a staple food crop, in five southern African countries. Measured surface ozone data are scare in the region so it was necessary to complement the monitoring with regional-scale photochemical modelling to achieve the objective. The Pennsylvania State and NCAR Mesoscale Model (MM5) is used to produce gridded meteorological data for 5 days in each month of the maize growing season, October to April, as input to the photochemical model, CAMx. Gridded anthropogenic emissions from industry, transport and domestic burning and gridded biogenic emissions from soils and vegetation are input to CAMx. The model estimations indicate large areas on the sub-continent where surface ozone concentrations exceed 40 ppb for up to 10 h per day. Maximum concentrations may exceed 80 ppb, particularly in the winter when mean ozone concentrations are higher. The areas where the 40 ppb threshold is exceeded coincide with maize growing areas in South Africa and Zimbabwe. It appears that neither anthropogenic emissions nor biogenic emissions are dominant in the production of surface ozone over southern Africa. Rather the formation of surface ozone over the region is attributed to the combined contribution of precursors from anthropogenic and biogenic origin.

Keywords: Photochemical modelling; CAMx; MM5; CAPIA; Botswana; South Africa; Mozambique; Zambia; Zimbabwe; Maize; Anthropogenic emissions; Biogenic emissions

1. Introduction

Southern Africa is a region of abundant sunshine, significant sources of ozone precursors and a dominant anticyclonic climatology that suppresses vertical mixing and favours the accumulation of pollutants. These conditions are favourable to the formation of ozone and suggest that ozone concentrations over southern Africa may be relatively high. Ozone is an important constituent in tropospheric chemistry (Jenkins and Clemitshaw, 2000). It is also associated with impacts to human health (Lippman, 1989), vegetation (Emberson et al., 2001; van Tienhoven and Scholes, 2003) and materials (Lee et al., 1996). Even so, measurements of

* Corresponding author. Tel.: +27 31 261 8161; fax: +27 31 261 2509.
E-mail address: mzunckel@csir.co.za (M. Zunckel).

1364-8152/$ - see front matter © 2005 Elsevier Ltd. All rights reserved.
doi:10.1016/j.envsoft.2005.04.004

ozone in the lower troposphere and at ground level are limited to a few campaign studies and at a limited number of monitoring sites in southern Africa.

In South Africa, surface ozone measurements have been made at Cape Point (Fig. 1) at the Global Atmosphere Watch (GAW) site since 1983 (Brunke and Scheel, 1998). Background concentrations typically vary between 15 ppb in summer and 30 ppb in winter with an annual average of approximately 22 ppb. Surface ozone is also monitored at a number of sites in the industrialised northeastern parts of the country (Annegarn et al., 1996) and monitoring is on going at Maun in Botswana where concentrations of 90 ppb and higher are not uncommon (Zunckel et al., 2004). During the period 1991–1993 a network of 20 monitoring stations was in operation in the eastern highlands of Zimbabwe (Jonnalagadda et al., 2001), where the mean annual surface ozone concentrations ranged between 37 and 49 ppb. Meixner and Helas (1994) measured ozone at Victoria Falls, Zimbabwe during the 1992 Southern Africa Fire Atmosphere Research Initiative (SAFARI-92) campaign. Surface ozone concentrations near the surface measured using tethersonde soundings during SAFARI-92 ranged between 40 and 70 ppb at Etosha, Namibia and 30–50 ppb over Irene, South Africa (Diab et al., 1996a,b). Surface ozone is also monitored at five stations as a component of the IGAC (International Global Atmosphere Chemistry) DEBITS (Deposition of Biogeochemically Important Trace Species) programme at background stations in Namibia and in South Africa and on the industrialised Mpumalanga highveld in South Africa (Zunckel et al., 2004). Kirkman et al. (2000) present an overview of measured ozone in the lower troposphere over southern Africa.

The objective of the Cross Border Air Pollution Impact Assessment Project (CAPIA) is to assess the possible impact of ozone on maize in five countries in southern Africa, namely Botswana, Mozambique, South Africa, Zimbabwe and Zambia (Fig. 1). The monitoring data are however too sparse in the region and do not immediately correspond to the areas where maize is grown (Zunckel et al., 2004; van Tienhoven et al., submitted for publication). It is necessary therefore to apply a photochemical transport model to estimate surface ozone concentrations over the region.

Maize is most susceptible to damage by ozone during the flowering season (van Tienhoven et al., submitted for publication), which varies temporally across the region with climate and with maize variety. In the Cross Border Air Pollution Impact Assessment Project (CAPIA, 2002), van Tienhoven et al. (submitted for publication) assessed the potential risk of damage to

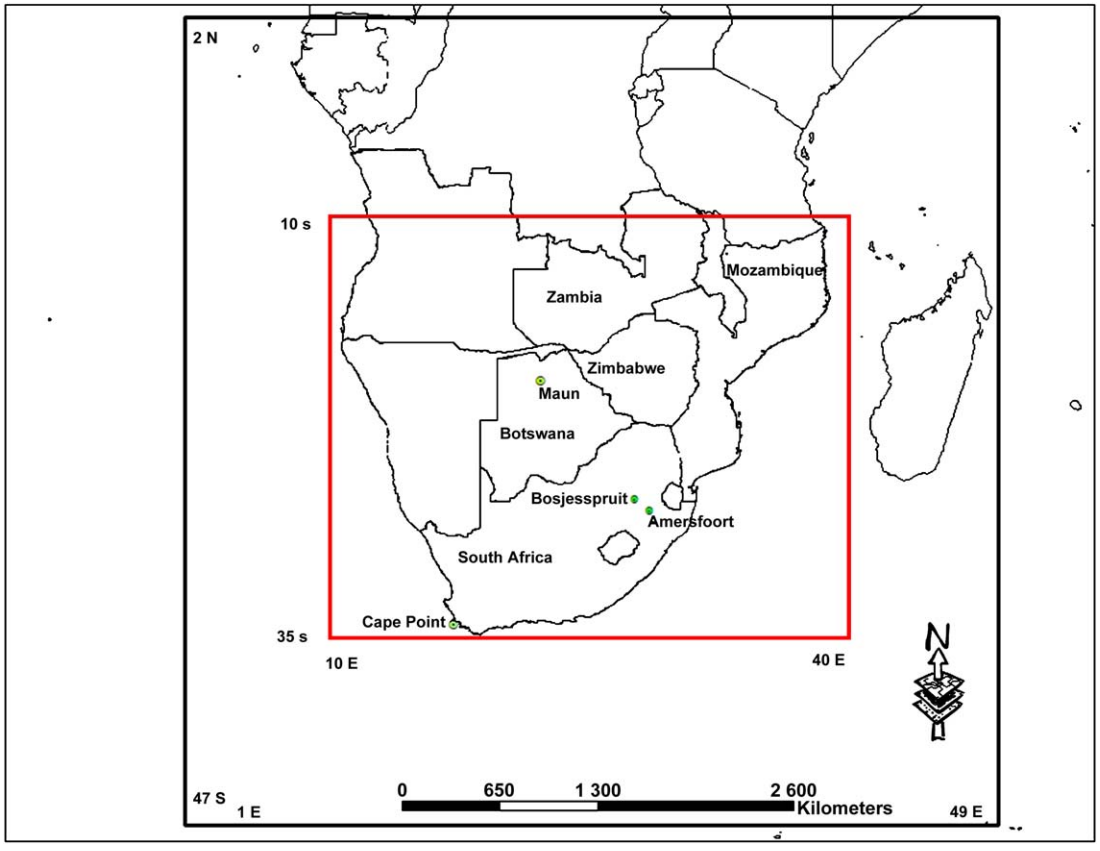


Fig. 1. The CAPIA study area indicating the participating countries and the extent of the larger and nested modelling domains.

maize by ozone using a threshold concentration of 40 ppb. Based on ozone concentrations alone they suggest that maize may be at risk in parts of southern Africa, including Zimbabwe and northeastern South Africa where this threshold is exceeded. This project focuses on the period October 2000 to April 2001 in order to assess the risk throughout the region during the entire growing period of maize. This paper elaborates on the modelling aspects of the CAPIA project and presents modelled spatial and temporal characteristics of surface ozone concentrations over southern Africa.

2. Methodology and data

2.1. Model selection

Many processes contribute to the observed concentrations of ozone at ground level. The processes are also influenced by meteorology, chemistry, emissions, and the nature of the underlying surface. Three-dimensional photochemical grid models simulate the complex non-linear interactions best (Hogrefe et al., 2001; Sokhi et al., in press). Of the available models, the Comprehensive Air Quality Model with extensions (CAMx), version 4.00 (ENVIRON, 2003; http://www.camx.com), was selected for the CAPIA modelling. CAMx is an Eulerian photochemical model that simulates the emission, dispersion, chemical reaction and removal processes in the lower troposphere by solving the chemical continuity equation for each species on a system of nested three-dimensional grids. The Carbon Bond 4 chemistry mechanism (Gery et al., 1989) is the most appropriate for regional-scale modelling (ENVIRON, 2003) and allows for 91 reactions and 36 species, providing an integrated assessment of gaseous and particulate air pollutants.

2.2. Model domain, grid resolution and modelling period

A modelling domain of 3000 km by 3000 km, extending from 10° to 35°S and from 10° to 40°E is defined to include the five participating countries (Fig. 1). The lowest 4 km of the troposphere are divided into 10 layers. The lowest layer is 70 m deep. Each layer of the modelling domain is divided into a 60 by 60 grid, giving 50 km grid resolution in both horizontal directions.

The data logistics of running CAMx and the meteorological model MM5 (http://www.mmm.ucar.edu/mm5) for every day of the period October 2000 to April 2001 are overwhelming and impractical. The modelling for this study is therefore limited to a 5-day period (120 h) in each month. The period of 10th to 14th each month was arbitrarily selected.

2.3. Input data

2.3.1. Meteorology

The synoptic scale meteorology of southern and central parts of southern Africa is largely influenced by the semi-permanent mid-latitude high pressure systems where the circulation is anticyclonic and subsident. Tropical easterlies have a stronger influence over the northern parts of the sub-continent. The atmosphere is also characterised by a number of permanent absolutely stable layers that are persistent and spatially continuous (Cosijn and Tyson, 1996). The absolutely stable layers inhibit vertical mixing and pollutants accumulate between the layers where they may be transported to great distances from source. Transport of pollutants over the sub-continent in the lower troposphere is controlled to a large extent by the semi-permanent synoptic scale anticyclonic gyre (Tyson et al., 1996; Garstang et al., 1996), which is characterised by recirculation over the sub-continent, an exit pathway off the sub-continent to the Atlantic Ocean at 20°S and an exit pathway at 30°S to the Indian Ocean. The duration and scale of the recirculation and the frequency of the westerly and easterly exit transport depends on season, level in the atmosphere and the dominant circulation type (Tyson, 1997).

Gridded global tropospheric reanalysis meteorological data (2.5° by 2.5° resolution) from the National Centre for Environmental Protection are used as initial input to the Pennsylvania State University and National Centre for Atmospheric Research mesoscale meteorological model, MM5. Surface and upper air observations were not used to nudge the predictions due to the scarcity of data. Other than over South Africa observed meteorological data are limited, or in the case of upper air data are non-existent. MM5 is run in a nested mode to generate gridded meteorological fields at 3-h intervals for each 5-day period. The larger domain of 5400 km by 5400 km, with a grid resolution of 150 km, is used to initialise the smaller nested domain of 3000 km by 3000 km, with resolution of 50 km. Terrain and landuse data from the U.S. Geological Survey are applied in both domains. Gridded meteorological data fields of height/pressure, wind speed and direction, temperature, water vapour, cloud/rain and vertical diffusivity are produced for input to CAMx. Meteorological processing in CAMx interpolates the 3-h MM5 data to hourly data for input to the dispersion module.

The MM5 meteorological fields compare favourably with measured data. Measured sea level pressure at Cape Town and Maputo at 12:00 GMT in October and November 2000 are compared with the modelled sea level pressure as an indicator of how well MM5 simulates the actual meteorology (Fig. 2). The correlation of 0.90 between the measure and modelled fields at the southern site (Cape Town) and the site towards the

north of the modelling domain (Maputo) indicates that the meteorology across the modelling domain is well simulated by MM5. The surface synoptic meteorology during each of the 5-day periods of the investigation is described below.

2.3.1.1. 10–14 October 2000. A low pressure trough is situated over the western parts of the sub-continent throughout this period, extending from southern Angola, southward through central Namibia to the central south coast of South Africa. On 10 October 2000 the strong Atlantic Ocean Anticyclone (AOA) (1032 hPa) is situated to the south east of the African sub-continent, ridging eastward on 11 October 2000, strengthening the Indian Ocean Anticyclone (IOA), and inducing an onshore flow over Mozambique during the remainder of the period.

2.3.1.2. 10–14 November 2000. A low pressure trough extends from western Zambia, southward across central Botswana and to the southeast coast of South Africa on 10 November 2000, with a cold frontal system situated to the southwest of the African sub-continent. The frontal system moves eastward on 11 and 12 November 2000, and is situated to the east of the sub-continent on 14 November 2000. During this time the trough over sub-continent also moves eastward, out into the Mozambique Channel. At the end of the period a trough is established over central Namibia.

2.3.1.3. 10–14 December 2000. A low pressure trough is situated over the western parts of the African sub-continent throughout this period, extending from eastern Angola, and southward across Namibia and Botswana to the south coast of South Africa. A weak AOA ridges south of the sub-continent on 11 December 2000 and induces a weak onshore flow over the eastern parts of South Africa and into southern Mozambique.

2.3.1.4. 10–14 January 2001. On 10 January 2001 a cold front is situated to the immediate southwest of the African sub-continent with a relatively deep trough further eastward over the sub-continent, extending from southern Zambia, across central Zimbabwe and over the eastern parts of South Africa. The front moves eastward on 11 January 2001, and is followed by a second front moving across South Africa on 13 and 14 January 2001. Further north the circulation is dominated by the low pressure trough over central Botswana.

2.3.1.5. 10–14 February 2001. A low pressure trough extends from southern Angola, across Namibia and western Botswana, to the south coast of South Africa on 10 February 2001. A weak frontal system moves eastward south of the sub-continent from 12 to 13 February 2001, moving the southern part of the interior trough to the east. By 15 February 2001 the trough axis is situated over eastern Botswana.

2.3.1.6. 10–14 March 2001. Weak pressure gradients exist over the entire region on 10 and 11 March 2001, but tighten on 12 March 2001 with the passage of a cold frontal system south of the sub-continent and with the formation of trough over central Botswana. The AOA ridges south of the sub-continent on 13 March 2001 and forces the trough to move westward over Namibia. The ridging also induces onshore flow over southern Mozambique.

2.3.1.7. 10–14 April 2001. A band of high pressure extends across the southern parts of the sub-continent on 10 April 2001, with a weak trough over central

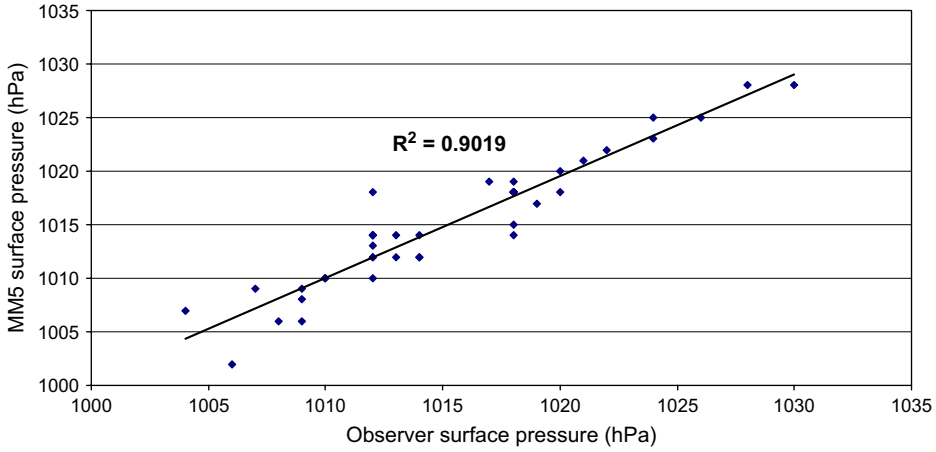


Fig. 2. Scatter plot of observed sea level pressure in hPa at Cape Town and Maputo with sea level pressure modelled by MM5 for two weeks in October and November 2000.

Namibia. Pressure remains high through the period over the southern parts as the AOA ridges eastward and up into Mozambique. Behind the ridging high the trough over Namibia deepens and extends southward to western Botswana by 14 April 2001.

2.3.2. Emissions

Annual average emissions of CO, SO_2, NO_x and total hydrocarbons resulting from anthropogenic activities (industry, transport and domestic burning) (Fleming and van der Merwe, 2002) are gridded to the defined CAMx grid. For most of the region the best available emissions data were used from 1990 National Emissions Inventories by countries that are signatory to the United Nations Framework Convention on Climate Change (UNFCCC). These inventories provide emissions data of several greenhouse gases by sector, such as transport and electricity generation. Emissions from area and linear sources were spatially disaggregated using ARC/INFO. The road and railway network together with population density were used to calculate emissions from the transport sector. Population density was used as a proxy for domestic biomass and coal burning to estimate emissions from domestic burning. Point source emissions were treated individually according to their respective geographic positions.

Very little data on inter-annual or diurnal variation was available in compiling the southern African greenhouse emissions inventory (Fleming and van der Merwe, 2002). Therefore in this study, annual emission rates in each grid cell are reduced to hourly emissions by simple division, i.e. it is assumed that no temporal variation occurs. This assumption may be valid for large industrial and mining sources, but it may have some limitations with regard to domestic biomass burning and transport which have seasonal and diurnal variations. The dominant activity in each grid cell is subsequently used to label the emissions from the cells as resulting from power generation, manufacturing, transport or domestic fuel burning. The defined activity and information on fuel type allows for speciation of the emissions in each cell into 16 species (ENVIRON, 2003).

Biogenic emissions of VOCs have been determined for the subequatorial region of Africa (Otter et al., 2003). Otter et al. (2003) used leaf area index, specific leaf mass and emission capacity with temperature and cloud cover data as input to algorithms described by Guenther (1999) to estimate biogenic isoprene and monoterpene emissions for one year. Similarly NO_x emissions from soils have also been determined (Scholes, 2002). These data are gridded for CAMx and reduced to hourly input fields. NO_x emissions from soil are dependent on temperature and soil moisture and vary with season and diurnally. VOC emissions from vegetation are dominated by isoprene and light-dependent monoterpenes. VOC emissions show daily and seasonal variations that peak with temperature and light intensity thus are not emitted at night. Stored monoterpene emissions are also temperature and humidity dependent with low emission rates at night and varying during the day with temperature.

Biomass burning in southern Africa is a seasonal phenomenon, occurring between May and September and peaking in July. Silva et al. (2003) report a total burned area of about 340 000 km^2 in southern Africa in July 2000, decreasing to less than 150 000 km^2 in September and to about 40 000 km^2 in October. By comparison, biomass-burning emissions after the onset of summer rains are small. The contribution by biomass burning to the formation of ozone during the CAPIA period, October to April, is not included in the emissions inventory.

2.3.3. Landuse and initial conditions

The variation in landuse across the modelling domain is captured by the distribution of one of 11 landuse types to each grid cell. These are the same land use data from the US Geological Survey that are used in the MM5 modelling runs. In this way, surface roughness and UV albedo are defined for each grid cell. Mean background concentrations of trace gases measured at the Global Atmosphere Watch station at Cape Point (Fig. 1) are used to initialise the first day of each month's 5-day model run. Concentrations from the last hour of day one are then used to initialise day two, and so on throughout the 5-day model run. These data are also used to define the boundary conditions.

3. Model results

Average modelled hourly concentrations of ozone over southern Africa range between 20 and 50 ppb in all months between October 2000 and April 2001 (Fig. 3). In all months the highest concentration occur in a broken band of between 30 and 40 ppb that stretches from west of Namibia, across the southern parts of Botswana, into northeastern South Africa and across Mozambique. In October and November and in March and April the relatively higher concentrations extend northwards over northern Mozambique and into northern Zambia. Pockets of higher concentrations exist within this band where the means reach 50 ppb. For much of the region the mean modelled ozone concentrations are between 20 and 30 ppb.

The modelled maximum hourly concentrations of surface ozone provide a different perspective (Fig. 4). Over much of South Africa, Botswana, Mozambique and Zambia the maximum ozone concentrations vary between 20 and 40 ppb. Similar to the average modelled concentrations, a band of higher concentrations is evident in all months where maximums exceed 40 ppb

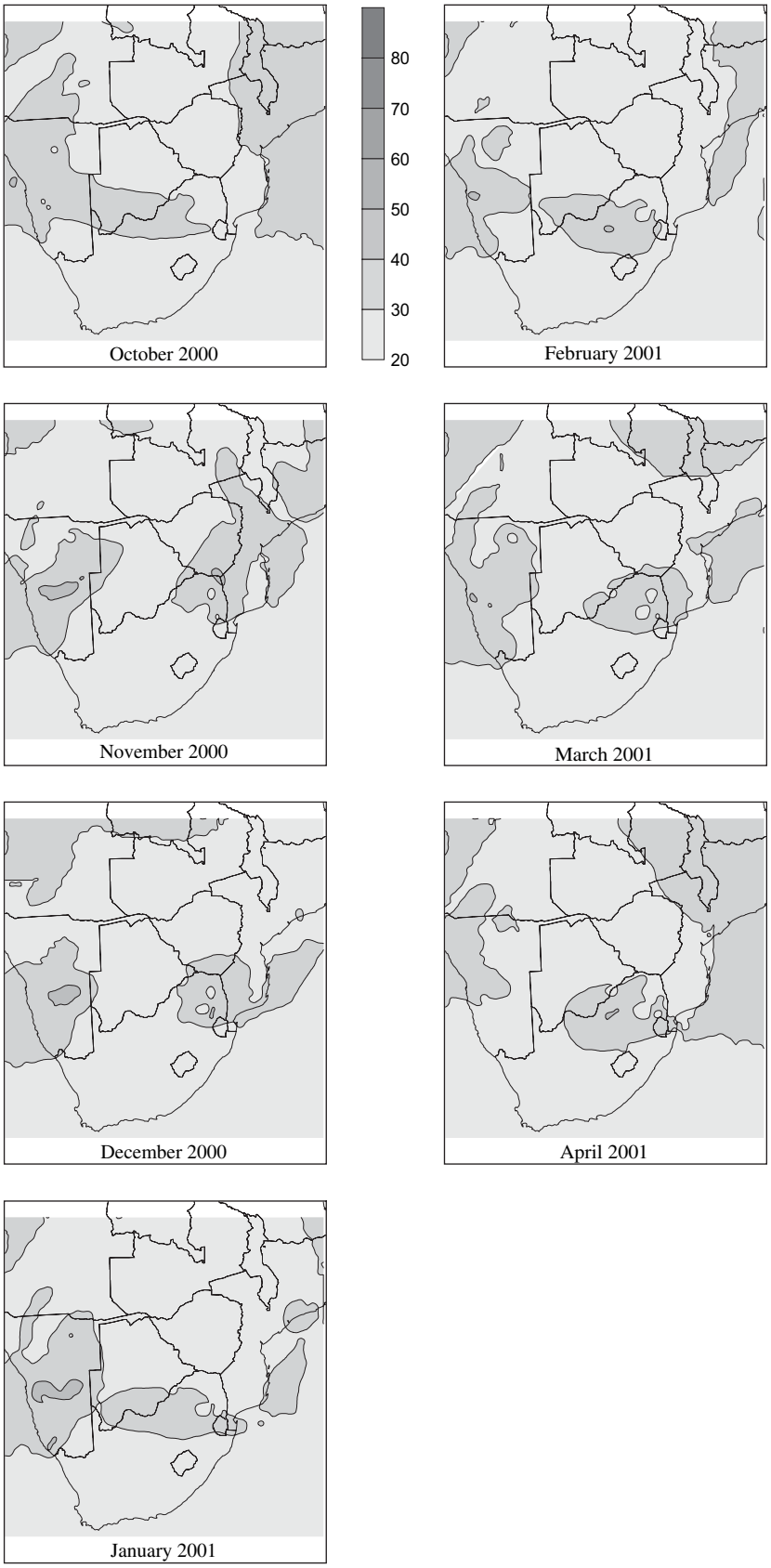


Fig. 3. Average modelled ozone concentrations over southern Africa in ppb for the 5-day period, 10th to 14th in each of the months, October 2000 to April 2001.

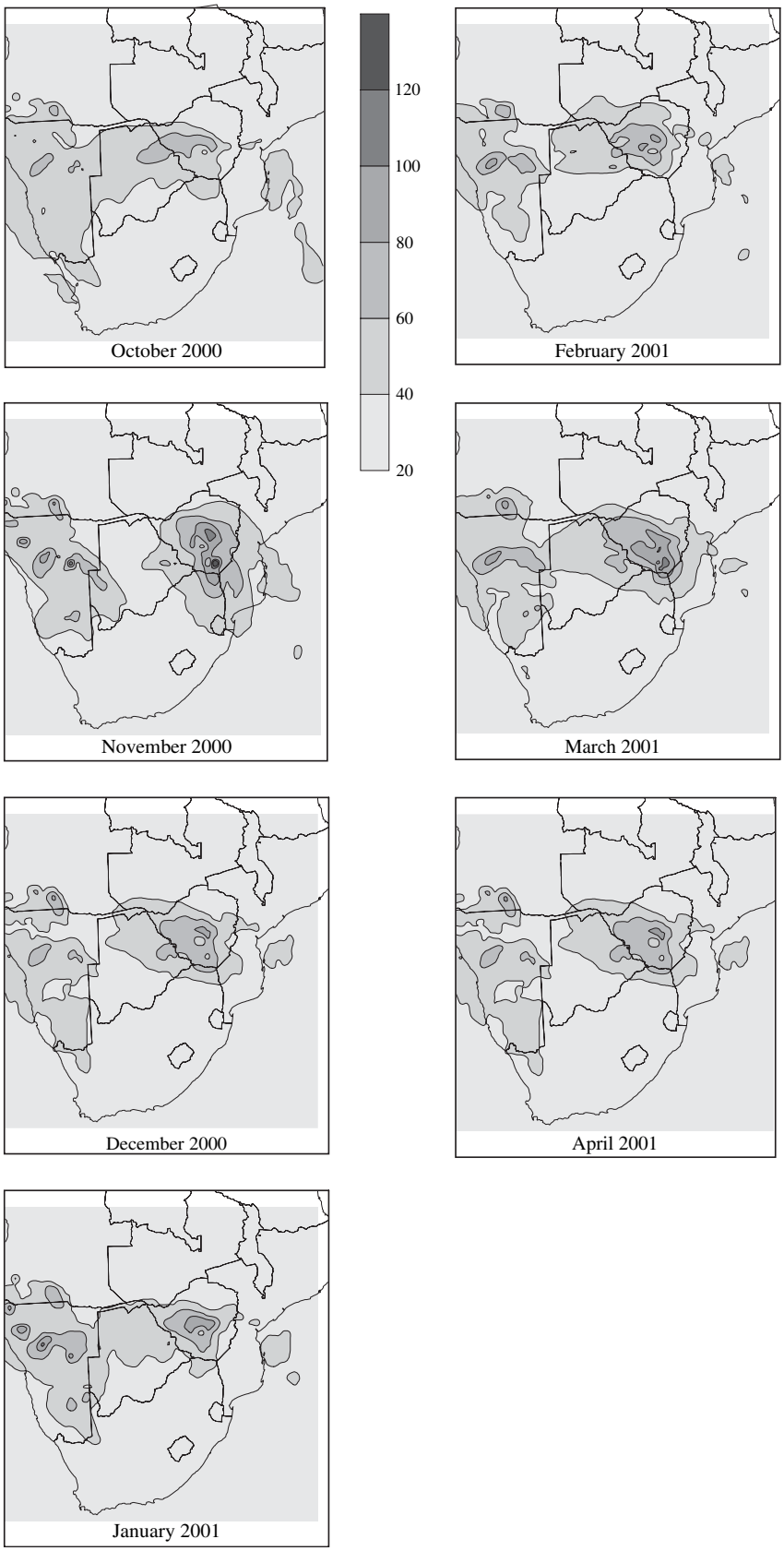


Fig. 4. Maximum modelled ozone concentrations over southern Africa in ppb for the 5-day period, 10th to 14th in each of the months, October 2000 to April 2001.

and reach more than 100 ppb in places. This band is different to the mean concentrations in that it is orientated from central Namibia towards the northeast across Botswana, over Zimbabwe and into the extreme western parts of Mozambique. The highest modelled maximum concentrations of 80 ppb and more occur over large parts of Zimbabwe in most months, stretching down into northeastern South Africa in March and April, and over northern Namibia in most months. The different spatial patterns between the average and maximum modelled ozone concentrations suggest that the maximums either occur infrequently, or they are associated with low concentrations during the rest of the modelling period which then contribute to relatively low average concentrations. This point is elaborated in the following discussion on the modelled diurnal variations.

Modelled ozone concentrations are compared in a single model grid cell in South Africa, Zimbabwe and Zambia for the 120 h period, 10th to 14th of each month from October 2000 to April 2001 in Fig. 5. The grid cell in each of the countries lies in the maize growing area and the cell has the highest modelled ozone concentration.

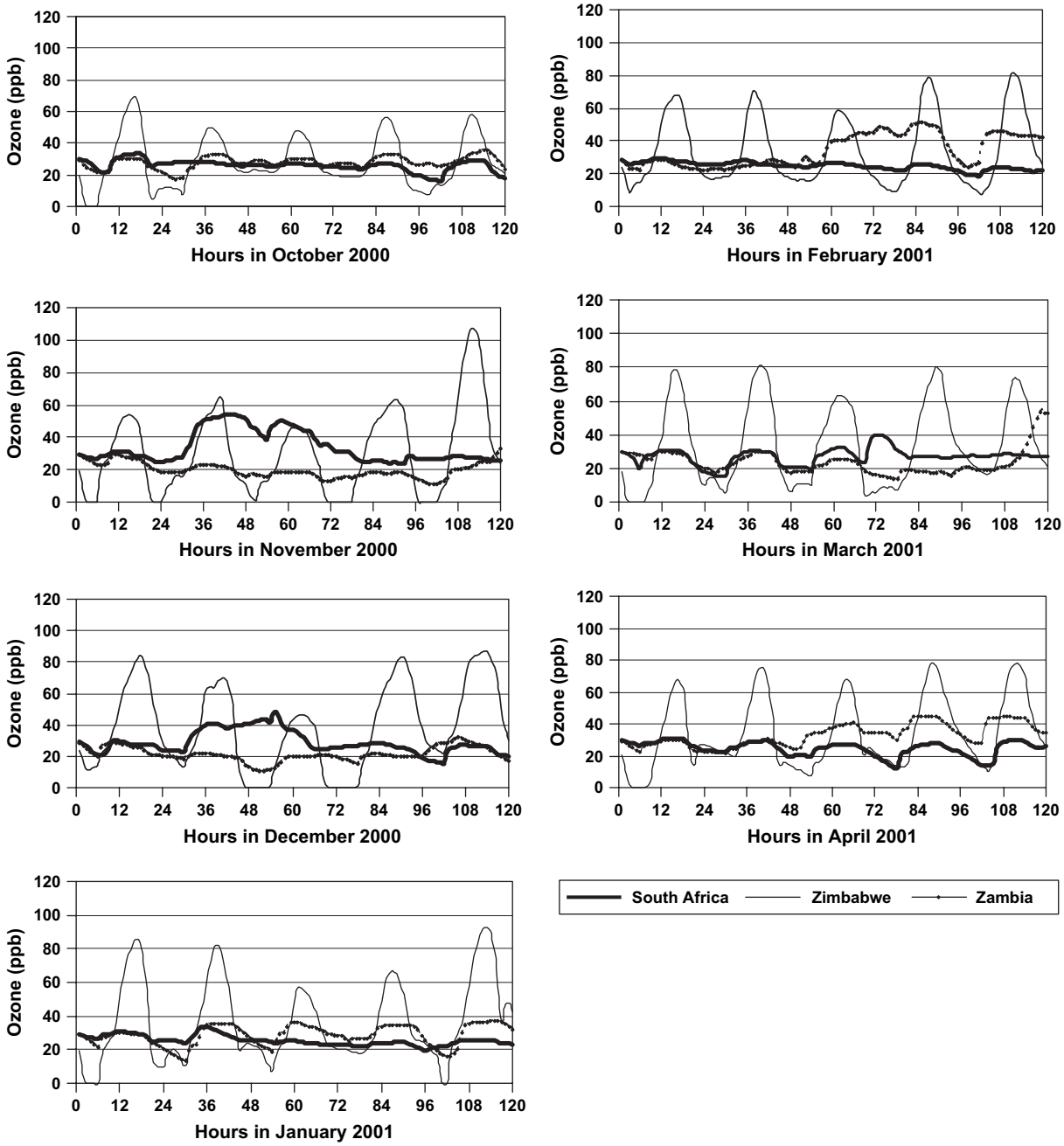


Fig. 5. Diurnal variation in ozone concentration (ppb) during the 120 model run from 10th to 14th in each month from October 2000 to April 2001 in the selected grid cell in the maize growing areas in South Africa, Zambia and Zimbabwe.

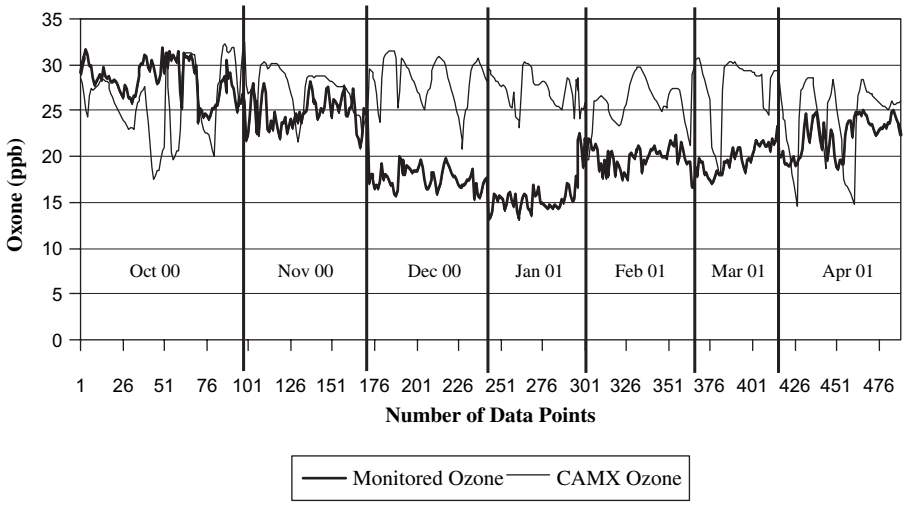


Fig. 6. Measured and modelled surface ozone at Cape Point during the period October 2000 to April 2001.

In each of the countries the modelled surface ozone concentrations show clear diurnal variations, varying from low concentrations at night and reaching maximums in the early afternoon. The diurnal cycle is most pronounced in Zimbabwe where the daytime peaks are consistently over 60 ppb in all months, reaching more than 100 ppb on 14 November 2000. At night the modelled ozone concentrations over Zimbabwe fall to zero. Over South Africa and Zambia the modelled concentrations vary less during the day than over Zimbabwe and generally range between 20 and 40 ppb, exceeding 40 ppb in South Africa in November 2000 and in February, March and April 2001 in Zambia. The mean concentrations in the selected cells in Zambia, Zimbabwe and South Africa are similar despite the large difference seen in the diurnal variation. The difference from month-to-month is also small.

The temporal variations provide an indication of the number of hours when ozone concentrations exceed the 40 ppb threshold. Over Zimbabwe this concentration threshold is exceeded in every month for a total of 308 of the 840 modelling hours. The longest period of exceedance is 54 of the 120 h in December 2000, and the shortest was 38 h in October 2000. A total of 78 h exceeded 40 ppb over Zambia between February and April 2001, with 49 of these in February 2001. Over South Africa there were only 48 h over 40 ppb in November and December 2000.

4. Comparison with measured data

A number of points should be borne in mind when comparing monitored data with the modelled concentrations. Firstly, monitoring stations are not selective and measure ozone results from emissions from all processes. Assumptions made in the CAPIA project limit the emissions used in the modelling of anthropogenic and biogenic sources. In turn, the anthropogenic emissions used in CAPIA may not include all sources. Furthermore, the selected VOC speciation represents the dominant anthropogenic activity of industrial processes in each grid cell. In reality there are a variety of industries present in the respective cells. The biogenic emissions are limited to only three VOC species. Emissions from biomass burning are excluded in the CAPIA project on the basis of them being low in the summer. A further point to consider is that monitoring stations measure surface ozone at a specific point while CAMx provides an estimate of the concentration in a grid block of 2500 km^2 and of 70 m deep.

Surface ozone is monitored at a few sites in southern Africa using both passive and active sampling (Zunckel et al., 2004). Zunckel et al. (2004) provide an overview of surface ozone measurements during the CAPIA project. The highest ozone concentrations occur over Botswana and the Mpumalanga highveld where the springtime maximums range between 40 and 55 ppb, but reached more than 90 ppb at times, mainly in the spring months. The monthly minimum in Botswana and the highveld is between 20 and 30 ppb. Maximum monthly ozone concentrations at the background International Global Atmospheric Chemistry DEBITS stations of Cape Point, at Etosha in Namibia and Ben MacDhui (Fig. 1) are between 20 and 30 ppb and the minimums range between 10 and 20 ppb.

There is reasonable agreement between monitored ozone concentrations at the background stations and the modelled concentrations. This agreement may be expected as these stations are well removed from major anthropogenic inputs which are subject to the greatest uncertainty considering the assumptions made regarding the emissions used in CAPIA. At Etosha and Ben MacDhui the mean modelled and measured

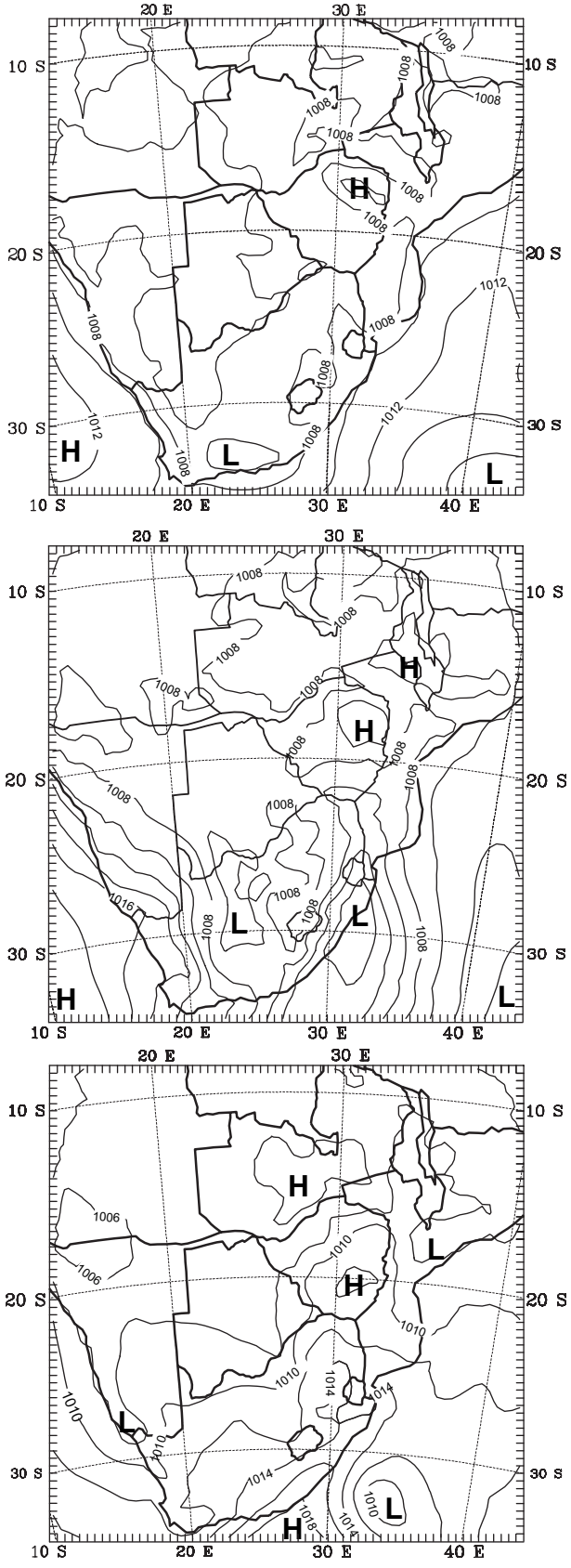


Fig. 7. Sea level isobaric charts over southern Africa at 12:00 GMT on 10 November 2000 (top), 11 November 2000 (centre), and on 14 November 2000 (bottom).

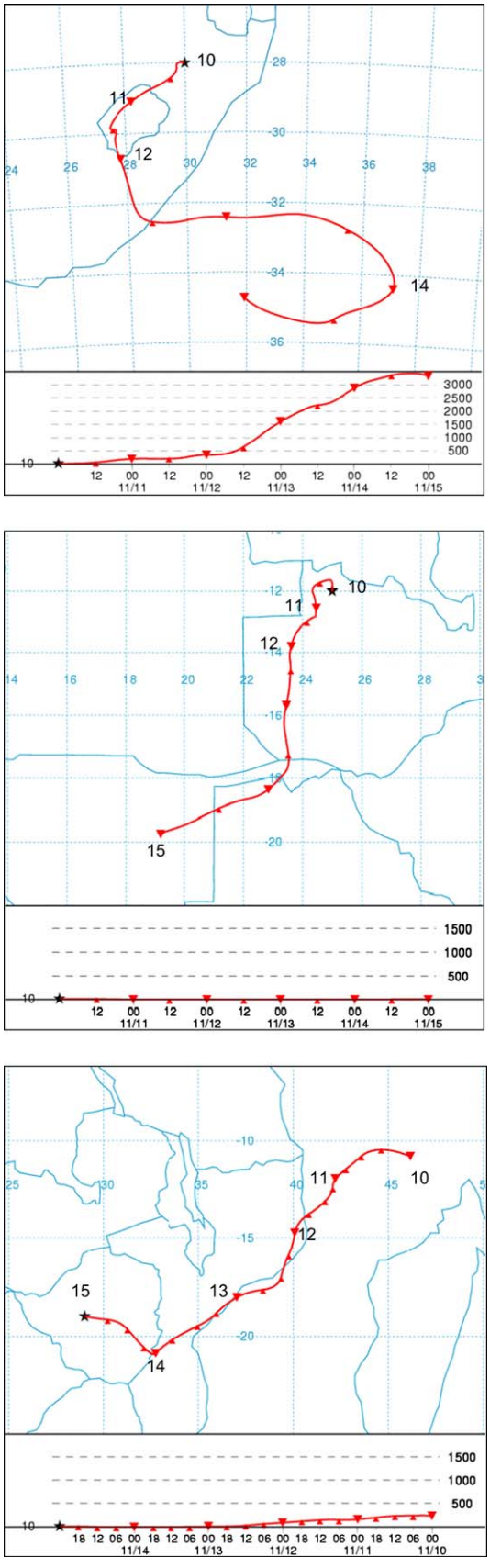
concentrations range between 20 and 30 ppb between October 2000 and April 2001 (Fig. 3). The annual mean concentrations at the background stations are 21 ppb at Etosha and 26 ppb at Ben MacDhui (Zunckel et al., 2004). Mean monthly ozone concentrations at DEBITS sites on the South African highveld range between minimum values in the region of 14 ppb near the main industrial source area at Elandsfontein and winter maximums of more than 40 ppb at Elandsfontein and at Palmer which is some 100 km north of the main industrial source region.

Ozone measured at Cape Point is mainly attributed to that which is formed naturally by ultraviolet in background marine air (Brunke and Scheel, 1998), while the ozone modelled by CAMx is a result of photochemistry with anthropogenic and biogenic precursors. Despite these differences and the uncertainties associated with comparing point measurements and modeled data over a 2500 km^2 area, the modelled ozone concentrations in October and November 2000 and during period at the end of April 2002 are compared at Cape Point (Fig. 6). The observed daily variation is modelled well by CAMx and despite a few outlying modelled concentration, the differences between measured and modelled concentrations are generally less than 5 ppb. During the mid-summer months, December 2000 to March 2001, the modelled surface ozone concentrations are consistently higher than the measure values, with difference between 7 and 14 ppb. This is possibly attributed to higher biogenic emissions during the summer resulting in higher modelled ozone. Similar comparisons at other stations is not possible due to either a lack of temporarily resolved data, or in cases, these data are not easily obtained.

5. Source–receptor relationships

The process of relating the observed surface ozone concentrations to any particular source-type or source-region is not that simple due to the number of non-linear process interactions that are involved, including chemistry and meteorology. The production of ozone requires NO_x and VOCs to exist in the correct ratios and sunlight for the photochemical reaction to take place. Depending on the prevailing air transport of the region, it may therefore be expected to find the highest ozone concentrations in or near to main ozone precursor source areas under a recirculation situation with strong anticyclonic flow, or some distance down wind of the anticyclonic gyre on the Atlantic Ocean or Indian Ocean exit pathways.

The major anthropogenic source regions of atmospheric pollutants in southern Africa are the mining and smelting activities on the Copperbelt in northern Zambia and emissions from coal combustion by industry on the



South African highveld (Fleming and van der Merwe, 2002). Emissions of NO_x from soils occur throughout the region while the emissions of VOCs from vegetation are highest in the moister tropical regions and in the east, decreasing towards the west and the south (Otter et al., 2003). It is therefore interesting to observe that the maximum modelled ozone concentrations occur over central Zimbabwe (Fig. 4) and not close to or downwind of the Zambian and South African industrial source areas.

The HYSPLIT-4 (Hybrid Single-Particle Lagrangian Integrated Trajectory) (Draxler and Hess, 1998) is used in a case study to evaluate the likely precursor source areas that result in the observed ozone maximum over central Zimbabwe in November 2000. At the start of the 5-day period a deep trough is initially situated through central Botswana, running southwards to a coastal low situated off the South African south coast (Fig. 7). The position of the trough axis is maintained over the northern parts of the region throughout the period, but it moves eastward over the southern parts with the passing and deepening of the coastal low.

A 5-day forward trajectory started on 10 November 2000 from the South African highveld (Fig. 8, top) initially moves towards the southwest along the trough line, continually gaining height, before turning to the west and exiting the sub-continent at about 32°S. Similarly, a 5-day forward trajectory initiated in northern Zambia on 10 November 2000, the other major anthropogenic source area, initially moves in a southerly direction for four days before crossing northern Botswana and into Namibia, remaining near ground level throughout the period. In this case it appears that neither of the two industrial source areas is responsible for the ozone maximum over Zimbabwe. A 5-day backward trajectory initiated from central Zimbabwe at midnight on 15 November 2000 indicates that air originates over the Indian Ocean 5 days earlier on 10 November 2000. It subsequently moves in a southwesterly direction near ground level along the Mozambique coast, before turning westward at the apex of the trough and moving across Zimbabwe. There are no major anthropogenic source areas along this trajectory path, but the biogenic VOC and NO_x emissions are relatively high along this trajectory, particularly in the moister east over Mozambique. This suggests that biogenic precursor emissions may play a significant role in the observed and modelled surface ozone concentrations over southern Africa.

Fig. 8. Five-day forward-trajectories from the South African highveld (top), the Zambian Copperbelt (centre) from 10 to 14 November 2000 and a 5-day backward trajectory from central Zimbabwe (bottom) from 15 to 10 November 2000. The triangles on the trajectories indicate the start of each day, i.e. midnight. The height of each trajectory is plotted in the lower part of each panel.

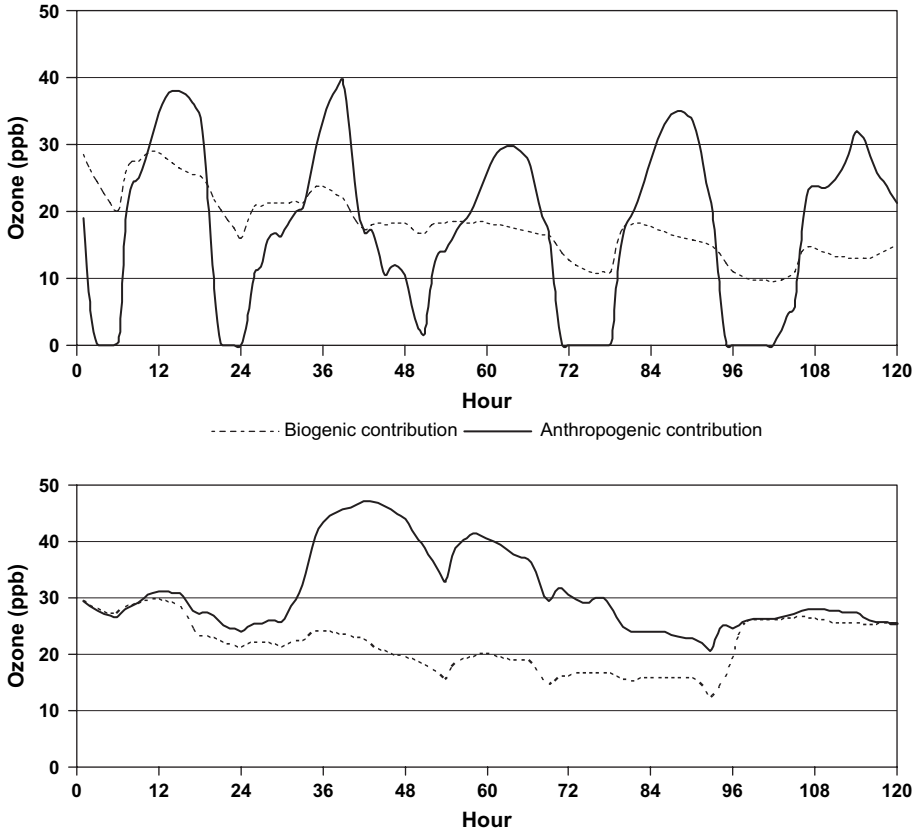


Fig. 9. Modelled surface ozone concentration (10–14 November 2000) over southern Zimbabwe (upper) and northeastern South African (lower) as a result the individual contributions of anthropogenic and biogenic emissions.

On the contrary, ozone that is formed in the Zimbabwean area of maximum ozone concentration as a result of anthropogenic emissions only is very similar to the ozone that is attributed only to biogenic emissions (Fig. 9, upper). For the November 2000 case study ozone formation for the individual emission types is modelled separately. The mean ozone concentration during this period for each emission type is 17.8 ppb. Ozone formed from anthropogenic emissions shows a far greater diurnal range, from 0 ppb at night to daytime maximums of between 30 and 40 ppb, despite the assumptions that the modelled emission sources have no temporal variation. The observed variation is therefore attributed to the solar cycle. Ozone formed from biogenic emissions has a weaker diurnal range, with nighttime minima varying between 10 and 17 ppb, and daytime maxima varying between 18 and 28 ppb. A similar situation is seen during the same period over northeastern South Africa (Fig. 9, lower) where ozone resulting from anthropogenic emissions only has a bigger daily range than that attributed to biogenic emissions. The mean ozone concentrations in this area are 31.0 and 21.6 ppb for anthropogenic and biogenic emissions, respectively.

It is apparent therefore that the modelled surface ozone over southern Africa, seen in Figs. 3–5, is a result of a combination of precursors from anthropogenic and biogenic sources and the solar cycle. Ozone formed from anthropogenic and biogenic sources individually occurs in lower concentrations when these source types occur together. It appears that collectively these source types contribute to the high concentrations of ozone over southern Africa.

6. Conclusion

The objectives of the Cross Border Air Pollution Impact Assessment Project (CAPIA) are to assess the potential risk of damage to maize by ozone in southern Africa. Ozone monitoring is conducted at only a few sites in the region that do not coincide with the maize growing areas. To meet the objectives of the project, ozone concentrations resulting from the chemical transformations between anthropogenic and biogenic emissions were modelled using the photochemical transport Comprehensive Air Quality Model with Extensions (CAMx), providing spatial and temporal characteristics of surface ozone concentrations over southern Africa.

Average modelled hourly concentrations of ozone over southern Africa range between 20 and 50 ppb during the maize growing season. Relatively higher

concentrations extend eastwards in a band from central Namibia, across the whole region and into Mozambique, then northwards into northern Zambia. During this time a band of higher concentrations is evident, reaching 40 ppb over large areas and more than 100 ppb in places. The highest maximum concentrations of 80 ppb and more are modelled over large parts of Zimbabwe, stretching down into northeastern South Africa and over Namibia.

In the CAPIA project a threshold concentration of 40 ppb is used to assess potential risk of damage to maize by ozone. Over Zimbabwe the 40 ppb threshold for surface ozone is exceeded in every month of the CAPIA study period with a total of 308 of the 840 modelling hours being exceeded. A total of 78 h of exceedance is modelled in Zambia and 48 h in South Africa. The implication at these levels of exposure is that southern African crops may indeed be at risk to damage by ozone. Southern Africa has an abundance of industrial and other anthropogenic sources. The modelling conducted here suggests that neither anthropogenic nor biogenic emission sources dominate in the formation of the high ozone concentrations over parts of the region. Rather the two emission source types contribute precursors collectively to the formation of ozone in concentration that exceeds 40 ppb over large areas. The predicted ozone concentrations may be underestimated considering that the model estimations do not include the potential contribution from biomass burning to ozone formation. Emission of ozone precursors from natural sources and those from anthropogenic origin play a combined and significant role in the production of surface ozone over southern Africa on a regional scale.

Acknowledgements

The South African Department of Science and Technology is acknowledged for the financial support of the CAPIA project through the Regional Science and Technology fund. The South African Weather Service is thanked for the Cape Point data. The Air Pollution Information Network – Africa (APINA) are acknowledged for their in-kind support. Gavin Fleming of CSIR is thanked for assisting with gridding the biogenic and anthropogenic emissions data.

References

Annegarn, H.J., Turner, C.R., Helas, G., Tosen, G.R., Rorich, R.P., 1996. Gaseous pollutants. In: Held, G., Gore, B.J., Surridge, A.D., Tosen, G.R., Turner, C.R., Walmsley, R.D. (Eds.), Air Pollution and its Impacts on the South African Highveld. Environmental Scientific Association, Cleveland, 144 pp. (Chapter 6).

Brunke, E.-G., Scheel, H.E., 1998. Surface ozone measurements at Cape Point (34°S, 18°E). In: Bojkov, R.D., Visconti, G. (Eds.), Atmospheric Ozone, Proceedings of the XVIII Quadrennial Ozone Symposium, L'Aquila, Italy, 12–21 September 1996, pp. 331–334.

Cosijn, C., Tyson, P.D., 1996. Stable discontinuities in the atmosphere over southern Africa. South African Journal of Science 92, 381–396.

Diab, R.D., Jury, M.R., Combrink, J., Sokolic, F., 1996a. A comparison of anticyclonic and trough influences on the vertical distribution of ozone and meteorological conditions during SAFARI-92. Journal of Geological Research 101 (D19), 23809–23821.

Diab, R.D., Thompson, A.M., Zunckel, M., Coetzee, G.J.R., Combrink, J., Bodeker, G.E., Fishman, J., Sokolic, F., McNamara, D.P., Archer, C.B., Nganga, D., 1996b. Vertical ozone distribution over southern Africa and adjacent oceans during SAFARI-92. Journal of Geological Research 101 (D19), 23823–23833.

Draxler, R.R., Hess, G.D., 1998. An overview of the HYSPLIT-4 Modelling system for trajectories, dispersion and deposition. Australian Meteorological Magazine 47, 295–308.

Emberson, L.D., Ashmore, M.R., Murray, F., Kuylenstierna, J.C.I., Percy, K.E., Izuta, T., Zheng, H., Sheu, B.H., Liu, C.P., Agrawal, M., Wahid, A., Abdel-Latif, N.M., van Tienhoven, M., de Bauer, L.I., Domingos, M., 2001. Impacts of air pollutants on vegetation in developing countries. Water, Air and Soil Pollution 130, 107–118.

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

Fleming, G., van der Merwe, M., 2002. Spatial disaggregation of greenhouse gas emissions inventory data for Africa south of the equator, <http://gis.esri.com/library/userconf/proc00/professional/papers/PAP896/p896.htm>.

Garstang, M., Tyson, P.D., Swap, R.J., Edwards, M., Kållberg, P., Lindsay, J.A., 1996. Horizontal and vertical transport of air over southern Africa. Journal of Geophysical Research 101, 23721–23736.

Gery, M.W., Whitten, G.Z., Killus, J.P., Dodge, M.C., 1989. A photochemical kinetics mechanism for urban and regional scale computer modelling. Journal of Geophysical Research 94, 925–956.

Guenther, A., 1999. Modelling biogenic volatile organic compound emissions to the atmosphere. In: Hewitt, C.N. (Ed.), Reactive Hydrocarbons in the Atmosphere. Academic Press, San Diego, California, pp. 97–118.

Hogrefe, C., Rao, T.S., Kasibhatla, P., Hao, W., Sistla, G., Mathur, R., Mc Henry, J., 2001. Evaluating the performance of regional-scale photochemical modelling systems: Part II-ozone predictions. Atmospheric Environment 35, 4175–4188.

Jenkins, M.J., Clemitshaw, K.C., 2000. Ozone and other secondary photochemical pollutants: chemical processes governing their formation in the planetary boundary layer. Atmospheric Environment 34, 2499–2527.

Jonnalagadda, S.B., Bwila, J., Kosmos, W., 2001. Surface ozone concentrations in Eastern Highlands of Zimbabwe. Atmospheric Environment 35, 4341–4346.

Kirkman, G.A., Piketh, S.J., Andreae, M.O., Annegarn, H.J., Helas, G., 2000. Distribution of aerosols, ozone and carbon monoxide over southern Africa. South African Journal of Science 96, 423–431.

Lee, D.S., Holland, M.R., Fella, N., 1996. The potential impact of ozone on materials in the U.K. Atmospheric Environment 30 (7), 1053–1065.

Lippman, M., 1989. Health effects of ozone: a critical review. Journal of the Air and Waste Management Association 39, 672–695.

Meixner, F.X., Helas, G., 1994. Meteorology, transport and boundary layer ozone at Victoria Falls (Zimbabwe): ground level and airborne observations. In: Proceeding of the AUG Fall Meeting 1994, AUG, San Francisco, 147 pp.

Otter, L., Fleming, G., Wiedinmyer, C., Guenther, A., Greenberg, J., Harley, P., 2003. Spatial and temporal variations in biogenic VOC emissions for Africa south of the equator. Journal of Geophysical Research 108 (D13), doi: 10.1029/2002JD002609.

Scholes, R.J. (Ed.), 2002. SAFARI 2000: nitrogen and sulphur deposition in southern Africa. Final Report to Department of Arts, Culture, Science and Technology, South Africa (Lead fund). CSIR Environmentek, Pretoria. ENV-P-C 2002-020, 88 pp.

Silva, J.M.N., Pereira, J.M.C., Cabral, A.I., Sá, A.C.L., Vasconcelos, M.J.P., Mota, B., Grégoire, J.M., 2003. An estimate of area burned in southern Africa during the 2000 dry season using SPOT-VETETATION satellite data. Journal of Geophysical Research 108 (D13), 8498, doi: 10.1029/202JD002325.

Sokhi, R.S., San José, R., Kitwiroon, N., Fragkou, E., Péres, J.L., Middleton, D.R., Prediction of ozone levels in London using MM5–CMAQ modelling system. Environmental Modelling and Software, in press. doi: 10.1016/j.envsoft.2004.07.016.

Tyson, P.D., 1997. Atmospheric transport of aerosols and trace gases over southern Africa. Progress in Physical Geography 21 (1), 79–101.

Tyson, P.D., Garstang, M., Swap, R.J., Kållberg, P., Edwards, M., 1996. Air transport climatology for subtropical southern Africa. International Journal of Climate 16, 265–291.

van Tienhoven, A.M., Scholes, M.C., 2003. Air pollution impacts on vegetation in South Africa. In: Emberson, Lisa, Ashmore, Mike, Murray, Frank (Eds.), Air Pollution Impacts on Crops and Forests. A Global Assessment. Air Pollution Reviews, vol. 41-86094-292-X.

van Tienhoven, A.M., Zunckel, M., Koosailee, A., Gwata, C., Lungu, C., Emberson, L.D. Preliminary assessment of risk of ozone impacts to maize (*Zea mays*) in southern Africa, submitted for publication.

Zunckel, M., Venjonoka, K., Pienaar, J.J., Brunke, E.-G., Pretorius, O., Koosailee, A., Raghunandan, A., van Tienhoven, M.A., 2004. Surface ozone over southern Africa: synthesis of monitoring results during the Cross Border Air Pollution Impact Assessment project. Atmospheric Environment 38, 6139–6147.