Modelled transport and deposition of sulphur over Southern Africa

M. Zunckel[a,b,*], L. Robertson[c], P.D. Tyson[a], H. Rodhe[d]

[a]*Climatology Research Group, University of the Witwatersrand, Johannesburg, South Africa*
[b]*CSIR Environmentek, P.O. Box 17001, Congella, South Africa*
[c]*Swedish Meteorological and Hydrological Institute, S-601 76, Norrköping, Sweden*
[d]*Department of Meteorology, Stockholm University, S-106 91, Sweden*

Abstract

Ambient SO_2 concentrations and atmospheric deposition of sulphur resulting from emissions on the industrialised highveld region of South Africa are estimated using the multi-scale atmospheric transport and chemistry (MATCH) modelling system, developed at the Swedish Meteorological and Hydrological Institute (SMHI), and compared with an inferential model driven by measured input quantities. Modelled SO_2 concentrations on the central highveld mostly range between 10 and 50 ppb, exceeding 50 ppb in source areas. Dry deposition rates for sulphur exhibit a similar spatial pattern to the ambient SO_2 concentrations and both are consistent with synoptic-scale transport patterns. Maximum dry deposition rates for sulphur of more than 10 kg S ha^{-1} a^{-1} occur over the central highveld with a well-defined gradient decreasing away from the source region. Despite the significant differences in modelling approaches, the estimates of dry deposition provided by MATCH are in reasonable agreement with those of the inferential model. The maximum modelled wet deposition rates occur over the South African highveld and its periphery and range between 1 and 5 kg S ha^{-1} a^{-1} and compare favourably with measurements from an acid rain network. Wet deposition generally exceed dry deposition on the highveld and the adjacent areas except in the central highveld source region. Over the drier western half of South Africa MATCH-modelled dry and wet deposition rates are again similar and are less that 1 kg S ha^{-1} a^{-1}. Wet deposition exceeds dry in the higher rainfall regions along the south and east coasts of South Africa. © 2000 Published by Elsevier Science Ltd. All rights reserved.

Keywords: South Africa; Deposition; Sulphur; MATCH model; Inferential technique

1. Introduction

Given the location of the subcontinent of southern Africa in the subtropical high-pressure belt, the mean circulation at 800 hPa over southern Africa is anticyclonic for much of the year, exceeding 80% in winter (Preston-Whyte and Tyson, 1988; Tyson et al., 1996) (Fig. 1). The general subsidence that prevails in the anticyclonic conditions ensures that fine weather predominates over much of the interior plateau (approximately 1500 m ASL). The inherently poor pollutant dispersion conditions attendant on anticyclonic flow patterns are exacerbated by the presence of semi-permanent, subsidence-induced absolutely stable layers at 700 and 500 hPa that extend over the subcontinent (Cosijn and Tyson, 1996). The semi-permanent south Atlantic anticyclone, the continental anticyclone over southern African, and the south Indian anticyclone have a dominant effect on the transport of aerosols and trace gases in the troposphere over southern Africa. Pollutants are trapped below the stable layers and their transport away from the highveld source region is largely controlled by

---

* Corresponding author. Tel.: + 31-261-8161; fax: + 31-261-2509.

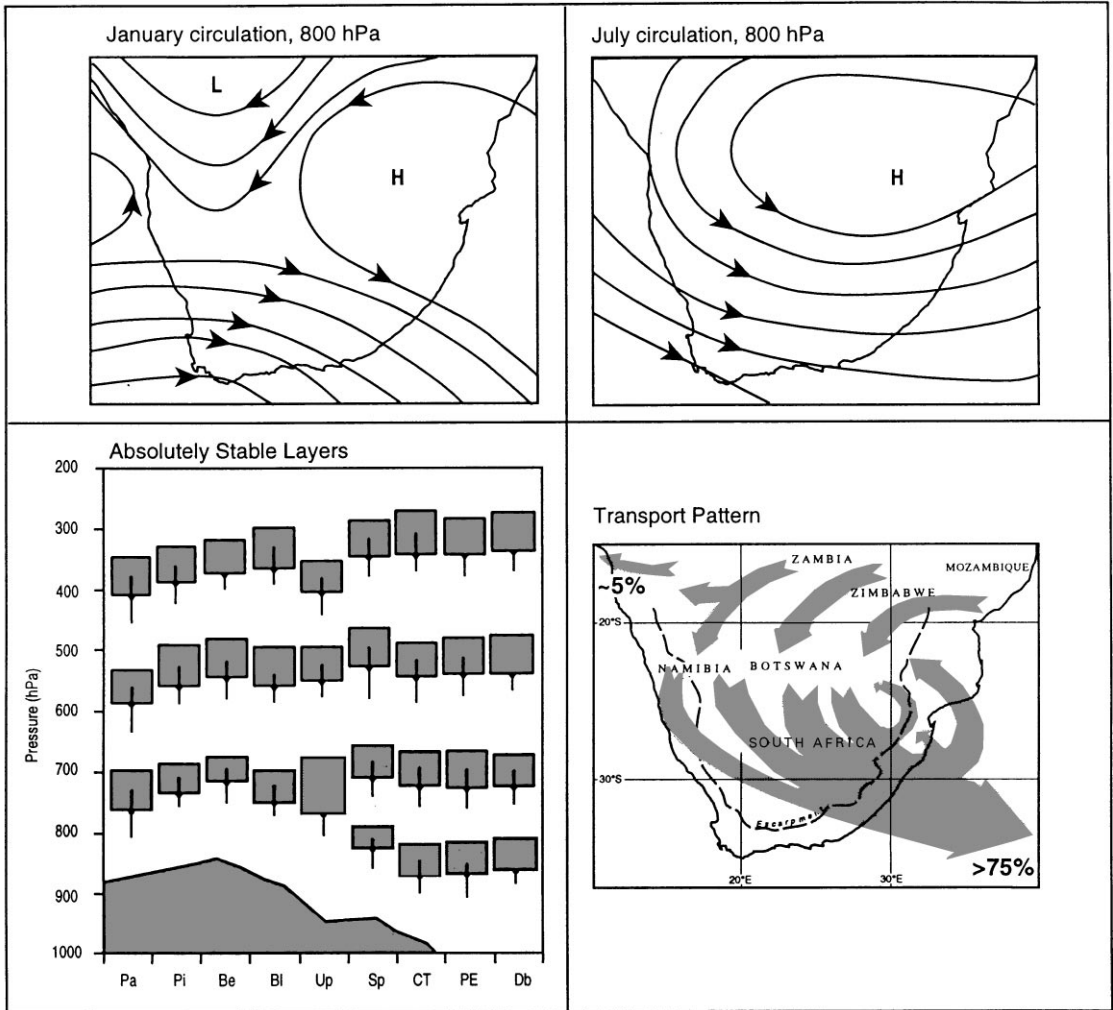*E-mail address:* mzunckel@csir.co.za (M. Zunckel).

Fig. 1. Mean 800 hPa circulation over southern Africa in summer (January) and winter (July) (after Tosen and Jury, 1986), temporarily consistent absolutely stable layers observed over southern Africa (after Cosijn and Tyson, 1996) and major transport pathways over southern Africa (after Tyson and Preston-Whyte, 1999).

the relative position of the anticyclones. The resultant anticyclonic transport dominates and leads to recirculation over southern Africa of varying scales (Fig. 1). It may be limited to the subcontinent or be of a larger scale with the re-circulated limb returning to the subcontinent after initially exiting to the Indian or Atlantic Oceans (Garstang et al., 1996; Tyson et al., 1996; Sturman et al., 1997; Tyson and D'Abreton, 1998). Little of the transport takes place to the Atlantic Ocean, whereas more than 75% of all transport over the subcontinent is to the Indian Ocean and beyond.

Sulphur dioxide (SO_2) is an air pollutant most often associated with coal combustion for power generation, fossil fuel refining and ore smelting. In the developing southern African region south of 18 S, the average annual emission density for sulphur is estimated to 3 kg S ha^{-1} and ranges from zero in the desert and bushveld areas to as much as 200 kg S ha^{-1} on South Africa's highveld region and in parts of Zimbabwe (Sivertsen et al., 1995). More than a decade of network measurements over South Africa have provided a reasonable understanding of wet deposition resulting from the highveld emissions (e.g. Bluff et al., 1991; Turner et al., 1996; Galpin and Turner, 1999a,b). Annual wet deposition rates range about 6 kg S ha^{-1} in the source region to less than 1 kg S ha^{-1} at background sites. Biomass combustion has a controlling effect on rain acidity, given natural background conditions, and the contribution from fossil fuel combustion by industry is superimposed on this natural source of acidity. Some work has been done towards

understanding the impacts of wet deposition on forests, soils and surface waters over the eastern parts of South Africa (Olbrich and Du Toit, 1993; Olbrich et al., 1994). Recently, deposition work in South Africa has focussed on estimating dry deposition rates for sulphur (Zunckel et al., 1996,1999; Zunckel, 1999). One of these studies (Zunckel et al., 1996), based on two-week summer and winter measurement campaigns, indicated that dry deposition rates for sulphur exceed wet deposition in the relatively dry South African highveld climate in a ratio of approximately 60 : 40. Annual dry deposition rates were shown to decrease rapidly from a maximum of more that 13 kg S ha^{-1} on the central highveld to about 4 kg S ha^{-1} and less on the highveld periphery and beyond (Zunckel, 1999).

Given the relatively high emission of sulphur from South Africa and the inhibiting dispersion climatology, it is important to quantify deposition of pollutants on a regional scale in order to begin to address their impacts. In this paper, dry deposition rates of sulphur resulting from emissions on the industrialised highveld region of South Africa are estimated using the multi-scale atmospheric transport and chemistry (MATCH) modelling system, developed at the Swedish Meteorological and Hydrological Institute (SMHI) (Robertson et al., 1999). Similar studies using MATCH have been conducted in other regions including Sweden (Langner et al., 1995), Asia (Robertson et al., 1995) and over the African and South American continents (Robertson, 1996). In the absence of a major network of direct measurements of dry deposition, regional-scale dry deposition estimates from MATCH are compared at five sites to those derived previously from the inferential method of Hicks et al. (1987,1991), by Zunckel et al. (1999) and Zunckel (1999). MATCH-modelled wet deposition rates are compared with network measurements and region-scale estimates of total sulphur deposition over southern Africa.

2. Methodology and data

MATCH is an Eulerian multi-layered three-dimensional model that includes horizontal and vertical transport, vertical diffusion, dry deposition, wet scavenging and chemical transformations (Robertson *et al.*, 1999). Emissions information and meteorology are input requirements for modelling transport and chemical transformations to estimate ambient SO_2 and SO_4^{2-} concentrations. In turn, the modelled ambient concentrations are used together with assumed deposition velocities and scavenging rates to estimate dry and wet deposition fluxes of SO_2 and SO_4^{2-}, respectively.

The dry deposition rate is the lower boundary condition of the vertical diffusion scheme. The MATCH model uses reference dry deposition rates, valid at 1 m (Table 1), with an assumed diurnal variation of the uptake of SO_2

Table 1
Deposition velocities, cm s^{-1}, used in the MATCH model. vd$_{min}$ is the night time dry deposition velocity and vd$_{min}$ + vd$_{amp}$ is the maximum value daytime. The solar height is used to make a smooth transition in between these two extremes. Note that the deposition velocity of sulphate has no diurnal variation

Surface type	SO_2		SO_4^{2-}
	vd$_{min}$	vd$_{amp}$	vd
Sea	0.8	0.0	0.05
Land	0.3	0.5	0.01

by vegetation during daytime (Eq. (1)). No inter-annual variation due to vegetation seasons is assumed. Atmospheric stability is accounted for by a vertical diffusion scheme and a sub-module for the effective dry deposition rate in the lowest model layer (− 65 m depth) (Robertson et al., 1999). The diurnal variation of the reference dry deposition rate of SO_2 is determined from

$$vd_{ref} = vd_{min} + vd_{amp} f, \qquad (1)$$

f = max (solar elevation/solar elevation at noon, 0), which then roughly accounts for the sensitivity to uptake by vegetation during daytime. vd$_{min}$ is the nighttime dry deposition velocity and vd$_{min}$ + vd$_{amp}$ is the maximum daytime value. Dry deposition of SO_4^{2-} is not assumed to have any diurnal variation. The wet deposition flux is given by the product of the scavenging coefficient, the precipitation intensity (mm h^{-1}) and the ambient concentration of the pollutant. Scavenging coefficients applied are 2.8×10^{-4} h mm^{-1} s^{-1} for SO_4^{2-} and 1.0×10^{-4} h mm^{-1} s^{-1} for SO_2, respectively. The latter includes an estimate of the oxidation of SO_2 to SO_4^{2-} in rain droplets.

The chemistry scheme is a linear transformation from SO_2 to SO_4^{2-},

$$dC(SO_2)/dt = - kCSO_2, \qquad dC(SO_4^{2-})/dt = kC(SO_2),$$

the transformation rate k of concentration C is prescribed to vary with latitude and season. It has a maximum and constant value 4.0×10^{-6} s^{-1} at the equator, while it is given a sinusoidal variation with the season at the south pole between 0.2×10^{-6} s^{-1} at the southern hemisphere winter solstice and 2.4×10^{-6} s^{-1} at the corresponding summer solstice. In areas between the equator and the south pole the transformation rate varies linearly with the distance from the equator (Tarrason and Iverson, 1998). The adopted values of k implicitly account for oxidation of SO_2 to SO_4^{2-} in cloud-free air and in non-precipitating cloud water.
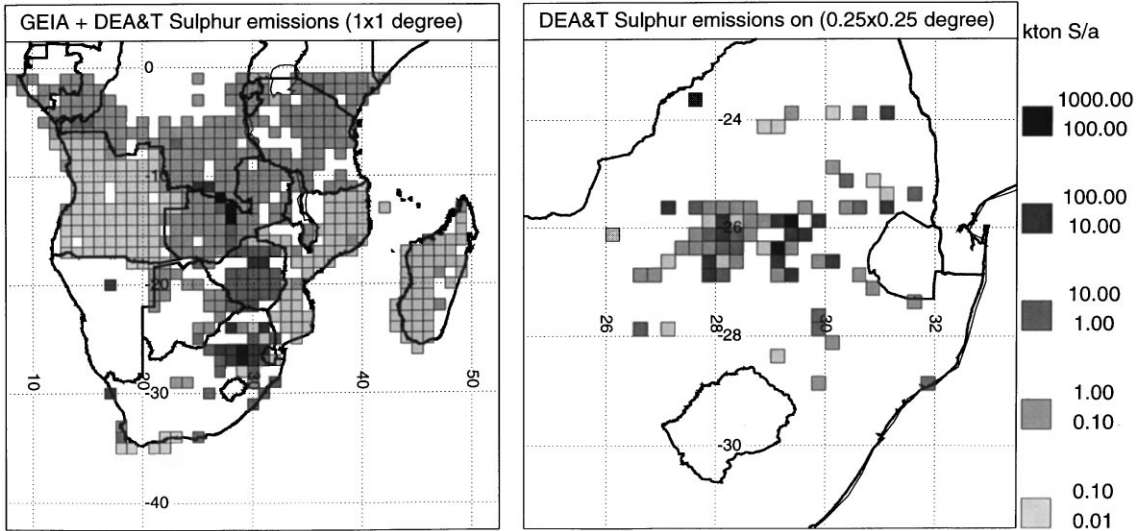
Fig. 2. Sulphur emission rates in kton a^{-1} for (left) southern Africa for the GEIA emissions on a $0.1° \times 0.1°$ grid, (right) highveld emissions from DEA and T on a $0.25_- \times 0.25_-$ grid.

The MATCH modelling domain is setup for $1° \times 1°$ and $0.25° \times 0.25°$ resolution grids extending over southern Africa south of the equator. Two emission data sets are used to calculate SO_2 concentrations and deposition rates for each grid. The first is the Global Emissions Inventory Activity (GEIA) $1° \times 1°$ emission inventory (Benkovitz, 1993), valid for 1990 and covers southern Africa south of the equator (Fig. 2 left). The second inventory is provided by the South African Department of Environment Affairs and Tourism (DEAT) (Wells et al., 1996) for 1993 (Fig. 2 right). The latter includes industrial emissions from the South African highveld defined from $26°$ to $32°E$ and $23°$ to $29°S$ in $0.25° \times 0.25°$ grid squares. The annual sulphur emission rate from southern Africa, excluding emissions from the highveld, is 1414 kton S a^{-1}. The highveld emissions account for an additional 884 kton S a^{-1}. Highveld emissions account for nearly 40% of all Africa's emissions south of the equator. In all cases the emissions are set initially as 95% of SO_2 and 5% of SO_4^{2-}.

The inferential model (Hicks et al., 1987,1991) uses a knowledge on dry deposition processes to infer dry deposition fluxes from routinely measured air pollutant concentrations and meteorological parameters. The dry deposition velocity is derived using a multiple-resistance transfer model where major resistances to atmosphere-surface exchange are modelled to infer the deposition velocity. The major resistances are the aerodynamic resistance which is determined by properties such as turbulent mixing and buoyancy, the quasi-laminar boundary layer resistance that accounts for molecular diffusivity in the vicinity of the receptor surfaces, and the surface or canopy resistance. This last resistance extends the resistance of a single leaf to represent the vegetation canopy. Deposition flux estimations made on a routine basis using the inferential technique are tested in two networks in the United States (Meyers and Sisterton, 1991; Clarke et al., 1997). In South Africa (Zunckel et al., 1996,1999; Zunckel, 1999), continuous measurements of SO_2 were used to calculate hourly average concentrations, while particulate samples were collected on open faced stacked-filter units (SFU). The SFU consisted of two equilibrated Nuclepore filters, a 8.0 μm filter in first stage and a 0.4 μm filter, to collect coarse and fine particulate matter, respectively. Separate filter units were run for the convective daytime (0600–1800), and the stable nighttime (1800–0600) periods.

Differences in the estimated dry deposition rates for sulphur between the inferential and MATCH modelling are to be expected as a consequence of basic differences in modelling approach. MATCH assumes two land cover types for the modelling domain (Table 1), namely land and sea. However, the roughness structure over the subcontinent partly accounts for the various vegetation types. MATCH assumes no change in land cover with seasons. Seasonal variation in the land cover is considered in the inferential model and the vegetation cover is specified in detail as combinations of grassland, maize cultivation, bushveld and forest. The diurnal and seasonal variation in modelled effective deposition velocity by MATCH are derived from atmospheric stability. The inferential technique estimates the resistance to transfer of the pollutant from the atmosphere based on in situ meteorological measurements, surface characteristics
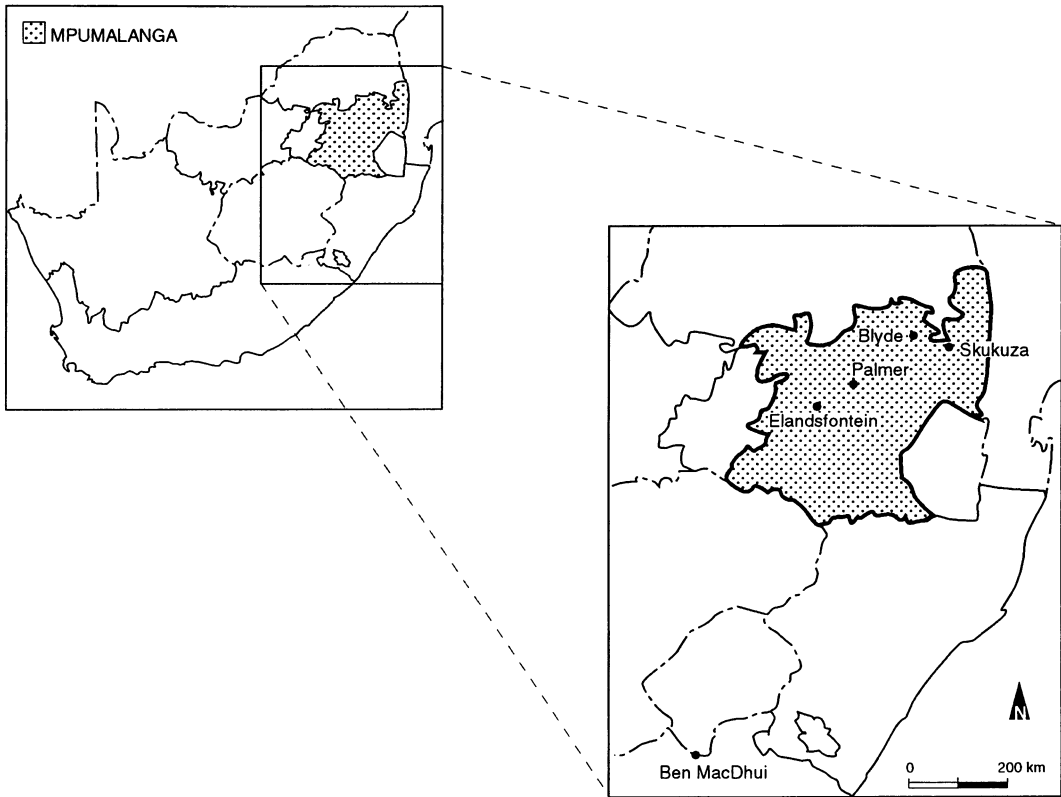
Fig. 3. Southern Africa showing the relative positions of the four field monitoring sites in the Mpumalanga province, and the remote site at Ben MacDhui.

and plant physiology to infer the deposition velocity. As a result, the inferred deposition velocity takes daily and seasonal meteorological and plant physiological variations into account. On summer days over the highveld the inferred dry deposition velocities for SO_2 range from 0.45 to 0.2 cm s^{-1}, while they approach zero under stable nighttime conditions throughout the year (Zunckel, 1999). The inferential technique estimates dry deposition fluxes at a specific location while MATCH provides a spatially continuous estimation of deposition fluxes. Although the monitoring equipment for single sites employing the inferential approach is relatively inexpensive, the costs of a monitoring network that will provide spatially continuous information are prohibitive. A few selected and representative monitoring sites in support of a spatial model is the ideal, cost effective way to obtain regional-scale results.

Three different model simulations are performed to derive SO_2 concentrations, dry and wet deposition rates and the relative loading from the highveld emissions. These are for southern Africa on the $1° × 1°$ grid omitting the major sources on the highveld, secondly for highveld emissions only on the $1° × 1°$ grid, and thirdly for the

highveld emissions only on the $0.25° × 0.25°$ grid. This enables evaluation of the relative impact from the highveld emissions on one hand and the resolution in the emission data on the other. Meteorological data for the MATCH modelling are derived from European Centre of Medium Range Forecasts (ECMWF) global analyses for five selected periods, namely 1 January–10 February 1994, 1 April–1 May 1994, 1 July–12 August 1994, 1 October–1 November 1994, as well as 1 January–1 February 1995. MATCH interpolates the ECMWF input fields a 1 h time resolution. The modelling periods are selected to allow comparison with an intensive two-week winter and summer field monitoring programme in July and August 1994 and in January 1995 at Elandsfontein on the central industrialised highveld of South Africa's Mpumalanga province (Fig. 3) where data are collected as input to the inferential model (Zunckel et al., 1996). Results from similar monitoring campaigns in Mpumalanga between June 1996 and May 1997 (Zunckel, 1999) and at a remote high-altitude site near the South African border with Lesotho in March and June 1996 (Zunckel et al., 1999) are also used. Atmospheric emissions from the major industries on the highveld are

relatively constant over time (Lloyd, 1997) and comparison of MATCH results with the later field experiments are representative. A monitoring network for acid rain is in operation over the northeastern parts of South Africa, centered on the highveld. Results that span up to a 10 yr period (Bluff et al., 1991; Turner et al., 1996) are compared with MATCH wet deposition predictions.

3. Results

3.1. Ambient SO₂ concentrations

The MATCH-modelled mean SO_2 concentrations over southern Africa are based on the GEIA and DEAT emissions on the $1° \times 1°$ grid reveal high concentrations extending from northern Zambia to the southeast coast of South Africa (Fig. 4 left). Two maxima are observed: one that is a result of industrial activity on the highveld of South Africa and the second further north over the mining areas of northern Zambia and the southern Congo. In the model run for the high-resolution distribution of SO_2 from the highveld of South Africa alone (Fig. 4 right) mean SO_2 concentrations over the central highveld range between 3 and 10 ppb and exceed 10 ppb near source areas. Further afield, e.g., over Lesotho and areas to the south and east concentrations decrease to less than 1 ppb. The modelled ambient SO_2 concentrations compare reasonably well with measured values. For a five-year data set, Turner (1990) reported mean ambient SO_2 values of 10 ppb on the central highveld that decrease to 4 ppb and lower on its periphery. Zunckel et al. (1999) measured SO_2 concentrations of less than 1 ppb at the high-altitude remote site at Ben MacDhui (3001 m ASL) in South Africa near the southeast border with Lesotho, ~ 600 km direct distance from the highveld industrial heartland of South Africa. The modelled SO_2 distribution from highveld emissions only is approximately concentric around the predominant anticyclone and is consistent with the observed transport patterns over the subcontinent given in Fig. 1.

3.2. Dry deposition

Dry deposition rates for sulphur are calculated by MATCH using the combined $1° \times 1°$ GEIA and DEAT emission data for southern Africa, the predicated SO_2 concentrations and assumed deposition velocities that are a reflection of the underlying surface (Table 1). They exhibit a similar spatial pattern to that of ambient SO_2 concentrations. Maxima of more than 10 kg S ha^{-1} a^{-1} are deposited over the highveld and northern Zambia (Fig. 5 left). Much of the southern African subcontinent experiences dry sulphur deposition rates of 0.1–0.5 kg S ha^{-1} a^{-1}. Deposition for highveld-only $0.25° \times 0.25°$ emissions (Fig. 5 right) exceeds 0.3 kg S ha^{-1} a^{-1} over most of southern Africa south of $\sim 18°$S. As with the coarser-resolution simulation, a core of maximum deposition rates in excess of 1 kg S ha^{-1} a^{-1} occur over the central highveld.
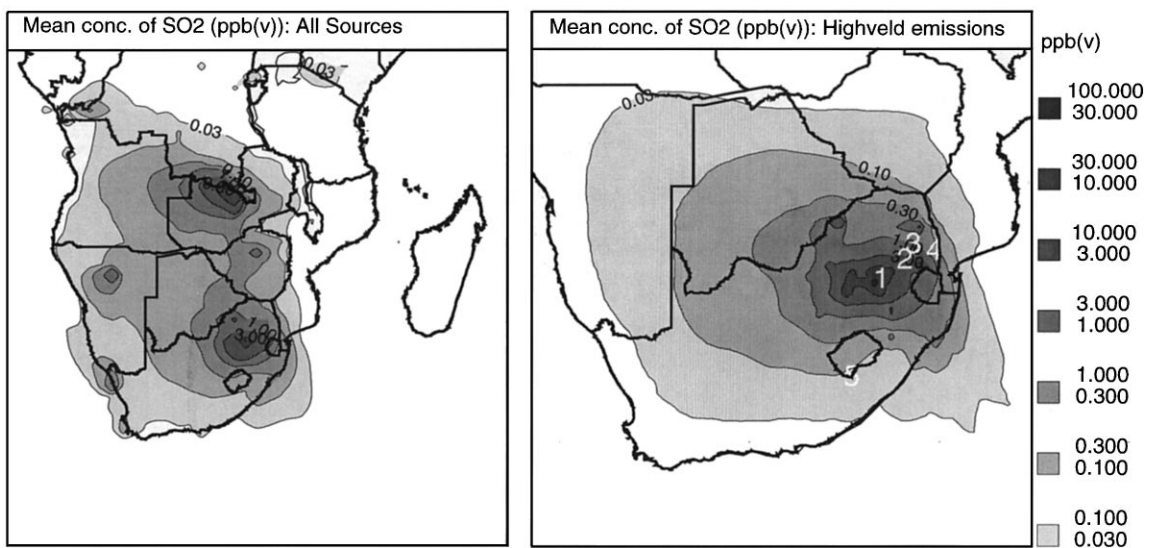

Fig. 4. MATCH-modelled annual ambient SO_2 concentrations in ppb for southern Africa (left) using total emissions from the combined GEIA and DEA and T emission data (on the $1° \times 1°$ grid) and (right) based on highveld-only emission (on the $0.25° \times 0.25°$ grid). The position of the five field stations are indicated as (1) Elandsfontein, (2) Palmer, (3) Blyde, (4) Skukuza, and (5) Ben MacDhui.
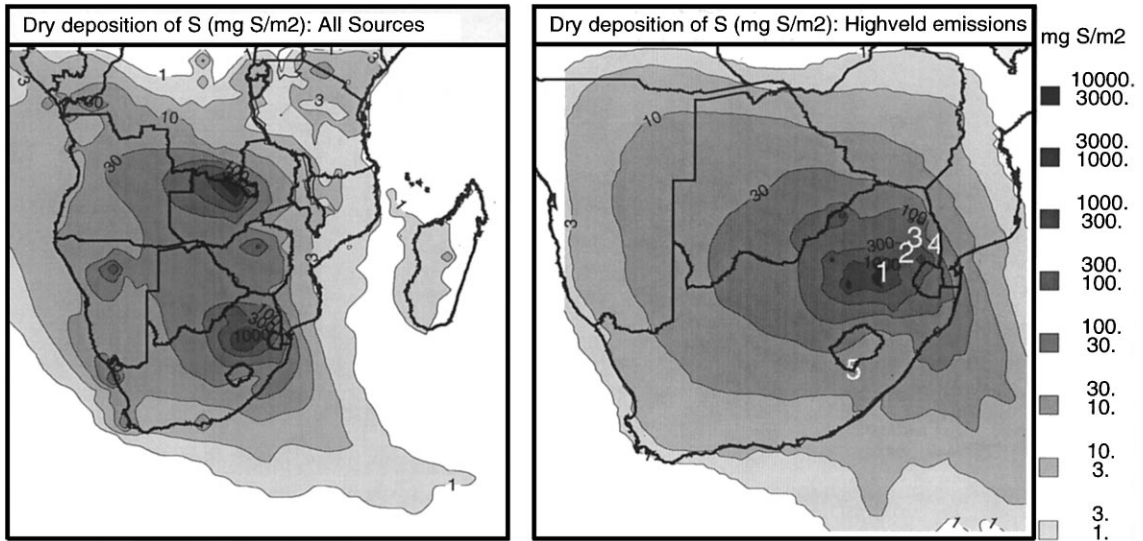
Fig. 5. MATCH modelled annual dry deposition rates for sulphur in mg m^{-2} for southern Africa (left) using combined GEIA and DEA and T emissions data (on the $1° \times 1°$ grid), (right) based on highveld-only emission (only on the $0.25° \times 0.25°$ grid). The positions of the five monitoring sites where the inferential model was applied are indicated on b.

Dry deposition rates for sulphur are available for five stations in South Africa (Figs. 3 and 5 right) from inferential modelling (Zunckel, 1999; Zunckel et al., 1999). These modelling studies found that dry deposition exceeded 13 kg S ha^{-1} a^{-1} at Elandsfontein in the central highveld region, but decreased to about 4 k S ha^{-1} a^{-1} the east over the eastern escarpment (at Palmer and Blyde Forest Station) and the lowveld of South Africa (at Skukuza). At the remote Ben MacDhui site, situated more than 1000 km away from the source region along the most frequent anticyclonic transport pathway, dry deposition is about 1 kg S ha^{-1} a^{-1}. The relatively flat highveld area is well represented by Elandsfontein and Palmer from a climatological perspective. Elandsfontein is located in the centre of the highveld's major industrial activity and is in close proximity to a number of significant point sources. From an air quality perspective it represents the upper extreme for the highveld. Palmer, on the other hand, is located on the highveld periphery some 100 km to the east of the major source region.

The MATCH model was run with the meteorological data for the same period as the inferential modelling. Dry deposition rates for sulphur were simulated for the five inferential sites using both the $1° \times 1°$ and $0.25° \times 0.25°$ emission data. The gradient in dry deposition rates across the highveld of South Africa is modelled well by MATCH. Maximum deposition rates at Elandsfontein decrease to approximately 25% of the maximum on the edge of the highveld at Palmer (Table 2). The weak

Table 2
Annual estimated dry deposition for total sulphur from the inferential method and MATCH modelling in kg ha^{-1}. The values in brackets are the percentage difference between the inferential and the MATCH simulations

Station	Inferential method	MATCH $1° \times 1°$	MATCH $0.25° \times 0.25°$
Elandsfontein	13.1	19.2 (47)	44.0 (236)
Palmer	3.1	5.8 (87)	5.1 (64)
Blyde	3.9	2.3 (-41)	1.8 (-54)
Skukuza	3.3	1.3 (-60)	1.2 (-64)
Ben MacDhui	1.1	0.3 (-73)	0.2 (-81)

gradient further eastward from the Blyde Forest Station to Skukuza is also well simulated. In the source region, MATCH overestimates dry deposition rates of total sulphur by nearly 50% on the $1° \times 1°$ grid and by more than 200% on the $0.25° \times 0.25°$ degree grid (Table 2). The model also overestimates deposition rates on the edge of the highveld at Palmer. The modelled dry deposition rates for sulphur from SO$_2$ and particulate sulphate (SO$_4^{2-}$) are generally underestimated away from the source region (Table 3). In general the simulation using the $1° \times 1°$ emission-data approximates the inferential dry deposition estimates better than the finer-resolution simulation.

The general overestimation of deposition by MATCH could be attributed to the assumed minimum deposition

Table 3
Annual estimated dry deposition of sulphur from SO_2 and SO_4^{2-} from the inferential method and MATCH modelling in kg ha^{-1}. The values in brackets are the percentage difference between the inferential and the MATCH simulations

Station		Inferential method	MATCH $1° \times 1°$	MATCH $0.25° \times 0.25°$
	SO_2	10.9	18.5 (169)	42.7 (291)
Elandsfontein	SO_4^{2-}	2.2	0.85 (-61)	1.35 (-39)
	SO_2	2.7	5.4 (200)	4.7 (74)
Palmer	SO_4^{2-}	0.4	0.4 (0)	0.4 (0)
	SO_2	3.3	2.0 (-29)	0.2 (-52)
Blyde	SO_4^{2-}	0.6	0.85 (42)	0.1 (-65)
	SO_2	3.0	1.1 (-63)	1.1 (-63)
Skukuza	SO_4^{2-}	0.3	0.2 (-29)	0.1 (-67)
	SO_2	1.0	0.19 (-81)	0.14 (-86)
Ben MacDhui	SO_4^{2-}	0.1	0.1 (0)	0.08 (-20)

velocity of 0.3 cm s^{-1} for all land surface, compared with the near zero deposition velocities that occur at night throughout the year (Zunckel et al., 1996; Zunckel, 1999). The assumptions that are inherent in any grid-based modelling approach with respect to topography, land use and boundary conditions may also contribute to the observed differences. Despite the significant differences in modelling approaches reveal the dry deposition of sulphur from SO_2 exceeds that from SO_4^{2-} at all the sites. Deposition of sulphur from SO_4^{2-} is fairly well modelled on the highveld at Elandsfontein and Palmer. This implies that the general overestimation of dry sulphur deposition by MATCH on the highveld is due to an overestimation of deposition from SO_2. Despite the significant differences in modelling approaches employed in the two models, the estimates they provide of dry deposition are in reasonable agreement.

3.3. Wet deposition

MATCH-modelled wet deposition rates for sulphur using the $0.25° \times 0.25°$ emissions data reveal concentrations generally exceeding 1 kg S ha^{-1} a^{-1} over the entire eastern half of South Africa, southern Mozambique, northern Zimbabwe, Botswana and Zambia (Fig. 6 top left). The maximum modelled wet deposition rates occur over the eastern parts of South African and exceed 3 kg S ha^{-1} a^{-1}, reaching 10 kg S ha^{-1} a^{-1} over the highveld and its periphery. In comparison, for the 5-yr period 1985–1990, Bluff et al. (1991) report annual wet deposition rates determined from an acid rain network of 5.8–4.7 kg S ha^{-1} for the central highveld and the periphery 150 km to the southeast. At the background site of Louis Trichardt, near the South Africa/Zimbabwe border and 300 km to the north of the major industrial source area of SO_2, wet deposition rates of 1.3 kg S ha^{-1} a^{-1} are observed (Bluff et al., 1991). Over the drier western part of South Africa the modelled wet deposition ranges from 0.3 to 1.0 kg S ha^{-1} a^{-1} and are lower along the west coast.

Differences between observed and modelled wet deposition rates are likely a result of models generally over predicting the number of rain events over the southern African region, particularly in the east (Joubert, 1998; Joubert et al., 1998). In addition, the limited horizontal resolution of the model produces an averaging effect for the isolated convective precipitation over much of southern Africa. Individual elements of heavier precipitation are smoothed out into more widespread and lighter precipitation fields. This, in turn, results in efficient extraction of pollutants by the model scavenging process. Instead of complete removal over a small area by convective rain, there is opportunity for removal over a much broader area.

3.4. Total sulphur deposition

The MATCH-modelled deposition rates for total sulphur reveal an expanded spatial pattern to that of wet deposition (Fig. 6 right). Deposition rates in excess of 1 kg ha^{-1} a^{-1} occur over the entire northeastern parts of South Africa. A broad band of relatively high deposition rates extends northeastwards over western Botswana, much of Zimbabwe and into Zambia and Angola. Maxima over the highveld and northern Zambia exceed 30 kg S ha^{-1} a^{-1}. By extrapolating inferential results from short field campaigns on the central highveld, Zunckel et al. (1996) found that dry deposition of sulphur could exceed that from wet deposition in a ratio 60 : 40. The MATCH model suggests that wet deposition rates are similar to dry deposition on the highveld and the adjacent areas (Fig. 6, left). Over the sparsely vegetated drier western half of South Africa MATCH-modelled wet deposition exceeds dry deposition, but these rates are relatively low. In the higher rainfall regions along the south and east coasts wet deposition exceeds dry.
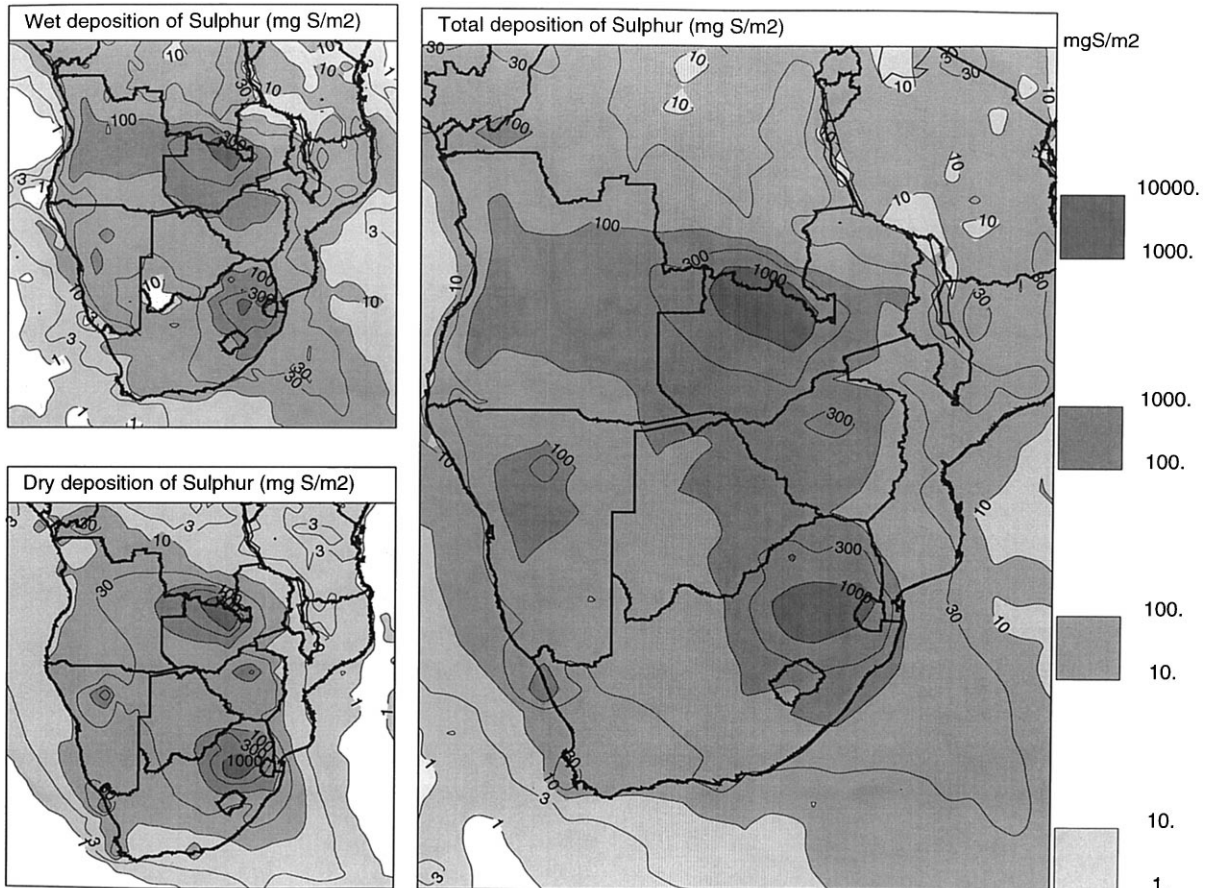
Fig. 6. MATCH-modelled annual wet deposition (top left), dry deposition (bottom left), and total sulphur deposition (right) in mg m^{-2} for the $1° \times 1°$ GEIA emission data.

Deposition rates of 0.3–1.0 kg S ha^{-1} a^{-1} are modelled from the highveld-only emissions over much of Botswana, the southern half Zimbabwe and Mozambique. Some sulphur deposition from South African emissions is apparent as far north as Angola and Zambia. The sulphur transport plume extending southeastward to the coast out into the Indian Ocean off the east coast of South Africa is clearly evident (Fig. 7) and compares with similar ozone (Fishman et al., 1991), carbon dioxide (Rayner and Law, 1995) and dust (Herman et al., 1995; Tyson and D'Abreton, 1998) plumes both observed and modelled. These plumes may extend to Australasia (Rayner and Law, 1995; Herman et al., 1995; Sturman et al., 1997). It is instructive to assess the relative concentration of South African highveld SO$_2$ emissions to the total sulphur deposition of southern Africa as a whole (Fig. 7). Except in the western, southwestern and southern regions, the highveld emissions contribute to more

than 80% to the total deposition over the whole of South Africa. Near the source region the percentage is considerably higher. Highveld emissions are also responsible for a high contribution to total sulphur deposition over the southern parts of Botswana, Zimbabwe and Mozambique. To the north the highveld contribution to sulphur deposition decreases rapidly.

4. Conclusion

Regional-scale ambient SO$_2$ concentrations and dry deposition rates of sulphur are estimated for southern Africa for the first time using the multi-scale atmospheric transport and chemistry (MATCH) modelling system. The model output is compared to estimations of dry deposition of sulphur using an inferential
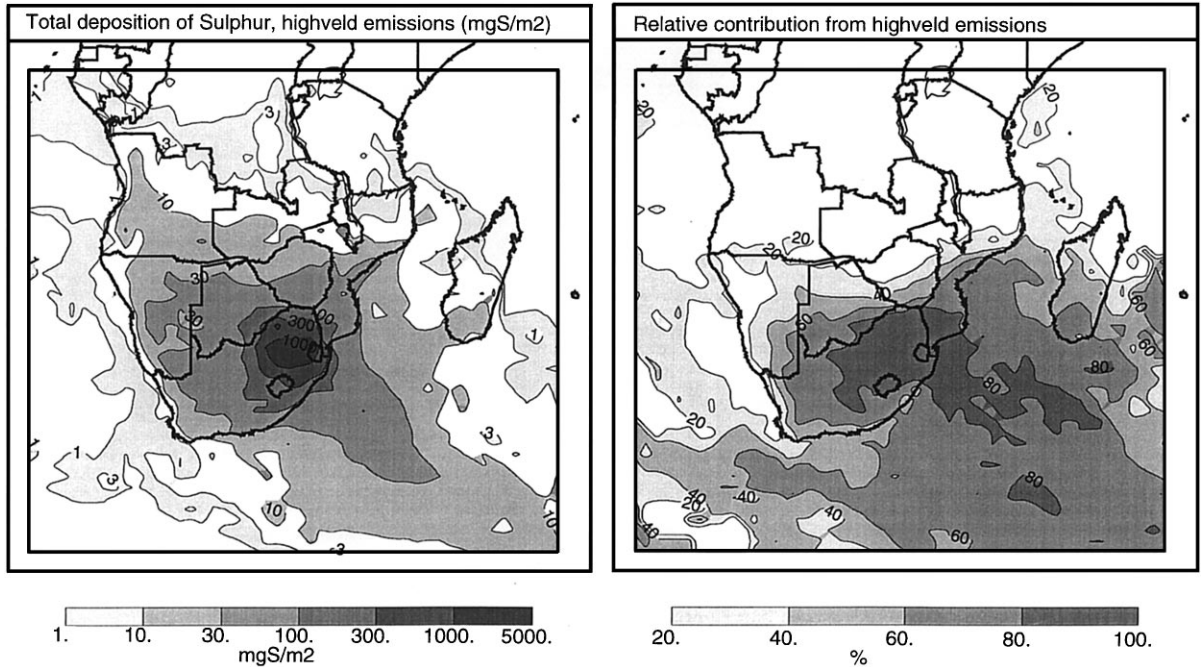
Fig. 7. (left) Total MATCH-modelled annual sulphur deposition in mg m^{-2} from highveld-only emissions, and (right) the relative contribution from highveld-only emissions to total sulphur deposition as a percentage.

model and to observations of both dry and wet deposition of the element. Despite the significant differences in modelling approaches, the MATCH and inferential model estimates of dry deposition are in reasonable agreement. Furthermore, MATCH-modelled wet deposition rates compare favourably with network measurements.

Spatial patterns of modelled SO_2 concentrations are consistent with the atmospheric transport modes prevailing over the subcontinent. They decrease from maximum concentrations of between 3 and 10 ppb over the central highveld to less than 1 ppb 600 km from of this industrial source region. MATCH-modelled dry deposition rates for sulphur exhibit a similar spatial pattern to the modelled ambient SO_2 concentrations. Maximum deposition exceeds 10 kg S ha^{-1} a^{-1} over the central highveld. Much of the southern African subcontinent experiences dry sulphur deposition rates of more than 0.3 kg S ha^{-1} a^{-1}. MATCH-modelled sulphur wet deposition rates reveal a spatial pattern that is generally consistent with the rainfall pattern over the subcontinent. Maximum values occur in the wetter east and minimum values are simulated in the west. Wet deposition exceeds 1.0 kg S m^{-1} a^{-1} over the entire eastern half of South Africa with the maximum modelled wet deposition rates over the South African highveld and its periphery of more than 10 kg S ha^{-1} a^{-1} in places. Over the drier

western part of South Africa the modelled wet deposition rates range from 0.1 to 0.5 kg S ha^{-1} a^{-1}.

South African emissions contribute more than 80 per cent of the total sulphur loading in the plume of material being transported off the subcontinent over the Indian Ocean towards Australasia. This is expected. What is unexpected is the modelling suggests that up to 20% of the loading is transported from as far afield as the Zambian copperbelt.

Emissions of SO_2, from whatever sources, in all the countries of the region contribute to the overall pattern of regional deposition of sulphur over the subcontinent of southern Africa. The two most significant sources of emissions come from the Zambian/Congo copperbelt region and from the highveld industrial heartland of South Africa. Deposition from transboundary atmospheric transport of sulphur occurs throughout the region. Atmospheric recirculation with anticyclonic circulation systems, together with the atmospheric stability attendant on these systems, are the primary factors governing the build-up of the atmospheric sulphur loading over the subcontinent before the major transport from the region takes place in the plume to the Indian Ocean towards Australasia. This plume must be recognised as a major feature of the southern hemispheric atmospheric circulation and transport climatology.

Acknowledgements

References

Benkovitz, C.M., 1993. Third GEIA Workshop on Global Emission Inventories. Amersfoort, The Netherlands, 31 January–2 February.

Bluff E., Turner, C.R., de Beer, G.H., 1991. Rain chemistry: 1985 to 1991. Eskom Engineering Investigations Report TRR/S/91/016.

Clarke, J.F., Edgerton, E.S., Martin, B.E., 1997. Dry deposition calculations for clean air status and trends network. Atmospheric Environment 31 (21), 3667–3678.

Cosijn, C., Tyson, P.D., 1996. Stable discontinuities in the atmosphere over South Africa. South African Journal of Science 92, 381–386.

Fishman, J., Fakhruzzaman, K., Cros, B., Mganga, D., 1991. Identification of widespread pollution in the Southern Hemisphere deduced from satellite analysis. Science 252, 1693–1696.

Galpin, J.S., Turner, C.R., 1999a. Trends in rain quality data from the South African interior. South African Journal of Science 95, 223–225.

Galpin, J.S., Turner, C.R., 1999b. Trends in composition of rain quality data from the South African interior. South African Journal of Science 95, 225–228.

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 Geophysical Research 101 (D19), 23721–23736.

Herman, J.R., Bhartia, P.B., Torres, O., Hsu, C., Seftor, C., Celarier, E., 1995. Global distribution of UV-absorbing aerosols from Nimbus-7/TOMS data. Journal Geophysical Research 102, 16911–16922.

Hicks, B.B., Baldocchi, D.D., Meyers, T.P., Hosker, Jr. R.P., Matt, D.R., 1987. A preliminary multiple resistance routine for deriving dry deposition velocities from measured quantities. Water, Air and Soil Pollution 36, 311–330.

Hicks, B.B., Hosker, R.P., Meyers, T.P., Womack, J.D., 1991. Dry deposition inferential measurements technique – I. Design and test of a prototype meteorological and chemical system for determining dry deposition. Atmospheric Environment 25A (10), 2345–2359.

Joubert, A.M., 1998. Forecasting rainfall and streamflow over South and southern Africa. Eskom Research Report, TRR/T98/046, 52pp.

Joubert, A.M., Katzfey, J.J., McGregor, J.L., Nguyen, K.C., 1998. Simulating mid-summer climate over southern Africa using a nested regional climate model. Journal of Geophysical Research, in press.

Langner, J., Persson, C., Robertson, L., 1995. Concentrations and deposition of acidifying air pollutants over Sweden: estimates for 1991 based on the MATCH model and observations. Water, Air and Soil Pollution 85, 2021–2026.

Lloyd, S.M., 1997. South African Department of Environment Affairs and Tourism, personal communication.

Meyers, T.P., Sisterton, D.L., 1991. Network measurements of dry deposition of atmospheric pollutants. NAPAP State of Science and Technology 1, 6-223–6-249.

Olbrich, K.A., Du Toit, B., 1993. Assessing the risks posed by air pollution to forestry in the Eastern Transvaal, South Africa. CSIR Report FOR-C 214.

Olbrich, K.A., Skoroszewski, R., Taljaard, J., Zunckel, M., 1994. An acid deposition risk advisory system: a test study of Region F to define actual and critical loads, CSIR Internal report FOR-1.

Preston-Whyte, R.A., Tyson, P.D., 1988. The Atmosphere and Weather of Southern Africa, 1st Edition. Oxford University Press, Cape Town, pp. 374.

Rayner, P.J., Law, R.M., 1995. A comparison of modelled responses to prescribed CO_2 sources. CSIRO Australia, Division of Atmospheric Research, Technical Paper No. 36.

Robertson, L., 1996. Modelling of anthropogenic sulfur deposition to the African and South American continents. SMHI Report No. 73, Norrköping.

Robertson, L., Rodhe, H., Granat, L., 1995. Modelling of sulfur deposition in the southern Asian region. Water, Air and Soil Pollution 85, 2337–2347.

Robertson, L., Langner, J., Engardt, M., 1999. An Eulerian limited area transport model. Journal Applied Meteorlogy 38 (2), 190–210.

Sivertsen, B., Matale, C., Pereira, L.M.R., 1995. Sulphur emissions and transfrontier air pollution in Southern Africa. Southern African Development Community, Environmental and Land Management Sector, Report Series 34.

Sturman, A.P., Tyson, P.D., D'Abreton, P.C., 1997. A preliminary study of the transport of air from Africa and Australia to New Zealand. Journal of the Royal Society of New Zealand. 27(4), 485–498.

Tarrason, L., Iverson, T., 1998. Modelling intercontinental transport of atmospheric sulphur in the northern hemisphere. Tellus 50B, 331–352.

Turner, C.R., 1990. A five year study of air quality in the Highveld region, Eskom Report TRR/S090/002, Eskom TRI, Johannesburg.

Turner, C.R., Wells, R.B., Olbrich, K.A., 1996. Deposition chemistry in South Africa, 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.

Tyson, P.D., D'Abreton, P.C., 1998. Transport and recirculation of aerosols off southern Africa – Macroscale plume structure. Atmospheric Environment 32 (9), 1511–1524.

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

Wells, R.B., Lloyd, S.M., Turner, C.R., 1996. National air pollution source inventory (inventory of scheduled processes). In: Held, G., Gore, B.J., Surridge, A.D., Tosen, G.R., Turner, C.R., Walmsley, R.D. (Eds.), 1996. Air Pollution and its

Impacts on the South African Highveld. Environmental Scientific Association, Cleveland.

Zunckel, M., 1999. Dry deposition of sulphur over eastern south Africa. Atmospheric Environment 33, 3515–3529.

Zunckel, M., Turner, C.R., Wells, R.B., 1996. Dry deposition of sulphur on the Mpumalanga highveld: a pilot study using the inferential method. South Africa Journal of Science 92, 485–491.

Zunckel, M., Piketh, S. and Freiman, T., 1999. Dry deposition of sulphur at a high-altitude background site in South Africa. Water, Air and Soil Pollution 115, 445–463.