DRY DEPOSITION OF SULPHUR AT A HIGH-ALTITUDE BACKGROUND STATION IN SOUTH AFRICA

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(Received 28 October 1997; accepted 4 November 1998)

Abstract. The inferential method is applied to estimate dry deposition rates of sulphur at a remote rural site on the South African south-eastern escarpment. Two 2-week intensive monitoring campaigns were conducted during the Ben MacDhui High Altitude Trace Gas and Transport Experiment (BHATTEX) when SO_2, particulates and meteorological data were collected. Total sulphur deposition rates of 4.9 mg m^{-2} in summer exceeds the winter value of 3.0 mg m^{-2}. The annual dry deposition rate for sulphur at the remote site of 1.1 kg ha^{-1} is about 13% of that occurring in the main source region, approximately 1000 km away along the most frequent transport pathway. Deposition of total sulphur on the south-eastern escarpment is a consequence of this large-scale regional transport of aerosols and trace gases over southern Africa.

Keywords: background, dry deposition, high-altitude, long-range transport, South Africa, sulphur

1. Introduction

Wet deposition of air pollution in South Africa has been studied previously (Turner, 1993). Work has focussed mainly on the industrialised Mpumalanga Highveld where approximately 47% of Africa's electrical energy is generated in 10 massive coal-fired power stations generating in excess of 24 GW. The Highveld region is responsible for 86% of particulates and 94% of all South Africa's SO_2 emissions (Wells *et al*., 1996). The possible significance of dry deposition in the largely arid South African climate has been alluded to in various reports (Wells *et al*., 1987; Tyson *et al*., 1988; Wells, 1989; Turner, 1993; Wells, 1993), but little studied, despite South Africa's interior plateau experiencing an average of only 60 rain days per annum.

Wells (1993) used long-term SO_2 concentrations for the central Mpumalanga Highveld region (Bluff *et al*., 1990) and suggested that sulphur deposition from dry deposition processes could range from one half to several times the value of that from wet deposition. Dry deposition has been measured in Mpumalanga by measuring canopy fall through (Olbrich and Du Toit, 1993). Piketh and Annegarn (1994) measured SO_4^{2-} concentrations in South Africa's Lowveld and escarpment region. By assuming a range of deposition velocities they estimated dry deposition rates for sulphate ranging from 0.56 kg ha^{-1} a^{-1} for grasslands to 6.7 kg

Water, Air, and Soil Pollution **115:** 445–463, 1999.

ha^{-1} a^{-1} in pine forests. Skoroszewski (1994) used rock runoff plots, polyethylene bulk samplers and SO_2 concentrations to estimate dry sulphur deposition in the Suikerbosrand catchment study and found it to be 80% of that of the wet deposition.

In a pilot project, Zunckel *et al*. (1996) measured SO_2 and SO_4^{2-} concentrations and micro-meteorological parameters for use in the inferential model of Hicks *et al*. (1991) to infer deposition velocity (V_d) and the dry sulphur deposition flux at Elandsfontein on the industrialised central Mpumalanga Highveld. Dry sulphur deposition in this source region was found to be approximately 60% of the total sulphur deposition of 13.9 kg ha^{-1} a^{-1}. Approximately 80% of the dry deposition of sulphur occurred through SO_2 deposition during the convective period of the day. Much smaller contributions to total sulphur loading were made by particulates during both the day and night.

The *B*en MacDhui *H*igh *A*ltitude *T*race Gas and *T*ransport *EX*periment (BHATTEX) provided an ideal opportunity to estimate dry deposition rates of sulphur at a remote rural site far removed from the major source region of Mpumalanga. Observations were made on Ben MacDhui mountain, the highest point on the South African south-eastern escarpment (3001 m), in March and June 1996. The sampling site at Ben MacDhui intercepts the high-level plume of aerosols and trace gases that exit the African continent into the Indian Ocean via anticyclonic transport (Garstang *et al*., 1996; Tyson *et al*., 1996). The experiment is described fully in Piketh *et al*. (1998). In this paper, dry deposition of sulphur at the remote rural site is examined using the inferential method (Hicks *et al*., 1987, 1991). The role of synoptic-scale meteorology in causing day-to-day variability in transport from the industrial source regions is also considered.

2. Site Description

The southern Drakensberg mountains were identified by Garstang *et al*. (1996) and Tyson *et al*. (1996) using trajectory analysis to be situated in the plume of aerosols and trace gases that originate over the southern African interior and exit the continent into the Indian Ocean via anticyclonic transport (Figure 1, bottom left and right). The sampling site near the Tiffendale Ski Resort on Ben MacDhui mountain (30°32′S; 27°58′E) is situated in the Eastern Cape province near the Lesotho border in the Drakensberg mountains (Figure 2). At an altitude of approximately 2870 m, just below the top of Ben MacDhui, the sampling site intercepts the planetary boundary layer which is capped by absolutely stable layers at 700 and 500 hPa. These are persistent temporally and are ubiquitous over southern Africa (Cosijn and Tyson, 1996).

The sampling site is some 600 km to the south-southwest of South Africa's major industrial regions of Mpumalanga and Gauteng. Rhodes is the nearest town and is situated approximately 1300 m below the sampling site and 25 km to the south-southeast with a population of less than 500. The nearest cities are East

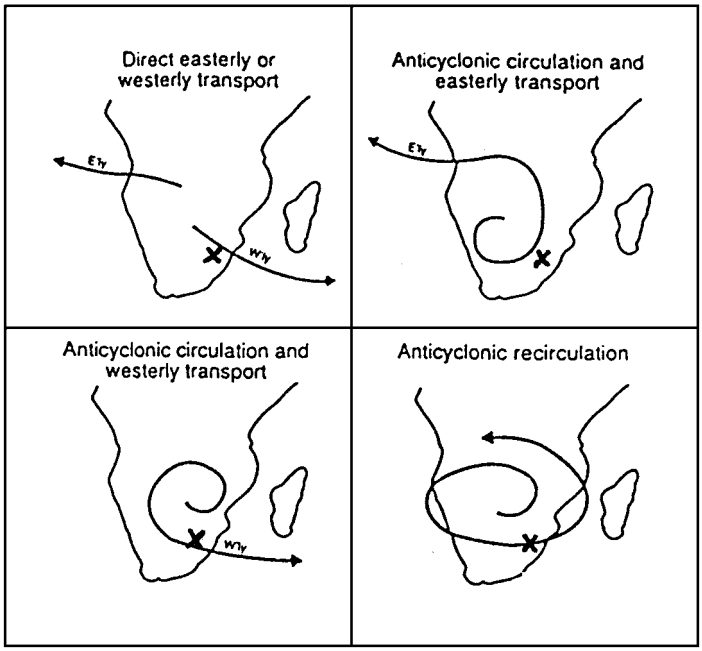

Figure 1. Schematic representation of major low-level transport trajectory modes likely to result in easterly or westerly exiting of material from southern Africa or in recirculation over the subcontinent (Garstang *et al*., 1996). The cross indicates the relative position of Ben MacDhui.

London and Port Elizabeth, situated approximately 300 and 400 km to the south and south-southwest, respectively. The site is thus ideally removed from potential industrial sources and local sources of contamination are limited.

The soils of the region are shallow and the vegetation is determined by the high altitude associated with low temperatures and snow in winter. The vegetation-region is defined as Alti Mountain Grassland that occurs between altitudes of 2500 and 3480 m on the steep, treeless, upper mountain region of Lesotho and the KwaZulu-Natal Drakensberg (Granger and Bredenkamp, 1996). It consists mainly of tussock grasses interspersed with sedges, ericoid or dwarf heath shrubs and creeping or mat-forming plants.

3. Data and Methodology

The factor that links the rate of dry deposition of a pollutant to the atmospheric concentrations is the deposition velocity (V_d), where

$$V_d = -F/C \tag{1}$$

F is the dry deposition rate and C the atmospheric concentration of the chemical species of interest. The negative sign conforms with the meteorological convention

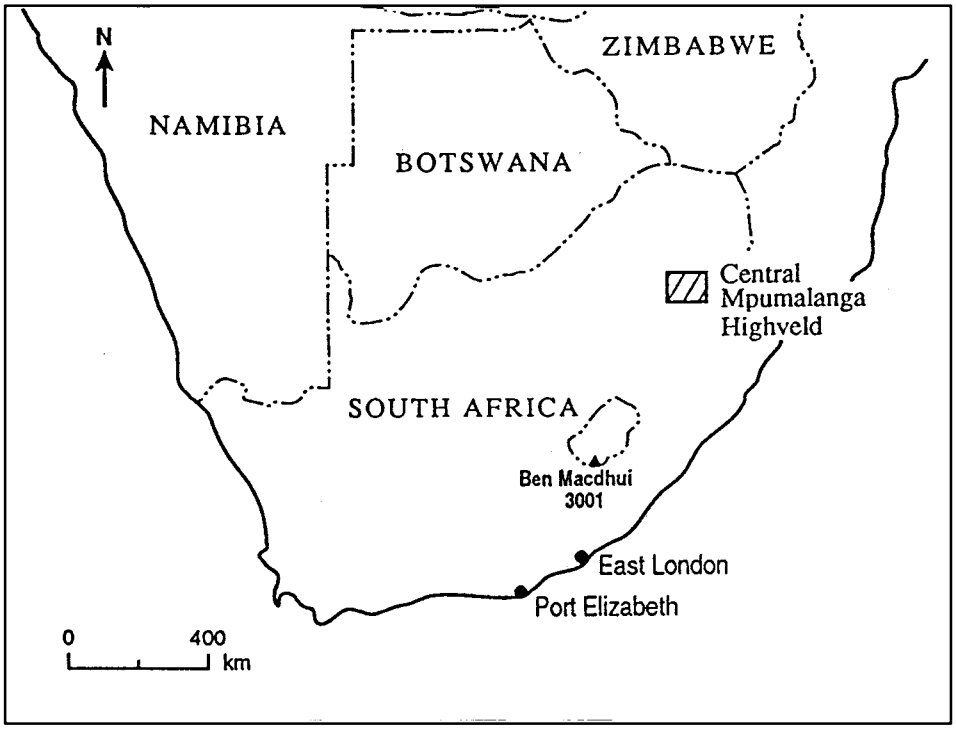

Figure 2. The position of Ben MacDhui relative to the industrialised central Mpumalanga highveld and the two coastal cities, East London and Port Elizabeth.

and indicates a downward flux. V_d is estimated with a technique referred to as the inferential method (Hicks *et al.*, 1987, 1991; Meyers *et al.*, 1991) developed by the National Oceanic and Atmospheric Administration's (NOAA) Atmospheric Turbulence and Diffusion Division (ATDD) and applied elsewhere to estimate site specific fluxes of sulphur dioxide (SO_2), ozone (O_3), nitric acid (HNO_3) and submicron particles (Meyers *et al.*, 1991; Meyers and Sisterton, 1991).

The inferential model simulates the resistance to transfer from the atmosphere through three resistance components acting in series: R_a is the aerodynamic resistance that is determined by atmospheric properties, predominantly turbulent exchange; R_b is the quasi-laminar boundary layer resistance that accounts for the molecular diffusivity affecting pollutant transfer in the vicinity of receptor surfaces; and R_c combines the consequences of all uptake processes involving individual leaf elements into a single number that is characteristic of the specific pollutant and the surface. The total resistance to transfer from the atmosphere is then

$$R = R_a + R_b + R_c \tag{2}$$

which leads to the relationship that

$$V_d = 1/(R_a + R_b + R_c) \tag{3}$$

R_a, R_b and R_c are estimated from routinely measured micro-meteorological parameters, a knowledge of the surface roughness and of the dominant vegetation. This makes the inferential method preferable over alternative high-cost and high-technology methods in routine monitoring and in projects such as BHATTEX.

The aerodynamic resistance, R_a, is controlled by atmospheric turbulence which is influenced by mechanical mixing and buoyancy. The standard deviation of wind direction (σ_θ) contains information on stability and surface roughness. When combined with measurements of mean wind speed it provides an estimate of R_a. The quasi-laminar boundary layer resistance, R_b, is the resistance in the layer in contact with the surface. Estimates of the friction velocity based on wind speed data and a knowledge of the surface roughness are used with the Schmidt number to evaluate the quasi-laminar transfer resistance. The surface or canopy resistance, R_c, extends the resistance network to the final receptor. The detailed interactions between the various resistance for a single leaf are integrated to provide an expression per unit horizontal area of the earth's surface, ie,. the resistance of a single leaf is extended to represent the canopy. R_c combines the stomatal resistance and the cuticular resistance. Cuticular resistance is the resistance to transfer of particulates and gases to the exterior of the leaf and is determined by factors such as wetness and waxiness. Stomatal resistance is the resistance imposed once gases enter the leaf through the dilated stomata of photosynthetically active vegetation.

Within BHATTEX two short intensive monitoring campaigns were held in the austral summer and winter from 8 to 22 March and 9 to 23 June 1996, respectively. Ambient SO_2 and particulate concentrations and micro meteorological data were collected as the basic input information for the inferential model.

At Oak Ridge, Matt and Meyers (1993) found that the dry deposition rate can be underestimated by as much as 40% when weekly averaged SO_2 concentrations are joined with weekly averaged deposition velocities. They suggested continuous or day-night monitoring as methods to reduce the error. During BHATTEX, SO_2 was monitored continuously by means of a Thermoelectron 43A UV-fluorescent analyser for the reason indicated by Matt and Meyers (1993) and because of the need to obtain time resolved data to establish the relationship between deposition and long-range transport. Continuous measurements were used to calculate hourly averages. The analyser was calibrated in the field before each monitoring period.

Particulate samples where collected by an open faced Stacked Filter Unit (SFU). The SFU consisted of two equilibrated Nuclepore filters. An 8.0 μm filter in the first stage collected aerosols above 2.5 μmad (aerodynamic diameter) and a 0.4 μm Nuclepore filter collected the fine fraction (μmad $<$ 2.5 μm) of the aerosol. The SFU sampler operated at 15–16 L min^{-1} and is subject to the same constraints as outlined in the literature for this sampling procedure (Cahill, 1977; Heidman, 1981; John, 1983). Besides monitoring for the coarse and fine particulate fraction, separate filter units were run for the daytime period (0600 to 1800), and for the night-time period (1800 to 0600). Particulate sulphur concentrations were determined using proton-induced X-ray emission (PIXE) analysis.

The micro-meteorological data consisted of 10 m wind speed and direction, the standard deviation of the horizontal wind direction (σ_θ), dry and wet bulb temperature, humidity, rainfall, global solar radiation and atmospheric pressure. Sampling was done every 10 seconds with a Campbell CR-10 data logger which, in turn, calculated and stored hourly averages. The parameter σ_θ is used by the inferential model as a proxy for turbulence and surface roughness. Surface wetness was determined by means of the dewpoint depression where the vegetation surface was assumed to be wet if the dewpoint depression at 1.2 m was less than 1.5 °C, an assumption previously tested successfully on the Highveld (Zunckel *et al*., 1996).

The inferential model allows for a ground cover mix of two species. The various grasses are grouped together and assumed to comprise the total vegetation cover. The inferential model is run for leaf area index (LAI) values of 0.5 and 1.5. The vegetation is assumed to have a full canopy in the summer, while in winter it is dormant. This scenario approach to leaf area index allows for an assessment of dry deposition under South Africa's highly variable rainfall, ie., for dry and wet years, respectively. Little of the required plant physiology data exists for South African vegetation (Smith, pers comm, 1997). As a result data on stomatal resistance and leaf temperature data for grasses at the Sequoia National Park and on the Pawnee Grasslands have been used as surrogates while the canopy height of 0.5 m is typical for Ben MacDhui. Despite the different continents, the grasses at these two sites and at Ben MacDhui have similar a physiology (Smith, pers comm, 1997). The assumed values of leaf area index at Ben MacDhui are conservative. The value of 0.3 for the Pawnee Grasslands is somewhat lower than that assumed at Ben MacDhui in dry conditions while the 2.8 for the Sequoia National Park is higher than the assumed value at Ben MacDhui for wet conditions.

4. Results

The mean SO_2 concentration during the 2-week summer period was 2.6 μg m^{-3}. During the winter period it was 2.3 μg m^{-3}. The low SO_2 concentrations can be attributed to the distance from the source region and loss through oxidation and other processes. Actual concentration values should be treated with caution, as the analyser was operating close to its minimum detection limit and the recorded concentrations fell within the manufacturers specified noise limits of 1 ppb ($\pm$2.7 μg m^{-3}). Nevertheless, the variation is relative and can be considered to be real.

Hourly average SO_2 concentrations during the summer and winter at Ben MacDhui (Figure 3) are low in comparison with those reported for the Mpumalanga Highveld and its peripheral areas (Turner, 1994). They are indicative of a remote site. Turner (1994) showed the long-term annual average concentrations for the central Mpumalanga Highveld to exceed 26 μg m^{-3} and to exceed 10 μg m^{-3} at 100 km distance. The diurnal variation of SO_2 at Ben MacDhui is similar to that reported for the Highveld. Maximum values occur soon after midday and minima

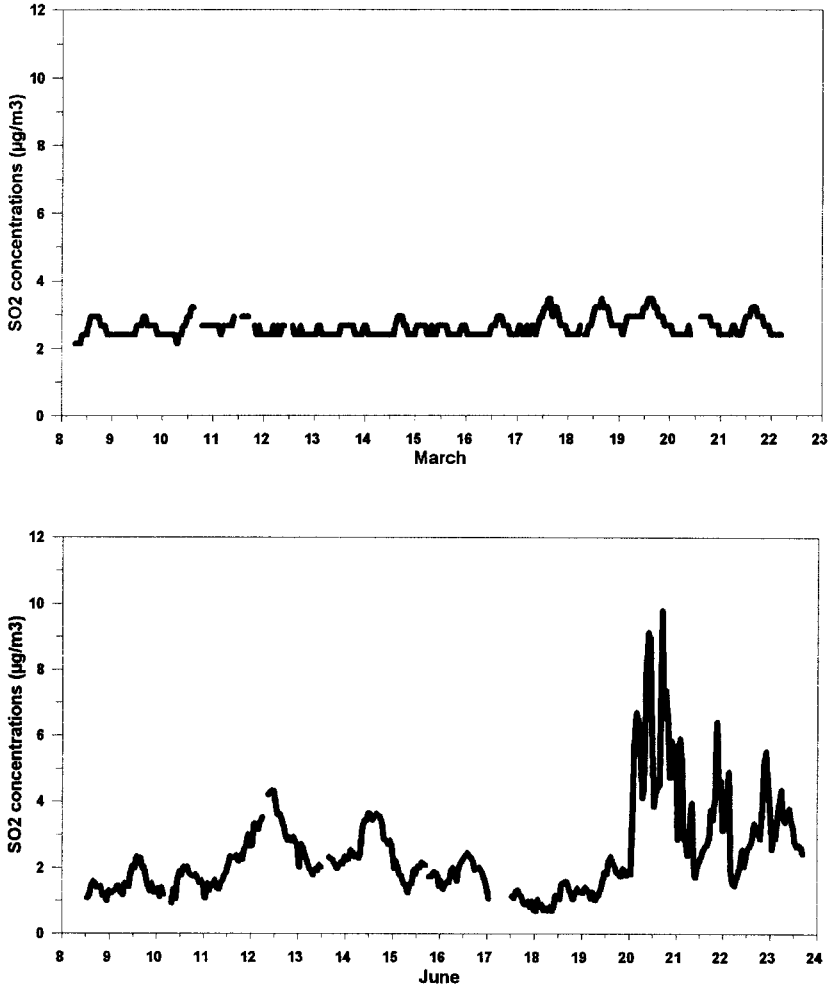

Figure 3. Hourly average SO_2 concentrations during the summer (left) and winter (right) monitoring campaigns.

at night. On the Highveld the diurnal cycle is attributed to the oscillation between daytime convection and strong thermal stratification near ground level at night. By day, the high-level plumes can mix down to ground level, while at night this mixing is inhibited as pollutants are released above surface temperature inversions. At Ben MacDhui mixing down by day of pollutants that are trapped beneath the 700 hPa absolutely stable layer (Cosijn and Tyson, 1996) may account for some of the diurnal variation. At night the surfaced-based inversion that persists over the subcontinent throughout the year (Tyson *et al.*, 1976) prevents any downward mixing of pollutants from aloft. Process such as dry deposition or chemical reaction could also contribute to the observed diurnal variation. The difference in the day-to-day range between summer (March) and winter (June) is meteorologically

TABLE I

Classification of the synoptic types prevalent over southern Africa during the summer and winter field campaigns

Summer															
March	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22
Synoptic-type	C	C	C	W	W	W	W	R	R	C	C	C	C	C	C

Winter																
June	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23
Synoptic-type	W	W	R	C	W	W	W	W	W	R	R	R	R	C	R	R

R: Ridging anticyclone; C: Continental anticyclone; W: Westerly wave.

driven. Summer is relatively undisturbed at a synoptic scale and the day-to-day variation does not vary much. By contrast, marked changes in synoptic flow in winter with strong ridging and frequent westerly intrusions result in a greater observer day-to-day variation in SO_2 concentrations.

In both seasons, most of the particulate sulphur is observed in the fine fraction, ie., $< 2.5\ \mu$m (Figure 4). In summer an average of 85% of the daily sulphate loading is in the fine fraction, ranging between 58 and 100% during the monitoring period. During the winter sampling period an average of 90% of the total sulphate loading occurs in the fine fraction, ranging between 59 and 100%. The major source of the fine sulphur is industrial emissions. The sulphur is emitted into the atmosphere as SO_2 and is oxidised at an average rate of 5% per hour in southern Africa in summer and less in winter (Pienaar and Helas, 1996). Given these oxidation rates and the distance that the material has typically travelled, it is expected that the sulphur would be observed mainly in the fine fraction ($< 2.5\mu$m). The path length around the continental anticyclone from the Highveld could be 1000 km, or more. Any sulphur that has accumulated in the coarse fraction has an atmospheric life of hours and will be deposited by gravitational settling and other processes soon after its formation. Unlike on the central Mpumalanga highveld, there is no indication of a diurnal cycle for particulate sulphur.

The day-to-day variation in SO_2 and particulate concentrations are associated with the large-scale synoptic circulation. A summary of the circulation types observed during the two field campaigns is presented in Table I. The continental anticyclone relates to the plume trajectories indicated in lower panels of Figure 1 and the transport of polluted continental air from the interior over the sampling site. The ridging anticyclone and westerly wave are mostly associated with transporting clean maritime air to the sampling site from the east and southwest, respectively. The peaks in fine particulate concentrations from 8 to 11 March (Figure 4, top-

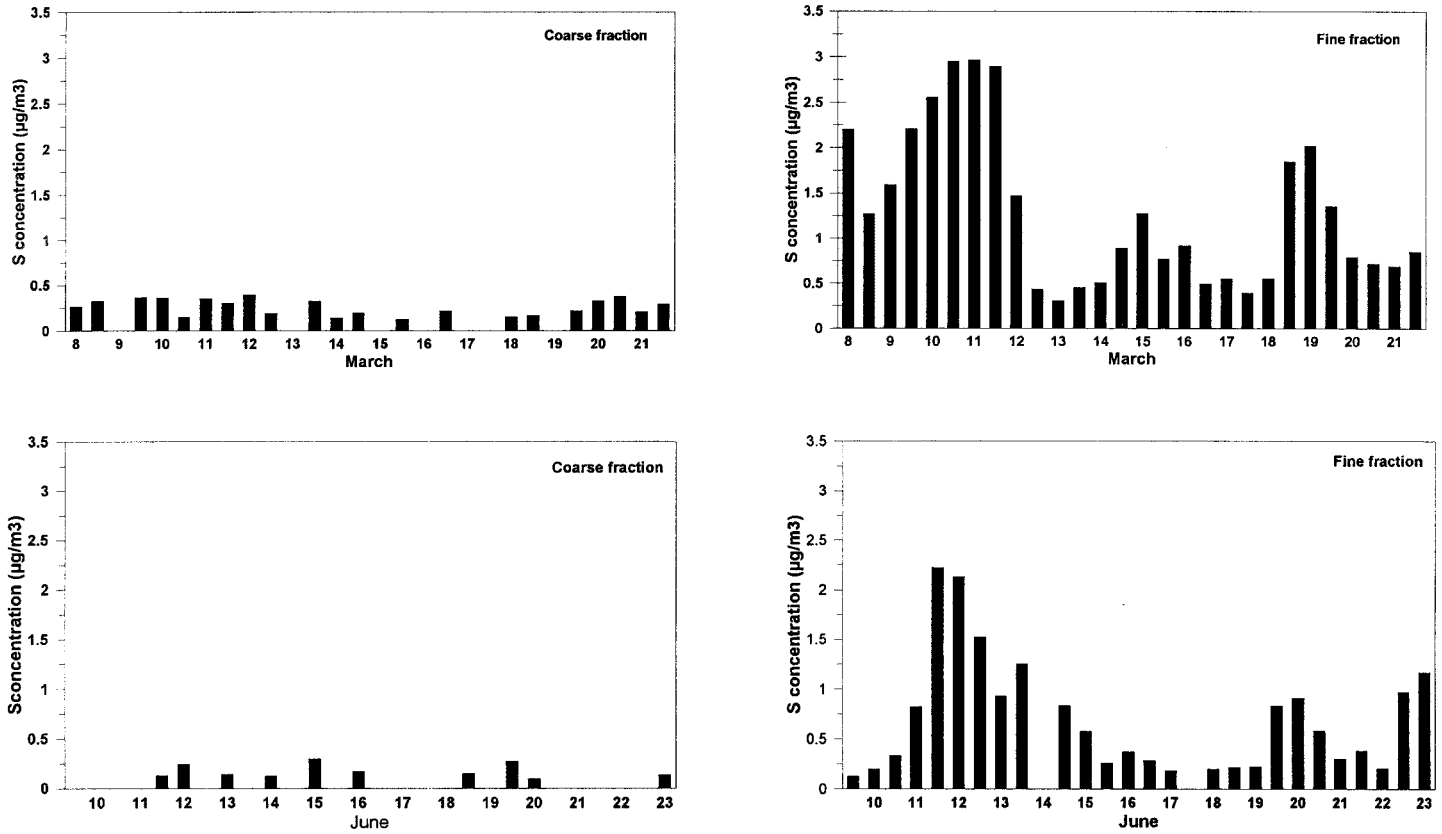

Figure 4. Coarse (left) and fine (right) particulate sulphur concentrations during summer (top) and winter (bottom). Each day is represented by a pair of bars where the bar under the date indicates the daytime period (0600 to 1800) and the undated bar immediately to its right represents the night-time period (1800 to 0600).

right) and around 19 March are indicative of transport in anticyclonic systems from the heart of the subcontinent to Ben MacDhui. By contrast, minimum concentrations are associated with air that has had a short residence time over the subcontinent and a different path to Ben MacDhui. Low concentrations on 12 March and from 16 to 18 March are associated with the passage of a westerly wave and with southerly sub-polar maritime airflow over Ben MacDhui, respectively.

Similarly in winter, low SO_2 and fine particulate concentrations are associated with westerly waves and a south-westerly maritime transport of air over Ben MacDhui. The episodes at the start of the winter campaign and from 15 to 16 June are examples of this situation (Figure 3, right and Figure 4, bottom-right). Low concentrations are also associated with anticyclonic ridging across the southern parts of the subcontinent with a westerly zonal flow such as from 17 to 19 June. The winter peaks from 11 to 12 June and around 20 June are associated with anticyclonic flow and the transport of polluted air from industrial sources from central interior.

The height of the 700 hPa absolutely stable layer in relation to the height of the sampling station also plays a role in the observed day-to-day variation as pollutants are trapped immediately below this layer. The pressure difference between the base of the absolutely stable layer and the surface pressure at Ben MacDhui is used to indicate their relative heights to each other (Figure 5). A negative pressure difference indicates that the absolutely stable layer is below the sampling site. On such occasions the pollutant concentrations should be low with any synoptic type as the sampling site lies in the cleaner air aloft. This was indeed so from 16 to 19 June (Figures 3, 4 and 5). Low concentrations should also occur in a deep well-mixed lower atmosphere when the absolutely stable layer is well above the sampling site such as 11 to 14 June. Data used to determine the height of the absolutely stable layer were recorded over Bloemfontein, the nearest radiosonde station to Ben MacDhui on the interior plateau.

Relatively high fine particulate sulphur concentrations occur under established continental anticyclonic conditions when the height of the 700 hPa absolutely stable layer coincides with Ben MacDhui, as on 10 and 19 March 1997. The peak in fine particulates and SO_2 concentrations around 20 June occurs under such conditions. Conversely, low pollutant concentrations occur when a small relative height difference exists during zonal flow induced by a westerly wave flow or a ridging anticyclone. Examples with a westerly wave are 12 March and 15 June and with a ridging anticyclone are on 17 March and 18 June.

The inferred deposition velocity exhibits a strong diurnal variation in summer and winter with daytime maxima and night-time minima (Figure 6). This is indicative of the dependence of V_d on photosynthesis, global radiation and in turn, on the turbulence proxy, σ_θ. Photosynthetically active vegetation in summer influences the deposition velocity for SO_2 (Figure 6, top and centre) when mean values range from 0.32 to 0.47 cm s^{-1} for a leaf area index of 0.5 and 1.5, respectively. Changes in leaf area index in summer have no influence on the deposition velocity for par-

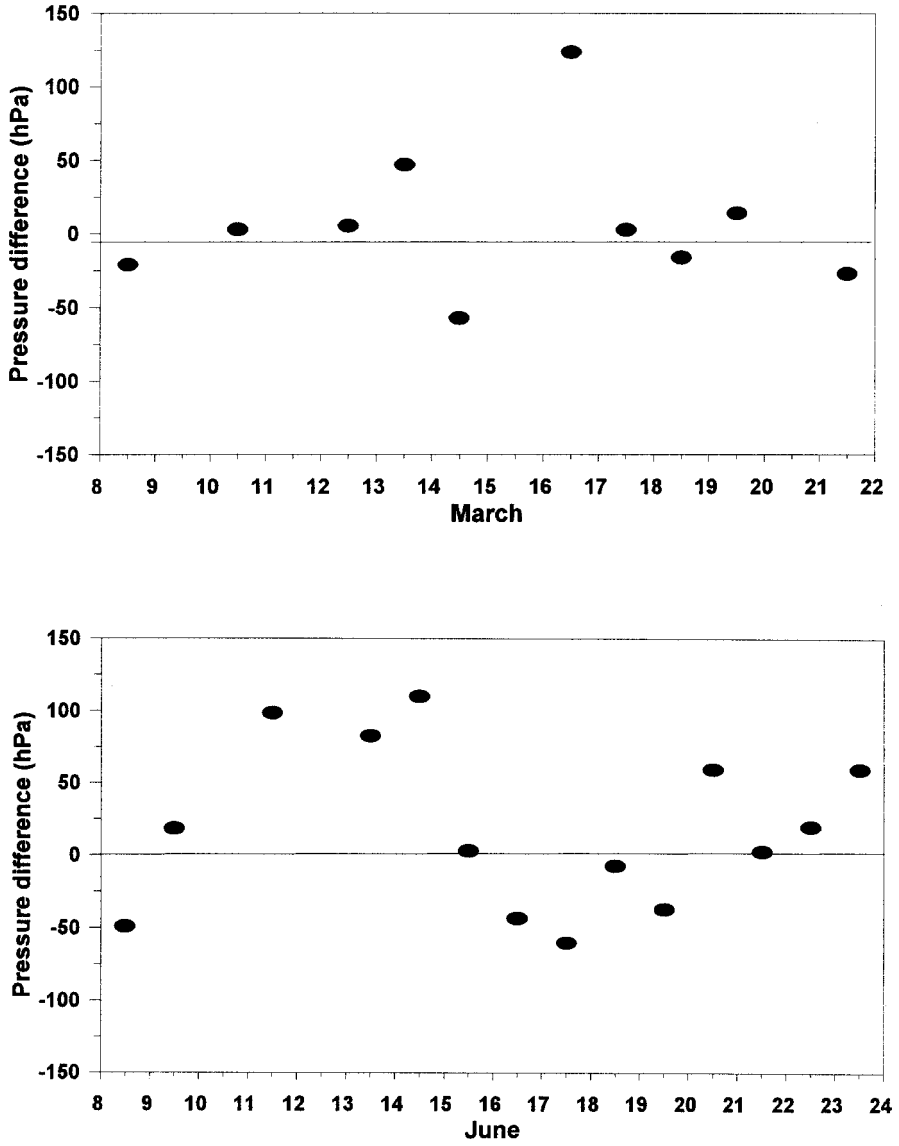

Figure 5. Relative pressure difference between the base of the 700 hPa absolutely stable layer and the sampling site on Ben MacDhui for summer (left) and winter (right). A positive difference indicates that the absolutely stable layer is above the sampling site.

ticulates when meteorology appears to dominate the process (Table II). In dormant winter vegetation the canopy resistance has a negligible influence on deposition velocity. As a result changes in leaf area index do not influence deposition velocity for either SO_2 or particulates in winter. Deposition velocity in winter is rather con-

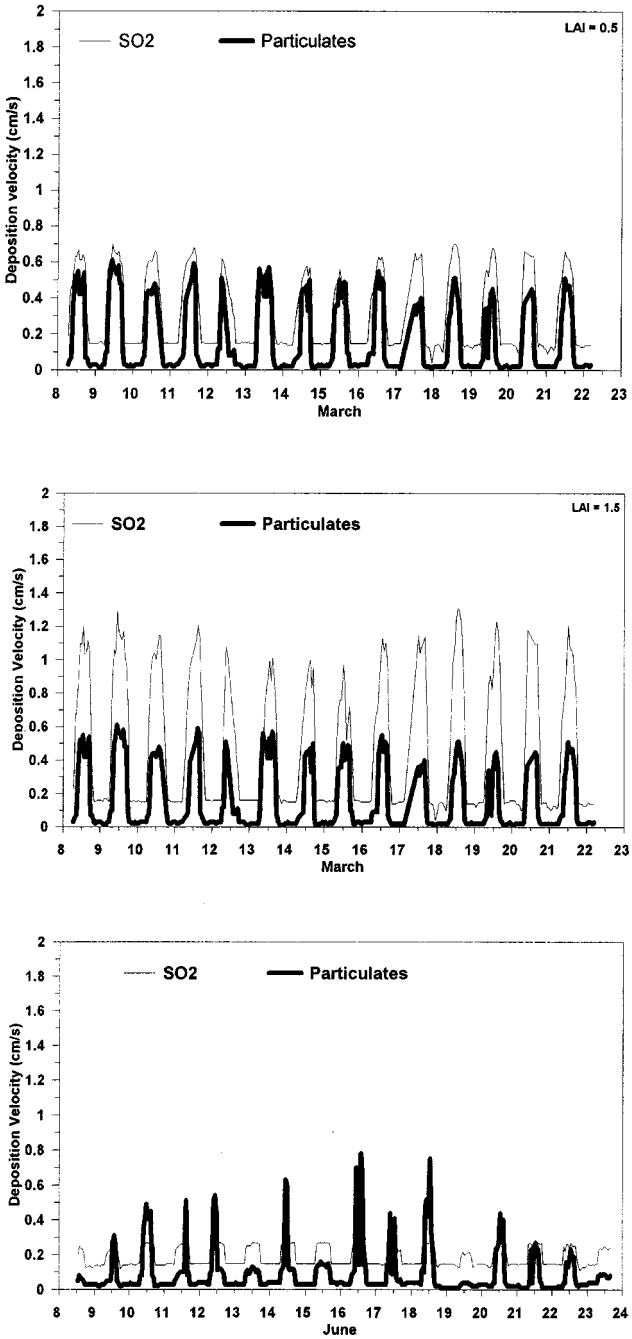

Figure 6. Mean hourly deposition velocities for SO_2 and particulates at Ben MacDhui for two leaf area index scenarios in summer (top two diagrams) and in winter (bottom).

TABLE II

Deposition velocity (cm s^{-1}). The range is between the minimum and maximum values

	LAI	SO_2		Particulates	
		Mean	Range	Mean	Range
Summer	0.5	0.32	0.66	0.15	0.60
	1.5	0.47	1.27	0.15	0.60
Winter	0.5	0.19	0.17	0.09	0.77
	1.5	0.19	0.17	0.09	0.77

trolled by the aerodynamic and quasi-laminar boundary layer resistances. Canopy resistance in winter plays a small role with deposition to the leaf surface only.

The meteorological conditions during the two monitoring periods are considered to be typical of summer and winter, respectively. Mild days and cool nights were experienced during the summer campaign with convective afternoon or evening showers on six occasions. No rain fell during the typically dry winter campaign when cool days and cold nights were experienced. Mean global radiation for the 2-week summer and winter monitoring campaigns was 253.4 and 148.7 W m^{-2}, respectively. The mean summer temperature was 10.9 °C in comparison to 3.1 °C in winter and mean summer relative humidity was 60.7% by comparison to 39.9% in winter. In contrast, the summer σ_θ value of 12.2 is only marginally higher than the 11.1 in winter. The fact that deposition velocities for SO_2 and particulates are larger in summer than in winter may be attributed to the higher values of radiation, temperature, turbulence and humidity in summer and to the photosynthetically active summer vegetation.

In summer, leaf area index influences the deposition flux and larger values of leaf area index are associated with higher deposition (Figure 7), ie., there is more photosynthetically active vegetation available. The mean hourly deposition flux ranges from 29.8 to 44.1 μg m^{-2} for leaf area index of 0.5 and 1.5, respectively. The total deposition of sulphur from SO_2 for the summer ranged from 4.64 to 6.86 mg m^{-2} for leaf area index of 0.5 and 1.5, respectively.

In winter, since changes in leaf area index in the dormant vegetation do not influence the deposition velocity for SO_2, they in turn, do not affect the deposition flux. The hourly mean deposition flux for the winter is 16.6 μg m^{-2} with a total deposition of sulphur from SO_2 of 2.94 mg m^{-2}. The strong relationship that exists between the pollutant concentration and deposition flux (Equation (1)) is emphasised in the SO_2 episode around 20 June (Figure 3 and Figure 7, bottom). Of the

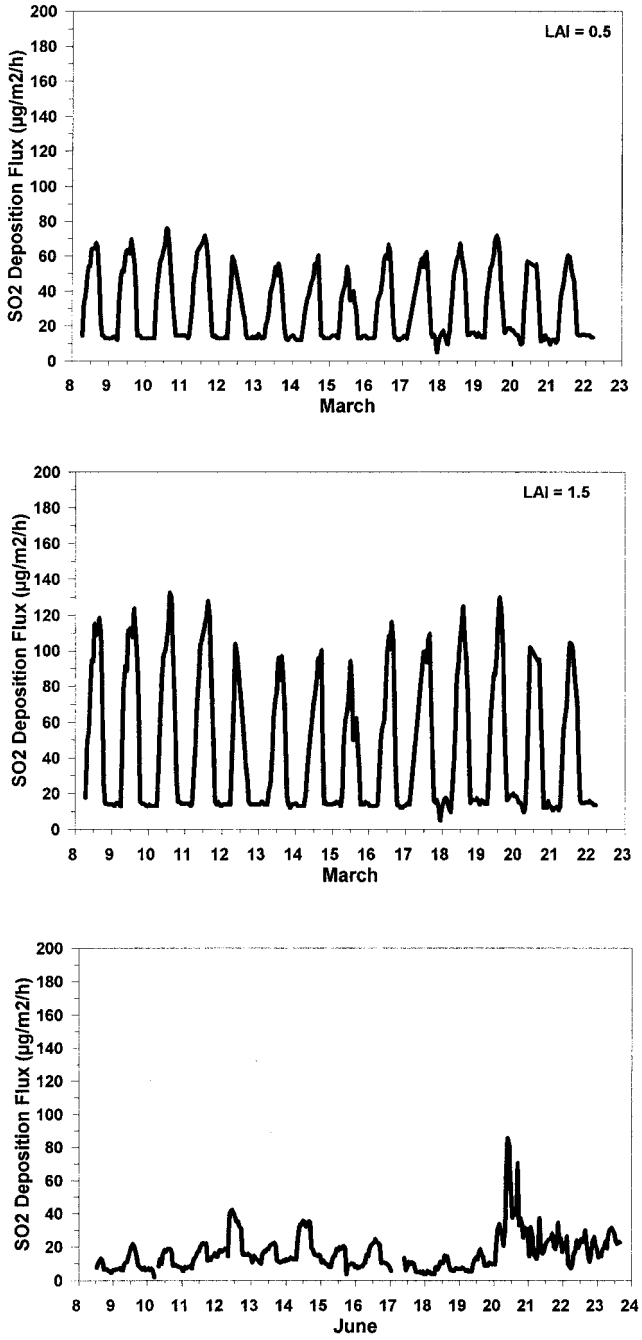

Figure 7. Mean hourly SO_2 deposition flux during summer for the two leaf area index scenarios (top and centre), winter (bottom).

TABLE III

Contribution to dry sulphur deposition by sulphur in SO_2 and particulate sulphur for summer and winter halves of the year. LAI is in parenthesis

	Summer (mg S m^{-2})		Winter (mg S m^{-2})
Gaseous	4.64 (0.5)	6.86 (1.5)	2.94
Particulate	0.21		0.06

total dry deposition of sulphur from SO_2 during the winter period, 39% occurred during this episode.

The mean hourly particulate sulphur deposition rate is 0.7 and 0.2 μg m^{-2} for the summer and winter, respectively (Figure 8). The deposition flux is low in both seasons and this may be attributed to the relatively low deposition velocities for particulates and to the relatively low particulate sulphur concentrations. The total particulate sulphur deposition is 0.21 mg m^{-2} in summer and 0.06 mg m^{-2}during winter. In the model leaf area index has no effect on the deposition velocity for particulates. In turn, it has no effect on the deposition flux for particulate sulphur.

The total deposition of sulphur during the summer is dominated by gaseous deposition and ranges from 4.9 to 7.1 mg m^{-2} for leaf area index of 0.5 and 1.5, respectively. During the winter the total sulphur deposition of 3.0 mg m^{-2} is also attributed mostly to gaseous deposition (Table III). With the majority of dry sulphur deposition associated with SO_2, the low deposition flux should not be confused with the high particulate loading reported at Ben MacDhui by Piketh *et al*. (1997).

An estimate of the annual deposition rate for sulphur can be made if the two-week summer and winter monitoring periods are each assumed to be representative of the summer and winter halves of the year. On this basis, the annual deposition rates for total sulphur range from 1.0 to 1.3 kg ha^{-1} for leaf area index of 0.5 and 1.5, respectively. These deposition rates compare favourably with those at remote sites in the U.S. Environmental Protection Agency (EPA) Clean Air Status and Trends Network (CASTNet) where the inferential technique is used (Clarke *et al*., 1997). On the Pawnee Grasslands which are well removed from the major source region in the U.S., Meyers *et al*. (1991) estimated annual deposition rates of total sulphur of 0.6 kg ha^{-1}.

A difference of 27% in dry sulphur deposition occurs between dry years (leaf area index = 0.5) and wet years (leaf area index = 1.5). This difference is relative given the low deposition rate for sulphur of less than 300 g ha^{-1} a^{-1}. Hence, South Africa's variable annual rainfall and its influence on vegetation density does

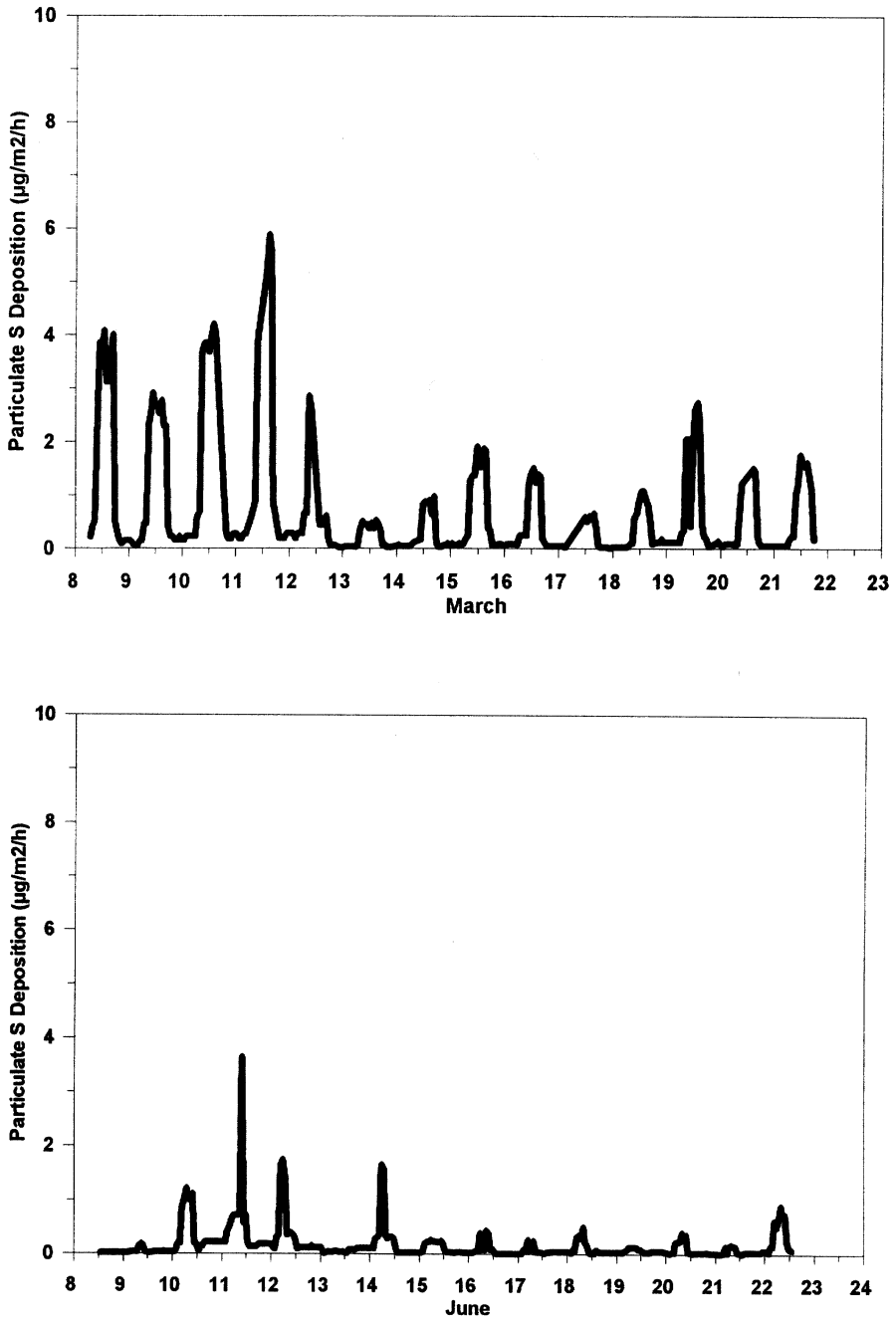

Figure 8. Summer and winter deposition flux for particulate sulphur.

not strongly influence dry sulphur deposition at the remote rural site. Based on a similar methodology on the central Mpumalanga Highveld, Zunckel *et al*. (1996) calculated dry deposition rates for sulphur of 8.2 kg ha^{-1} a^{-1}. Dry deposition of sulphur at the high-altitude background station is approximately 13% of that in the source area.

5. Conclusions

Dry deposition of sulphur at a high-altitude background station in South Africa has been established and related to day-to-day variability in the transport of trace gases and aerosols over the area. In summer, the total deposition of sulphur from SO_2 ranges from 4.64 to 6.86 mg m^{-2} for a leaf area index of the vegetation of 0.5 and 1.5, respectively. In winter, when leaf area index does not influence the deposition rate of SO_2 and the total sulphur deposition from SO_2 is 2.94 mg m^{-2}. The total particulate sulphur deposition is 0.21 mg m^{-2} in summer and 0.06 mg m^{-2} in winter. Annual deposition rates for total sulphur range from 1.0 to 1.3 kg ha^{-1} for leaf area index of 0.5 and 1.5 and are a consequence of large-scale regional transport from the Mpumalanga Highveld source region. The dry deposition of sulphur at Ben MacDhui is approximately 13% of that measured in the source region of the central Mpumalanga Highveld.

Higher summer than winter deposition velocities for SO_2 and particulates account for deposition rates for sulphur in summer being more than double those in winter. In summer, the meteorology and the photosynthetically active vegetation influences the rate of dry deposition of sulphur from SO_2. A relatively small difference occurs in dry sulphur deposition between dry and wet years, hence, South Africa's variable rainfall and concomitant vegetation density does not strongly affect dry deposition of sulphur at the remote rural site.

The relative height of the 700 hPa absolutely stable layer at Ben MacDhui together with varying transport pathways associated with changing synoptic-scale circulation influences the day-to-day variability in SO_2 and particulates concentrations, as well as sulphur deposition rates. Episodes of high SO_2 and fine particulate concentrations and high dry deposition of sulphur occur with anticyclonic circulation over the central interior of South Africa, and when the height of the absolutely stable layer coincides with the sampling site. This results in the long-range transport from the Highveld source region. Episodes of low concentrations and sulphur deposition occur with anticyclonic circulation over the subcontinent, but the 700 hPa absolutely stable layers situated either below or well above Ben MacDhui. Low SO_2 and fine particulate concentrations are associated with westerly waves and ridging anticyclones regardless of the height of the stable layer.

The major transport corridor from South Africa to the Indian Ocean and beyond passes over the remote, rural, high-altitude site at Ben MacDhui. The site is perfectly situated to monitor background levels of sulphur products from the

industrialised Highveld as they are transported from the source region. This transport occurs in well defined pathways undulating within the African haze layer that extend on a majority of days in a year to about 500 hPa. The horizontal transport is largely determined by the atmospheric circulation types prevailing, whereas vertical transport is a function of the occurrence of persistent stable layers in the troposphere.

Acknowledgements

The author acknowledges the assistance of Dr Bruce Hicks, Director of the Air Resources Laboratory at Silver Spring, Maryland and Dr Ray Hosker, Director of ATDD at Oak Ridge, Tennessee for supplying the inferential model. Thanks also to Dr Tilden Meyers and Dr Detlef Matt of ATDD at Oak Ridge. CSIR are acknowledged for financial support. The staff and management Tiffendale Ski Resort, the BHATTEX project team and CSIR's Dawid Swanepoel and Janusz Lisowski are thanked for logistical assistance. Prof Peter Tyson is thanked for his comments and criticisms on the initial draft.

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