The Ben Macdhui High Altitude Trace Gas and Aerosol Transport Experiment

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The Ben Macdhui High Altitude Aerosol and Trace Gas Transport Experiment (BHATTEX) was started to characterize the nature and magnitude of atmospheric, aerosol and trace gas transport paths recirculating over and exiting from southern Africa to the Indian Ocean. This field campaign, a multi-institutional, interdisciplinary atmospheric study conducted in the southern Drakensberg during 8–22 March and 9–23 June 1996, aimed to characterize the nature and amount of the aerosols transported, to verify the accuracy of trajectory models used to predict the observed aerosol transport and finally to determine the extent to which anthropogenic sulphur can be found to affect a remote site in southern Africa. An overview of the BHATTEX project, as well as a report on some results obtained during the initial long-term time-resolved particulate sampling and the two intensive sampling campaigns, are presented.

Determination of the transport and deposition of aerosols and trace gases in southern Africa has been the focus of several international research programmes: Southern African Fire-Atmosphere Research Initiative-92 (SAFARI-92),[1,2] Transport and Atmospheric Chemistry near the Equator-Atlantic (TRACE-A),[3] and Southern African Atmospheric Research Initiative-94 (SAARI-94).[4] The influence of aerosols and trace gases on the air quality on the South African highveld has been the subject of a recent review.[5]

The nature and extent of aerosol and trace gas transport out of the industrialized highveld region over southern African are not well documented. Atmospheric recirculation of aerosol and trace gases over the interior plateau had previously been suggested from investigations of southern African atmospheric particulate and trace gas chemistry.[5–9] Difficulties arose, however, in attempting to confirm these findings without appropriate large-scale meteorological data. Atmospheric recirculation occurring on a subcontinental scale over southern Africa was demonstrated for the first time during SAFARI-92 and provided new insight into the manner of air-parcel transport over the subcontinent.[10,11]

The Ben Macdhui High Altitude Aerosol and Trace Gas Transport Experiment (BHATTEX) was started in response to the need to explore and confirm the nature and extent of atmospheric recirculation over the South African highveld. Foremost among the objectives of BHATTEX was the characterization of the nature and magnitude of atmospheric, aerosol and trace gas transports recirculating over and exiting from southern Africa to the Indian Ocean. Specifically, this multi-institutional, interdisciplinary study aimed to characterize the nature and amount of the aerosols transported, to verify the accuracy of trajectory models used to predict the observed aerosol transport, and finally to determine the extent to which anthropogenic sulphur affects remote areas of southern Africa. The purpose of this paper is to provide the scientific community with an overview of the BHATTEX project, as well as to report on some of the preliminary results of the the long-term time-resolved particulate sampling and two intensive sampling campaigns (8–22 March and 9–23 June 1996).

Background

Four major synoptic circulation types are responsible for most of the atmospheric transport of southern African aerosols: semi-permanent subtropical anticyclones, transient mid-latitude ridging anticyclones, westerly baroclinic disturbances, and barotropic quasi-stationary tropical easterly disturbances.[10,11] Anticyclonic-type recirculation over southern Africa is the dominant transport mode (approximately 44 % on average) of atmospheric constituents[10] (Fig. 1). This type of atmospheric circulation is able to transport material at varying spatial scales that range from hundreds to thousands of kilometres for 10 or more days at a time.[11] It is persistent throughout the atmosphere to about the 500 hPa absolutely stable layer (Fig. 2).[10,11]

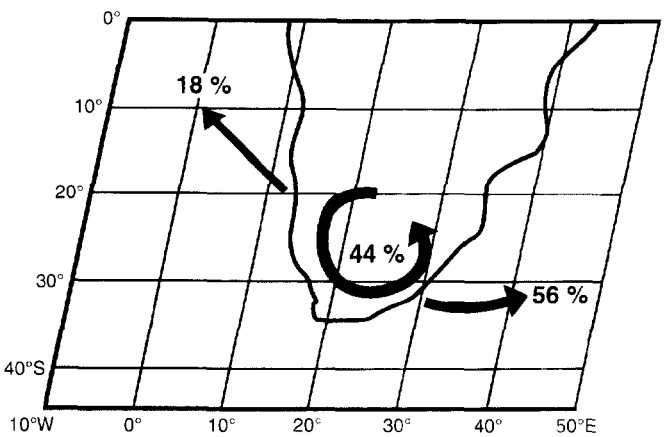

Fig. 1. Average transport as predicted by kinematic trajectory analysis for all synoptic circulation types between the surface and 500 hPa (after Tyson *et al.*[18]).

It can be expected that atmospheric transport and recirculation over temporal scales of 10 days and spatial scales of up to 4000 km have implications for the radiative balance, photochemistry, and the biogeochemistry of southern Africa. With the increased interest of the scientific community in the radiative effects of aerosols, especially fine sulphate particles,[1] these transport patterns become a subject of growing concern. Over the short term, the transport of aerosols and trace gases may affect radiative balances and lead to regional-scale forcing of climatic variables by sulphate aerosols.[13]

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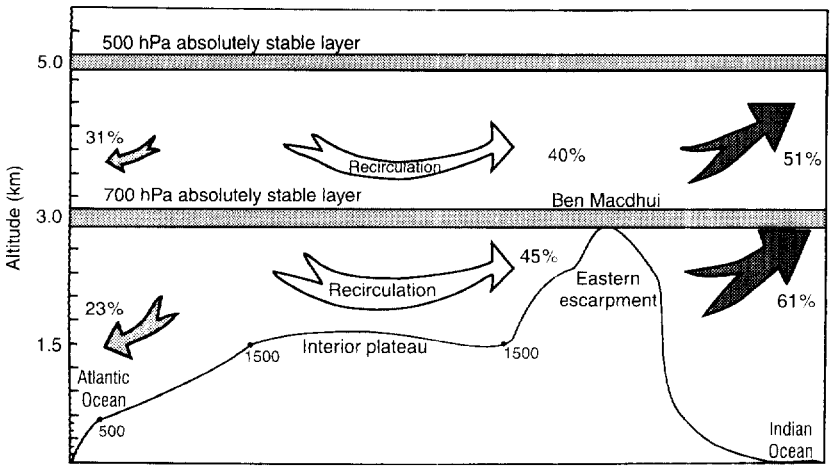

Fig. 2. Variation with height of average transport from the southern African interior to the two adjacent oceans (after Tyson *et al.*[18] and Garstang *et al.*[12]).

The long-range transport of aeolian material is an important component of regional biogeochemical and nutrient dynamics of far-removed ecosystems in other parts of the world.[14] Evidence suggests the same may be true of southern African ecosystems.[15,16] The total amount of aerosols transported over the southern African subcontinent is in the order of 135 Tg per year.[11] Of this amount, approximately 60 Tg of aerosols is transported, redistributed and deposited over southern Africa, approximately 30 and 45 Tg are transported westward to the Atlantic Ocean and eastward to the Indian Ocean, respectively.[11] Much of this aeolian material contains biogeochemically important species.[9,16,17] The transport may have lasting effects on the various biogeophysical cycles of ecosystems downwind. The above results, based on theoretical models and findings of the SAFARI-92, TRACE-A and SAFARI-94 experiments, provide a useful framework to test and verify hypotheses concerning atmospheric transports of pollutants such as ozone, ozone precursors, sulphates and biogeochemically important species of nitrogen and carbon over southern Africa.

Rationale

A major export corridor of air parcels, off the southeastern margin of the subcontinent, occurs at about 30° S near the height of a semi-permanent, absolutely stable layer (700 hPa).[10,19] Initially, sampling was conducted for one year using a time-resolved, particulate streaker sampler to test for signals of the long-range transport of industrial emissions from Gauteng and Mpumalanga in this exit corridor. The site, located on Ben Macdhui in the Eastern Cape Province (Fig. 3), was commissioned on 6 July 1995 and comprised a streaker sampler capable of month-long operation and a wind direction and wind-speed sensor.

Preliminary data from the first two months of streaker sampling at Ben Macdhui suggested that episodes of elevated sulphur concentrations, most probably of industrial origin, could be detected periodically (Fig. 4). However, it was not possible to identify these episodes as having exclusively industrial sources. The implementation of a series of intensive field programmes to address the following scientific questions became necessary:

- Is large-scale atmospheric recirculation occurring over southern Africa and could chemical and meteorological evidence of this atmospheric recirculation over the subcontinent be found at Ben Macdhui?
- Could sources, whether the result of industry, biomass burning or biogenic, be isolated from observational data gathered at the site?

Site description

An essential component of this project was suitable siting of the sampling efforts. The site had to be located in a region previously determined to lie in the middle of a recirculating limb of anticyclonic transport. The site also had to be as far removed as possible from potential industrial sources to minimize the possibility of local contamination. Trajectory analyses undertaken for the SAFARI-92 study indicated that the southern Drakensberg was ideally situated to observe atmospheric recirculation, with a location along the axis of maximum aerosol transport between 30 and 35° S.[16,17,19] The sampling site chosen was located on Ben Macdhui (30° 32′S, 27° 58′E, ~2780 m asl) near the border of Lesotho and the Eastern Cape Province (Fig. 3).

At an altitude of 2780 m above sea level, the sampling site intercepts the southern African planetary boundary layer, which is vertically capped by the presence of multiple layers of absolute stability at 700 and 500 hPa.[19,20] Ben Macdhui is situated over 600 km (straight line distance) from South Africa's industrial centres of Gauteng and Mpumalanga; this avoided any direct contamination of the samples by their industries. Potential local sources of aerosol and trace gases included the Tiffindell Ski Resort, located approximately 1.5 km downwind of, and some 40 m lower in altitude than, the intensive-sampling site. The nearest town, Rhodes, with a population of less than 500, is located some 1300 m lower and 25 km south-southeast of Ben Macdhui.

Methodology

The BHATTEX project was initiated by the Climatology Research Group at the University of the Witwatersrand. It soon became evident, however, that a multi-institutional, international and interdisciplinary approach would be necessary to optimize the results. Meteorologists, aerosol scientists, inorganic and organic geochemists, biogeochemists and systems

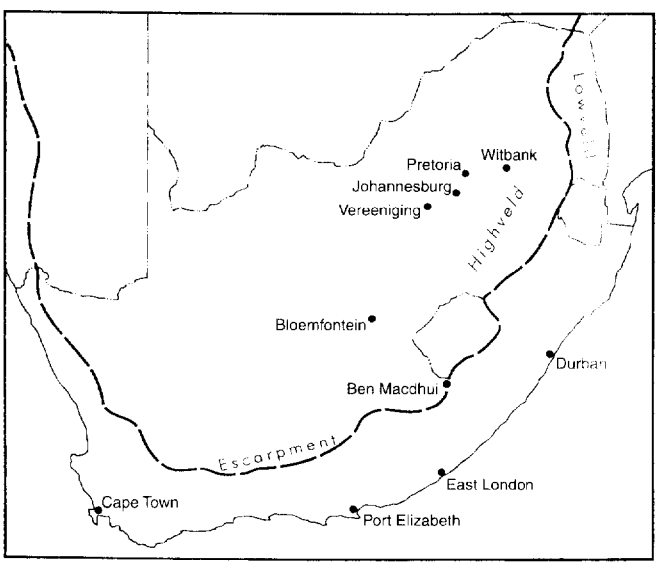

Fig. 3. Location of the sampling site for both the long-term sampling (June 1995 to January 1997) and the two intensive sampling campaigns (March and June 1996).

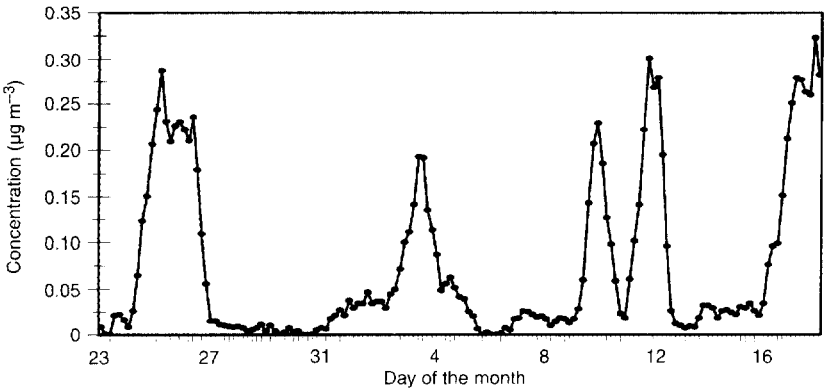

Fig. 4. Time series plot of industrially derived aerosols detected at Ben Macdhui during August 1995.

ecologists from 17 research groups are currently involved in this project (Table 1). Meteorological and chemical analyses are combined to provide an integrated approach to solving the problem of detailing aerosol characteristics, sources, transports and fluxes at Ben Macdhui. Semi-diurnal aerosol sampling is combined with daily kinematic trajectory modelling.

An investigation of the scientific questions necessitated intensive sampling campaigns of suitable duration to allow for sampling during each of the four major synoptic circulation types over southern Africa. The two short, intensive field campaigns that comprised BHATTEX maximized the climatological likelihood of achieving the sampling goals. The summer intensive field campaign was conducted during the austral summer, the season thought to have the lowest frequency of atmospheric recirculation (8–22 March 1996), whereas the intensive field campaign in winter was conducted during the high recirculation season (9–23 June 1996). Each intensive field effort lasted 14 days. Aerosol and trace gas samples were collected every 12 hours. A particulate streaker sampler, with a sample time resolution of four hours, was also operated at the site for the two intensive field campaigns. The chemical sampling scheme covered temporal periods that ranged across interacting scales of motion from micro-scale (<1 km) to synoptic scale (<10^4 km).

Because multiple sources can emit similar elemental signatures, source attribution of the aerosol samples on the basis of one or two elements is tenuous at best. BHATTEX used a wide range of elemental, inorganic, organic and isotopic analyses to identify different particulate and trace gas sources and to provide as broad and robust a chemical approach as feasible. During the summer field campaign, nine samplers were used (six particulate, three trace gas), while during June 10 samplers were used (Table 2). The combined data from the samplers will provide approximately 50 components for each 12-hour period.

Aerosol and trace gas samplers and analyses

Streaker sampler. The streaker sampler was initially commissioned at Ben Macdhui during July 1995. It was operated with 12 V d.c. power to enable the use of solar energy to maintain continuous sampling over a period of 28–30 days. The sampling head consisted of a PIXE International Inc. streaker unit, capable of collecting size-fractionated and time-resolved aerosol samples in the coarse [10 < aerodynamic diameter in micron (μmad) > 2.5] and the fine fraction (μmad <2.5). The coarse fraction was collected on a Vaseline-coated impaction stage of Kapton or Mylar foil. The fine material was trapped on a 0.4 μm Nuclepore filter. Both were loaded into a sampling cassette that revolved over a sucking orifice. This ensured that both size fractions remained aligned in time. The sampling and analysis (Table 2) gave the inorganic elemental composition of each millimetre of the exposed sample, which corresponds to four hours of sampling.

Stacked and single-stage filter units. Stacked and single-stage filter units (SFU and SSFU) were used in the two intensive sampling experiments. The SFU cassettes were loaded with two Nuclepore filters (8.0 μm and 0.4 μm). The top filter captured the coarse material and the second filter the fine material. The samples were all pre- and post-weighed to determine the gravimetric mass that was deposited after 12 hours of exposure. Details of sampling and analysis are given in Table 2.

The SSFU cassettes were loaded with 2 μm and 1 μm Teflon filters during the summer and winter experiments, respectively. These samples were exposed for 24 hours at flow rates of about 28 l min^{-1}. Material collected on these filters was not size fractionated and is representative of the total suspended particulate matter. The object was to obtain as much material on the filters as possible for instrumental neutron activation analysis (INAA).

Total suspended particulate samplers. Surface particulates were collected on several different filter media over semi-diurnal time scales. Teflon, Nuclepore, cellulose and pre-combusted quartz (1 h at 700 °C) filter media were used for the various analyses (Table 2). Sampling flow rates varied from 20 l min^{-1} to approximately 1400 l min^{-1}. Non-size-discriminate aerosol samples were collected with a high-volume sampler (approximate flow rate

Table 1. List of participating laboratories for the two intensive sampling campaigns.

Participating group	Analytical contribution
University of the Witwatersrand, Climatology Research Group	Kinematic trajectory analysis
University of the Witwatersrand, Schonland Research Centre	PIXE analysis
CSIR	Micro-meteorology, SO_2 and O_3
Eskom	Pilot balloon soundings
University of Port Elizabeth	Alkyl nitrate/trace gas sampling
University of Potchefstroom, Department of Chemistry	Passive trace gas sampling
University of the Witwatersrand, Hugh Allsop Laboratory	Radiogenic isotopic analysis
University of Virginia, Department of Environmental Sciences	Bulk and compound specific stable isotopic analysis
University of New Hampshire, Institute for the Study of Earth, Ocean and Space	Ion chromatography
Lawrence Livermore National Laboratory, California	Atomic absorption spectrometry
Desert Research Institute, Nevada	X-Ray fluoresence analysis
University of Ghent	Neutron activation analysis

Table 2. Analytical and sampling information for the intensive sampling campaigns.

Chemical analysis	Filter substrate	Sampling apparatus	Flow rates	Sampling duration
PIXE long-term	Coarse fraction: Mylar Fine fraction: Nuclepore	PIXE International Inc. streaker unit	~1 l min^{-1}	1 month – 4 hour time resolution
PIXE intensive	Nuclepore (8.0 & 0.4 μm pore size)	Stacked filter unit	~20 l min^{-1}	11.5 hours
IC	90 mm Zeflour Teflon filter	Dual Teflon sampling head	~60 sl min^{-1}	11.5 hours
IRMS	Pallflex Quartz filter	Sierra high volume sampler	1400 l min^{-1}	11.5 hours
TIMS	Pallflex Quartz filter	Sierra high volume sampler	1400 l min^{-1}	23.5 hours
INAA	Teflon filters	Single stage filter unit	28 l min^{-1}	23.5 hours
AMS	Pallflex Quartz filter	Sierra high volume sampler	1400 l min^{-1}	11.5 hours

of 1.4 m^3 min^{-1} over 11.5 h). Ninety-millimetre Teflon and cellulose filters were also used to collect samples for both ion chromatography and X-ray diffraction (XRD). Flow rates of these samplers were in the order of 60 sl min^{-1} (standard litres per minute).

Water-soluble, ionic species compositions and concentrations were determined using the ion chromatography analysis techniques for aerosols developed by Talbot *et al.*[21,22]

The molar concentrations for the following particulate species were determined for both the summer and winter components of BHATTEX: Cl^-, PO_4^{3-}, NO_3^-, SO_4^{2-}, C_2O_4, Na^+, NH_4^+, K^+, Mg^{2+} and Ca^{2+}. Concentrations are expressed as atmospheric mixing ratios in parts per trillion by volume (pptv).

Bulk stable isotopic analyses of atmospheric constituents were used to help indicate parent materials of carbon and nitrogen fractions of the particulates.[23] These techniques are used in a somewhat novel fashion in that they are combined to differentiate surface particulate samples from different sources.

Bulk and compound-specific stable isotope ratio mass spectrometry of carbon and nitrogen isotopes was performed on both the organic and total particulate nitrogen fractions of total suspended particulates. These analyses use the standard nitrogen and carbon techniques as well as newly developed compound-specific approaches for the analyses of atmospheric particulates.[24] Surface particulates were collected on pre-combusted (1 h at 700–725 °C), Pallflex ultra-pure quartz fibre 8 × 10" filters using a Sierra high volume (~1.4 m^3 min^{-1} inflow rate) total suspended particulate (TSP) sampler for periods of approximately 11.5 hours. The samples were sent to the University of Virginia and analysed for bulk stable carbon and nitrogen and compound-specific stable carbon isotopic signatures.[16,24] Stable isotopic ratios are reported as the ratio of $\delta^{13}C$ to $\delta^{12}C$ of the sample relative to the PDB standard and as $\delta^{15}N$ to $\delta^{14}N$ relative to the atmospheric standard. Concentrations were determined from empirically derived relationships between known concentrations and manometer deflections and ion-beam gauge readings.

Radiogenic isotopic analysis

In addition to recording stable isotopes of carbon and nitrogen, samples were also taken for the determination of radiogenic isotopic ratios of C, Sr, Nd and Pb by atomic mass spectrometry (AMS) for $\delta^{14}C$ and by thermal isotope mass spectrometry (TIMS) for Sr, Nd, Pb and Ur. Samples for $\delta^{14}C$ analysis were taken from filters used for the stable isotopic analyses of particulate carbon and nitrogen fractions. Samples for TIMS analysis were collected during the winter campaign only, using a Sierra high-volume sampler and Pallflex ultra-pure quartz fibre filters but for a sampling duration of 24 hours owing to uncertainties concerning particulate loading of the desired species. These samples were analysed using TIMS at the Hugh Allsop Laboratory, University of the Witwatersrand.

Trace gas samplers

Sulphur dioxide and ozone. Sulphur dioxide (SO_2) concentrations, indicative of fossil-fuel burning, and ozone (O_3), an indicator of multiple combustion sources, particularly biomass burning, were determined continuously during the two intensive campaigns by real time electronic O_3 analysers and UV-absorption SO_2 analysers[4,26]. Additionally, SO_2 and sulphate were monitored to determine the rate of sulphur deposition. SO_2 is the main precursor of sulphate aerosols.

Alkyl nitrates and halocarbons. Recent evidence has suggested that alkyl nitrates and halocarbons may be useful tracers of natural and anthropogenic emissions to the troposphere.[27,31,33,34] It has been proposed that alkyl nitrates be used as tracers of anthropogenically affected air masses transported to the remote troposphere.[28,33,34] To this end, alkyl nitrates and halocarbons were collected and analysed based on the method described by Atlas and Schauffler.[35]

Meteorology

The meteorological analyses used rely on methods that range across interacting scales of motion. Daily synoptic charts at the surface and 700 hPa levels, daily thermodynamic profiles of the regional atmosphere from radiosondes, multiple, daily pilot balloon launches (winter only), micrometeorological information at the sampling site and three-dimensional trajectory analyses, were all used to characterize the atmospheric environment during BHATTEX. Daily synoptic charts supplied by the South African Weather Bureau were used to classify the synoptic circulation patterns over the Eastern Cape region. The micrometeorological data collected at 10 m included wind direction and speed, dry and wet-bulb temperature, global solar radiation and surface pressure. These were used in turn to calculate the humidity, surface wetness and the standard deviation of the wind direction. Monthly three-hourly wind roses were constructed to determine diurnal and seasonal winds and potential pollution source patterns. The wind roses indicate direction and speed frequencies for 22.5° sectors. Winds with speeds below 2 m s^{-1} were regarded as calm and discarded from the analysis.

The southern African haze layer is known to extend over much of southern Africa, including the region of Ben Macdhui.[10,20] The depth through which the aerosols and trace gases that are present in the haze layer are transported is controlled by the presence of multiple layers of absolute stability.[10,17,20] Capping atmospheric layers of absolute stability over the sampling region were determined from daily South African Weather Bureau radiosonde observations at Bloemfontein, Pretoria, Port Eliza-

beth and Durban. Since the bulk of pollutants are transported below the first two capped atmospheric layers of absolute stability, namely the 700 hPa and 500 hPa levels, and the 850 hPa layer over coastal regions, only these three layers were examined.[10,11,16,20,29]

Pilot balloon ascents (PIBAL) were launched thrice daily, at 08:00, 13:00 and 16:00, during the winter campaign from 9 to 23 June 1996, in order to determine the extent of orographically induced controls on the local circulations at the sampling site. Upper-air winds were recorded at a site 300 m below the summit of Ben Macdhui at an altitude of 2700 m. A total of 45 ascents were undertaken during this period.

The ascents were grouped into distinct categories based on similarities in the wind-speed and direction profiles. Of the total ascents, 78 % were categorized. Wind direction and velocity vectors for each categorized ascent were resolved into components and the mean profiles determined for each category.

Preliminary results and discussion

Atmospheric chemistry. Results have been obtained for the long-term streaker sites and the two intensive sampling campaigns. Particulate streaker data for the year 1995/96 have been analysed and apportioned to the contributing sources. Four sources were identified in the coarse and fine fractions: soil, industrial, biomass burning, and marine (Fig. 5). In the coarse fraction the soil and marine components are most prominent, contributing 40–60 % of the total detected inorganic aerosol. Coarse-fraction sulphur was identified although the nature of the source is unclear. In the fine fraction, fine sulphur and iron, comprising the industrial components, fine potassium, related to biomass burning emissions, and fine silicon and aluminium from the soil, were most abundant. The industrial component was by far the largest contributor (47 %) to the fine-fraction aerosol load. Surprisingly, the total mass fraction of the fine fraction was equal to and in some instances even exceeded that of the coarse fraction (Fig. 6). This is an unexpected result as it suggests that the contributions of distant industrial emissions at this remote site are at least as abundant as the contribution from the local soil.

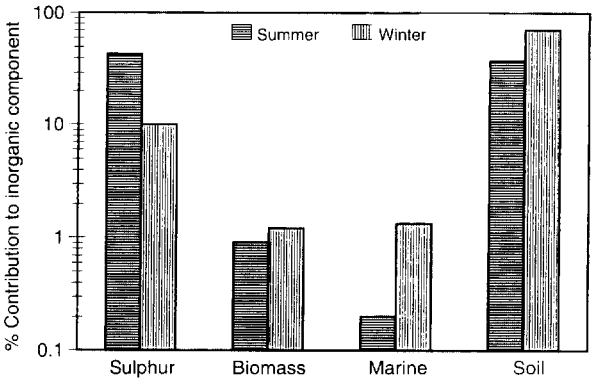

Fig. 5. Source apportionment results of both the long-term streaker sampling and the intensive sampling.

Analysis for ionic species found PO_4^{3-}, SO_4^{2-}, NO_3^-, C_2O_4, NH_4^+, Cl^- and K^+ during both the winter and summer intensive-sampling campaigns. The sulphate concentrations track the elemental sulphur data closely. In addition the SO_4^{2-} and NH_4^+ concentrations are highly correlated to one another ($r^2 = 0.7$) (Fig. 7). It is expected that most of the particulate SO_4^{2-} occurs over the continent as ammonium sulphate [$(NH_4)_2SO_4$]. The ammonium acts as a neutralizing agent of the acidic H_2SO_4. The degree of neutralization depends on the supply of NH_3 relative to the rate of formation of H_2SO_4.[26] The molar ratio NH_4^+/SO_4^{2-} ranges between 1 and 2. Sources of atmospheric ammonium include NH_4SO_4 and $(NH_4)_2SO_4$. The molar ratio for the Ben Macdhui data set ranges between 1 and 3, with the majority of the ratios (60 %) being close to 2. This implies that the dominant sulphate species at remote sites in southern Africa is $(NH_4)_2SO_4$.

The nitrogen ($\delta^{15}N$) and carbon ($\delta^{13}C$) stable isotope data for the summer period showed distinctive ratios for given periods. The $\delta^{15}N$ data can be separated into two categories, –2.1 to –4.3 ‰ and 5.4 to 12.9 ‰. The $\delta^{13}C$ data can also be divided into two distinctive categories, values above –26 ‰ and below –26.5 ‰ occurring at similar times to the $\delta^{15}N$ division. $\delta^{13}C$ and SO_4 data, however, showed the closest correlation. Low

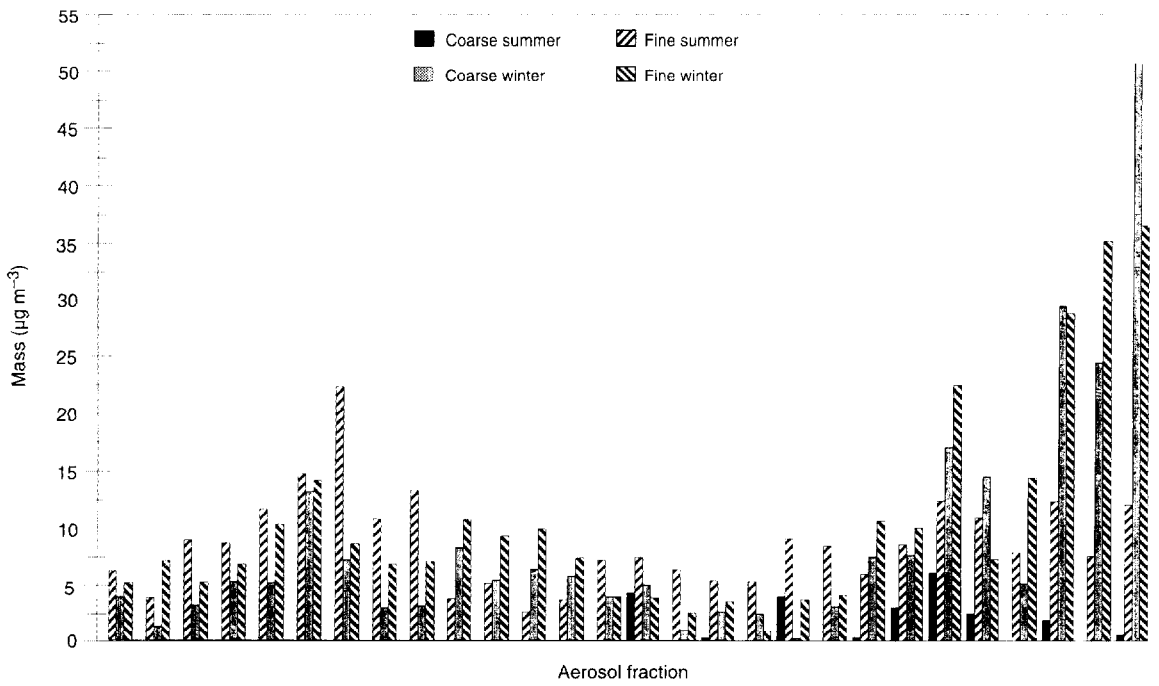

Fig. 6. Total mass loading in both the coarse and the fine fraction of the stacked filter unit samples. The fine-fraction mass equalled or exceeded the coarse-fraction mass.

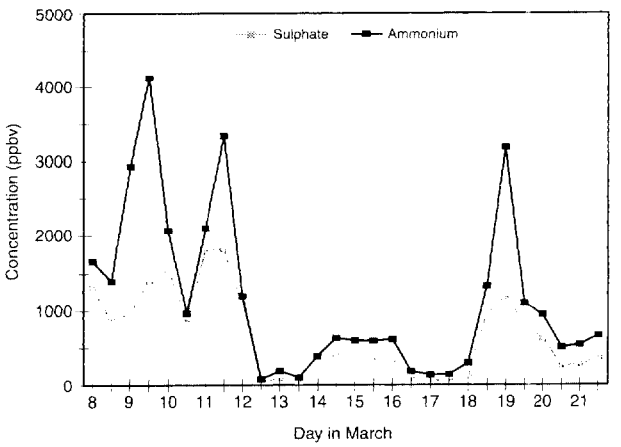

Fig. 7. Time series of SO_4^{2-} and NH_4^+ showing a high correlation between the two species in the atmosphere for the summer intensive-sampling campaign (8–22 March 1996).

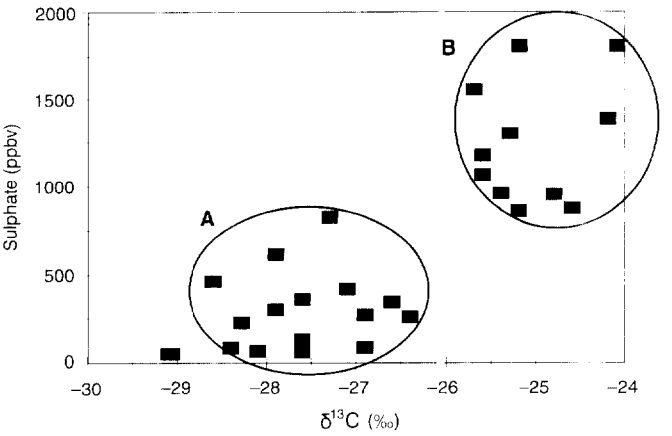

Fig. 8. A high correlation exists between the $\delta^{13}C$ and SO_4^{2-} data series with a clear grouping of $\delta^{13}C$ values related to both low and high concentrations of SO_4^{2-}.

concentrations of SO_4 were associated with the first $\delta^{13}C$ category, while elevated SO_4 concentrations corresponded to $\delta^{13}C$ ratios between –24 and –26 ‰ (Fig. 8). Previously, $\delta^{13}C$ ratios between –20 and –40 ‰ have been ascribed to fossil fuels, plants and marine organisms.[25] The present data set can now be used to refine this classification for southern Africa. $\delta^{13}C$ ratios between –24 and –26 ‰ are likely to be related to fossil-fuel burning.

Low-level airflow. During the months July 1995 to June 1996, mean day- and night-time surface winds over Ben Macdhui were predominantly northerly to north-northwesterly (with a mean frequency of 53 %), with northwesterly winds (with a mean of 21 %) being the next most frequent. This wind structure is largely attributable to the high-pressure system that prevails throughout the year over the interior of southern Africa. The dominant northerly winds at Ben Macdhui were observed less frequently during the austral summer (Fig. 9). Throughout the sampling period, low-level wind speeds from all directions were below 9 m s^{-1}. The only exceptions were the gradient winds from the north and northwest, when winds reached speeds exceeding 30 m s^{-1} and 17–30 m s^{-1} respectively.

Orographically-induced modifications to the wind field. The wind velocity profiles over Ben Macdhui indicated the occurrence of two major flow types: lee eddies and jets as the air descended over the Drakensberg mountains toward the coast (Fig. 10a,b), and low-velocity flow off the southern African plateau and up the mountain slopes within an absolutely stable layer (Fig. 10c,d). These wind systems operating over the complex terrain of the Eastern Cape are described in detail in Freiman *et al.*[30]

Absolutely stable layers. The daily occurrence of the absolutely stable layers during the months of March and June 1996 fell into one of the three broad categories chosen for this analysis: the lowest layer occurred below the mean height of the escarpment at about 850 hPa, the second was observed at approximately 700 hPa, and the next at approximately 500 hPa. All layers appeared as a wave-like continuity throughout the months. The layers assigned to the 850 hPa level were exclusively over the coastal stations. Table 3 illustrates the mean percentage frequency of occurrence of each layer for March and June 1996. The absolutely stable layers at both 700 and 500 hPa were present at all stations for more than 75 % of the time. These results correspond well with the results published by Cosijn and Tyson.[20]

The mean spatial characteristics of the elevated absolutely stable layers are illustrated along two transects, Pretoria – Durban and Pretoria – Port Elizabeth (Fig. 11a,b). The mean absolutely stable layers were generally lower at both the plateau and coastal stations during June 1996. The depths of the 700 hPa and 500 hPa layers over the interior were greater in March 1996.[39] The persistance of the 700 hPa absolutely stable layer would ensure that emissions on the highveld would be trapped and transported over South Africa in increasing concentrations. Peak concentrations of sulphate were always associated with the existence of a stable layer at 700 hPa.

Discussion

The chemical data show four sources influencing the site: dust, industry, biomass burning, and marine. Detection of an industrial source and related atmospheric conditions provides a useful tool to integrate the various data sets. Figure 12 illustrates

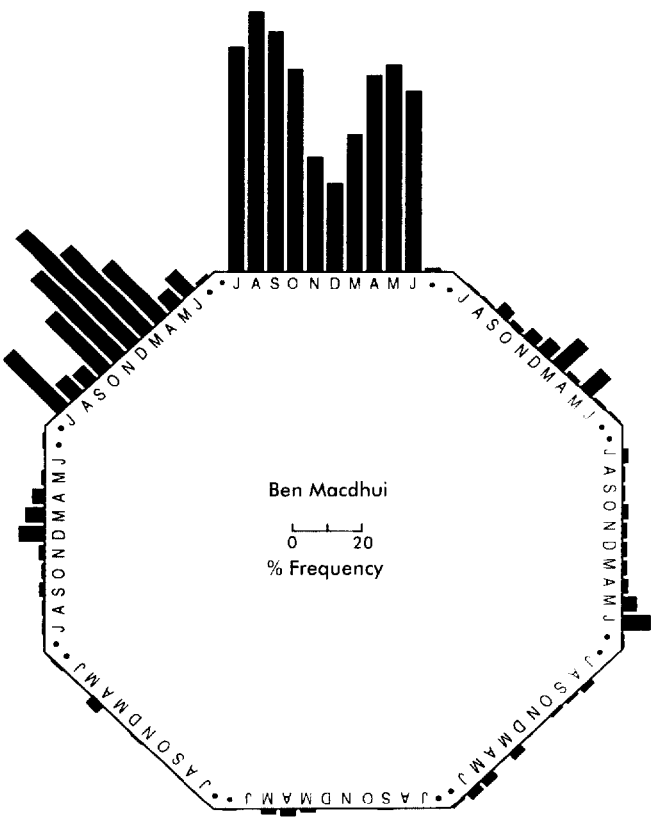

Fig. 9. Monthly variation of wind direction frequencies at Ben Macdhui (July 1995 – June 1996). Letters represent the names of the month (July – June).

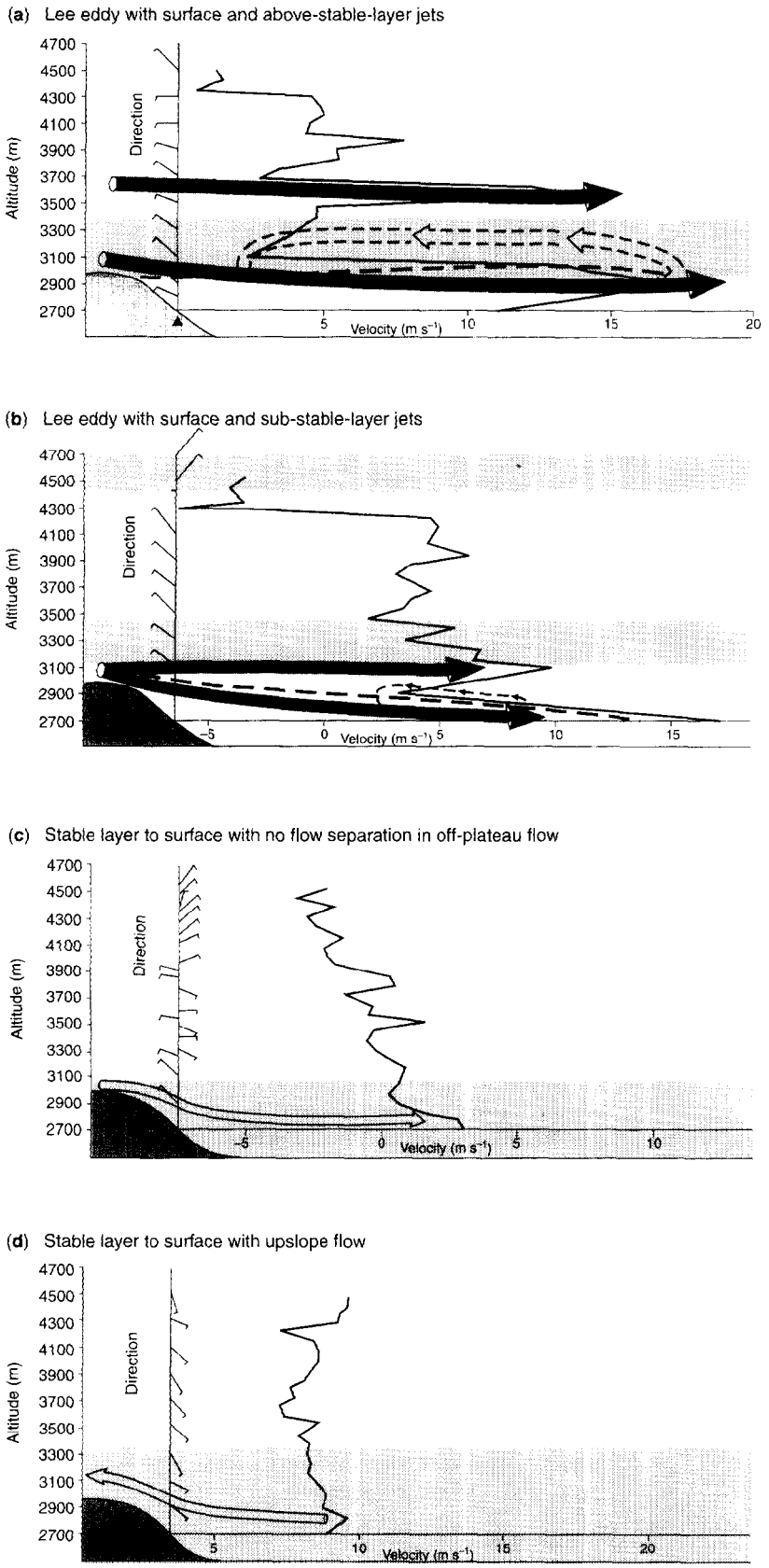

Fig. 10. a, b: lee eddies and upper-air jets as the air descends over the Drakensberg mountains. Low-velocity flow also occurs either down (**c**) or up (**d**) the mountain slope. Shaded arrows represent the direction of high-velocity airflow. Unshaded arrows represent the direction of low-velocity airflow. Positive wind speed represents westerly airflow. Wind feathers indicate wind direction into *y*-axis. Shading indicates the mean altitude of absolutely stable layers. Thick dashed lines indicate the boundary of flow separation and thin dashed lines represent return flow.

the concentration of fine sulphur detected at Ben Macdhui as well as the daily synoptic conditions and stability structure of the atmosphere during March 1996. The 500 hPa absolutely stable layer was temporally persistent irrespective of the synoptic influence at Ben Macdhui. The 700 hPa absolutely stable layer influences both the atmospheric chemistry over southern Africa and the prevailing wind systems for a given region. Sulphur concentrations increase steadily with time as a high-pressure system develops over the subcontinent. The presence of the absolutely stable layers, particularly at 700 hPa, facilitates increased concentrations of industrial sulphur long distances from the interior plateau source region. On the days when the mean absolutely stable layer at 700 hPa was located at or above the summit of Ben Macdhui and jet flow was observed on the lee side (Fig. 10a,b), the 700 hPa synoptic conditions over the mountain were dominated by a strong winter continental high-pressure system and occasional westerly trough. The mean wind direction was northwest to west.[30] On days when airflow off the escarpment was light (Fig. 10c) or was directed upslope (Fig. 10d) within coupled 700 and 850 hPa absolutely stable layers, synoptic conditions were dominated by ridging anticyclones. Industrial sulphur concentrations dropped, as onshore airflow was from the southern Indian Ocean. The 700 and 850 hPa absolutely stable layers break down with the passage of a cold front. The atmosphere over Ben Macdhui and the interior as a whole is then cleansed of anthropogenic aerosols and trace gases.

Summary

To test the hypothesis that atmospheric material is circulated and recirculated over southern Africa, a long-term time-resolving particulate sampler was commissioned at Ben Macdhui in the Eastern Cape. Episodes of elevated, industrially related aerosols were identified. Two intensive sampling campaigns were organized to obtain detailed chemical characterization of the industrial emissions detected at the sampling site.

Initial results from the summer and winter campaigns indicated four contributing sources at Ben Macdhui: soil, industry, biomass burning, and marine. The industrial component was the dominant source in the fine fraction of the aerosol. The contribution from soil was related to both local and remote sources. Soil in the fine fraction was considered to be of a more remote origin.

The ionic species identified in the sampled aerosols included PO_4^{3-}, SO_4^{2-}, NO_3^-, $C_2O_4^-$, NH_4^+, Cl^- and K^+. These species confirmed the apportionment of the inorganic elements. The good correlation between SO_4^{2-} and NH_4^+ as well as their related molar ratio, indicates that $(NH_4)_2SO_4$ is the dominant component for

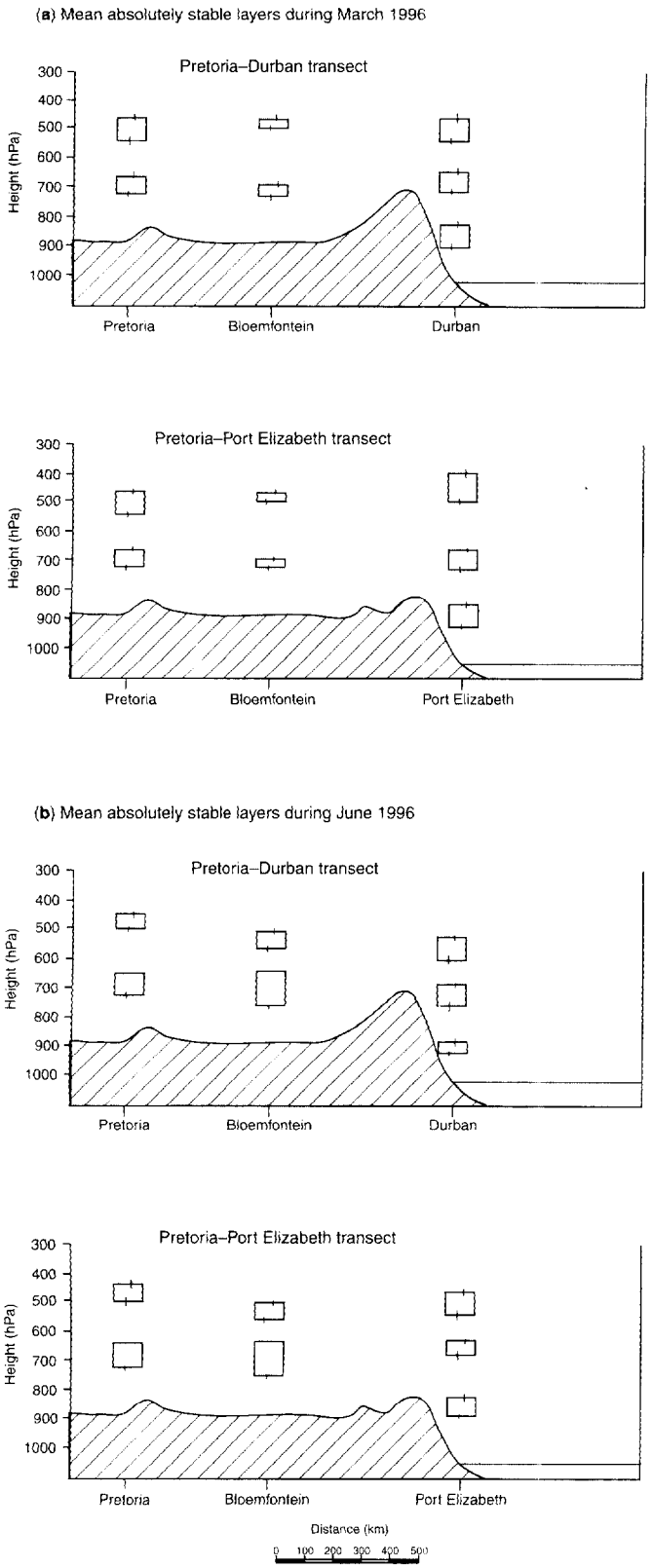

Fig. 11. Mean absolutely stable layers and standard deviation along two transects during (**a**) March and (**b**) June 1996.

atmospheric sulphate at a remote site in southern Africa.

Stable isotopic ratios, $\delta^{15}N$ and $\delta^{13}C$, showed distinctive characteristics for periods of elevated industrial particulates. In particular the industrial component, which is mainly related to fossil-fuel burning, is characterized by $\delta^{13}C$ ratios of between –24 and –26 ‰.

Table 3. The mean percentage frequency of occurrence and standard deviation of the absolutely stable layers during March and June 1996.

Pressure level	850 hPa		700 hPa		500 hPa	
Month	March	June	March	June	March	June
Plateau						
Pretoria	—	—	81	97	97	87
Bloemfontein	—	—	65	93	84	73
Mean	—	—	73 (8)*	95 (2)	90 (6)	80 (7)
Coast						
Durban	100	87	77	83	90	77
Port Elizabeth	90	83	81	67	90	73
Mean	95 (5)	85 (2)	79 (2)	75 (8)	90 (0)	75 (2)

*(Standard deviation).

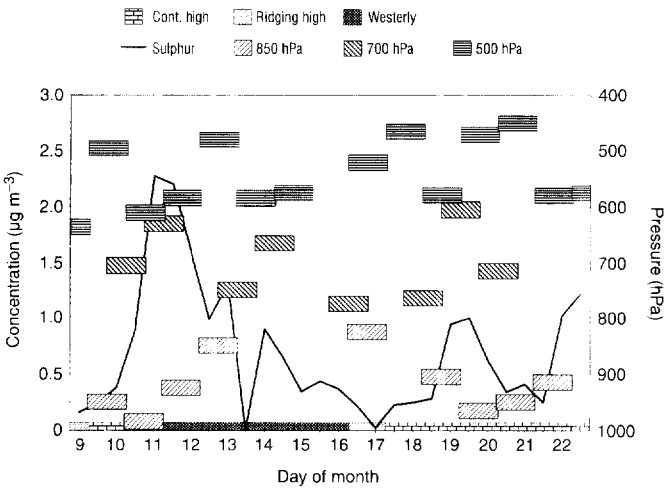

Fig. 12. Absolutely stable layers over Durban in June 1996. Shaded bars represent the vertical extent of the identified absolutely stable layer. Solid lines indicate continuity between days.

The dominant surface-level air flow was northerly to north-northwesterly (53 %) and northwesterly (21 %). This flow regime is related to the persistent anticyclonic circulation that prevails over the South African interior. Lee eddie jets and low-velocity flow were found to occur over Ben Macdhui from PIBAL ascents.

Analysis of the absolutely stable layer above Pretoria, Bloemfontein, Durban and Port Elizabeth showed spatially and temporally persistent layers at 700 and 500 hPa at the four stations, and additionally at 850 hPa at the coastal stations.

The results of trajectory analysis, trace gas data and characterization of the local meteorology will be presented in future papers. From the data presented here it is clear that the methods used for testing our hypothesis were ideal. Continuing research at Ben Macdhui aims to characterize the long-term trends of trace gas concentrations at the site as well as the optical properties of the troposphere.

Funding for this research was provided by Eskom Technology, Research and Investigations and the University of the Witwatersrand. S. Macko (University of Virginia) and R. Talbot (University of New Hampshire) are thanked for sampling equipment and analysis of samples. P. Sephton, J. Sephton and I. van Eck are thanked for logistical support.

Received 24 February 1998. Accepted 13 September 1998.

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