

Characterization and transport of aerosols over equatorial eastern Africa

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Abstract. Measurements of the composition of aerosol partitioned into two size fractions, fine (particle aerodynamic equivalent diameter, $d_p \leq 2.5 \mu\text{m}$) and coarse ($2.5 < d_p \leq 10 \mu\text{m}$) were made at a high-altitude site over equatorial eastern Africa on Mount Kenya to study long-range transport of aerosol and to determine the extent of interhemispheric transport at the equator. The two size fractions allow long-range transport aerosol to be distinguished from those more locally derived. Background midtropospheric elemental aerosol concentrations observed on Mount Kenya, for example, $S = 0.0157 \mu\text{g m}^{-3}$, are lower than commensurate findings for remote areas in industrialized countries and elsewhere in Africa but are 3 factors higher than concentrations reported from the South Pole. Locally derived aerosols are frequently transported up the slopes of Mount Kenya by day in valley winds but are rarely transported at night in the stable conditions characterized by downslope katabatic flow. Nocturnal conditions favor the measurement of long-range, midtropospheric transport products to Kenya from afar. Case studies reveal that anthropogenically derived S and Fe aerosols from both southernmost and central southern Africa may be observed on Mount Kenya after transport over distances $>6000 \text{ km}$. So-called pristine marine air from the central Indian Ocean is shown to contain aged and recirculated Si that could only have been derived from land areas after transport over long distances. The nature of the aerosols measured on Mount Kenya depends critically on regional patterns of aerosol transport. Interregional transfers seem to be a feature of the transport climatology. Likewise, interhemispheric transport across the equator in east Africa is observed.

1. Introduction

Interregional, intercontinental, and hemispheric transports of aerosols are known to occur frequently. For example, in the Northern Hemisphere, mineral dust from the Sahara Desert has been reported thousands of kilometers from the source in the Caribbean, Amazon, Europe, and the Near East [Darwin, 1846; Pitty, 1968; Carlson and Prospero, 1972; Schütz, 1980; Talbot *et al.*, 1986, 1996; Swap *et al.*, 1992]. In the Southern Hemisphere, transport of continentally derived material from southern Africa has been observed over the middle of the Indian and Atlantic Oceans [Bigg and Turvey, 1978; Heintzenberg and Bigg, 1990; Moody *et al.*, 1991; Balkanski *et al.*, 1993; Garstang *et al.*, 1996; Swap *et al.*, 1996; Tyson *et al.*, 1996a, 1996b; Herman *et al.*, 1997]. In contrast, few studies have reported interhemispheric transport of aerosols and trace gases, particularly over equatorial Africa. Studies in the region such as Dynamique et Chimie Atmosphérique en Fôret Equatoriale (DECAFE) and Fires of Savannah (FOS) (special issues of *Journal of Geophysical*

Research, 97(D6), 6123–6218, 1992 and *Journal of Atmospheric Chemistry*, 22, 1–148, 1995) focused on aerosol and trace gas emissions from rain forest and savanna but not on interhemispheric transfers. A study demonstrating the potential for such transfers is that of Krishnamurti *et al.* [1996], which shows that biomass products from Asian monsoon activity areas may be transported to equatorial and tropical eastern Africa. The recent Indian Ocean Experiment (INDOEX) [Lelieveld *et al.*, 2001] demonstrates long-range transport of air pollution from south and Southeast Asia toward the Indian Ocean to the location of the Intertropical Convergence Zone (ITCZ), the location of which varied between the equator and 12°S during the dry monsoon season in January to March 1999. Similar transport had been observed during the pre-INDOEX period conducted in January–February 1996 [Krishnamurti *et al.*, 1998]. Considering the potential impact of aerosols on regional and global climates [Charlson *et al.*, 1987; Schwartz, 1988; Charlson *et al.*, 1992; Andreae, 1995; Intergovernmental Panel on Climate Change, 1996; Andreae and Crutzen, 1997], a need exists to determine the extent to which major transequatorial transfers of aerosols may take place.

In order to address this question, an equatorial, high-altitude remote site was established on Mount Kenya for the sampling and characterizing of aerosols transported to and from the site. In this paper the nature of aerosols reaching Mount Kenya will be determined, and an attempt will be made to show the nature of interhemispheric regional atmospheric transport of aerosols across the equator in Africa. Previous studies of transport over Mount Kenya have shown that transport corridors to Kenya are clearly bounded and are well defined [Gatebe *et al.*, 1999]. Air reaching the country originates mainly from the Saharan region and northwestern Indian Ocean of the Arabian Sea in the Northern Hemisphere and from the Madagascan region of the Indian Ocean in the Southern Hemisphere. Air reaching Kenya between 700 and 500 hPa is mainly from the Sahara and northwest Indian Ocean in the

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months of January–March, which gives way to southwest Indian Ocean flow in May–November.

2. Data and Methods

Aerosol data for the period August 12 to September 9, 1997, have been used in this study. They were collected on the equator at an altitude of ~ 4420 m on the southwestern slope of Mount Kenya (1°S , 37.2°E at summit altitude 5199 m), approximately 480 km inland of the Indian Ocean and 190 km northeast of Nairobi. The sampling site is assumed to be representative free-air, midtropospheric conditions and largely removed from the influence of industrial sources, especially during the night when katabatic flow down the mountain [Winiger, 1986] ensures air is uncontaminated by local sources. Sampling was undertaken 3 m above the surface on a remote site covered by grassy patches and rock debris and at an altitude above the trade wind inversion, which occurs on average at ~ 3600 m [Winiger, 1986]. During the day, well-mixed air masses may be sampled during late morning and afternoon with anabatic upslope flow [Schnell *et al.*, 1978; Davies *et al.*, 1977].

A two-stage streaker sampler [cf. Annegarn *et al.*, 1988; Gatebe, 1999] was used to collect samples at a rate of approximately 1 L min^{-1} . Consequently, particles were partitioned separately into fine-fraction, particle-size equivalent aerodynamic diameter $d_p \leq 2.5 \mu\text{m}$ and coarse fraction $2.5 < d_p \leq 10 \mu\text{m}$ at mean sea level. However, at our sampling altitude, 4420 m above mean sea level, a pressure of 588 hPa (0.58 atm) and assuming a Brailsford pump maximum capability of 0.4 m (17 inches of vacuum), the flow rate is expected to average $\sim 0.5 \text{ L min}^{-1}$ and a particle-size equivalent aerodynamic diameter, $d_p \leq 2.1 \mu\text{m}$ for fine fraction and $2.1 < d_p \leq 8 \mu\text{m}$ for coarse fraction (W. Nelson, personal communication, PIXE International Corporation, Tallahassee, Florida, United States, 1999). The samples were continuously taken over a period of 28 days with a time resolution of ~ 4 hours.

Samples were analyzed at PIXE Analytical Laboratories, Tallahassee, Florida, using a 2.4-meV proton beam from a Tandatron accelerator [Bauman and Nelson, 1985]. The circular streaker sample frame was inserted into a sample manipulator that automatically steps the filter in 168, 1 mm-wide increments, equivalent to a time resolution of 4 sampling hours for each step. Each sample was irradiated, with a 1×7 mm collimated beam, for 6 min (120 s with the X-ray absorber out and 240 s with the absorber in). Because the streaker orifice moves and samples continuously along the filter, whereas the PIXE analysis is performed on consecutive 1-mm segments of the sample streak, every measurement is serially correlated by 25% with the immediate and subsequent measurements [cf. Darzi, 1982]. Therefore each concentration is only 50% representative of the ambient concentration during the 4-hour time step. The calculations of amounts were made using a calibration based on thin-film standards of the elements, and analytical detection limit for each element was based on 2 standard deviations of the background noise in the corresponding region of the spectrum [cf. Maenhaut and Vandenhaut, 1986; Bodhaine *et al.*, 1987; Gatebe, 1999]. Twenty-one elements: Na, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, As, Se, Br, Sr, and Pb were detected at the 95% confidence level (see Tables 1 and 2).

To understand the composition of aerosol sources at Mount Kenya, various interpretative techniques were employed, including trajectory analyses, elemental enrichment factors, and source identification using multivariate models. In this study we use a gridded three-dimensional wind vector field from the European Centre for Medium-Range Weather Forecasts (ECMWF) [Lambert, 1988] to compute, at best, an approximate indication of the mean motion of an advected air parcel. The ECMWF data are 6-hourly operational analyses of the three-dimensional wind field at 1000-, 925-, 850-, 700-, 500-, 400-, 300-, 200-, and 100-hPa geopotential

height levels. The data provide the basis for deriving daily air transport fields to and from Mount Kenya [Gatebe *et al.*, 1999]. The ECMWF data have been shown to be of high quality for this kind of analysis [Pickering *et al.*, 1994, 1996]. Transport pathways were determined by Lagrangian kinematic trajectory modeling using the *D'Abreton* [1996] model. The method is based on advection of air parcels with u , v , and w wind components with iterative steps of 15 min to produce 10-day back trajectories to and forward trajectories from the point of origin. Vertical velocities are determined from nonlinear mode initialization, which permits the diabatic as well as the adiabatic processes to be taken into account. The boundary condition of zero vertical motion at the surface and taking into account atmospheric stability, permits trajectories to follow the terrain or circumvent obstacles when trajectories approach the surface. An ensemble of five trajectories spaced 1° of latitude and longitude apart, with an origin at the centroid on Mount Kenya, were determined for each run. The origin has been taken to be at 500 hPa, a mean level approximating the altitude of the sampling site.

3. Results

3.1. Aerosol Characterization

As theory requires [Kemp and Möller, 1981; Johansson, 1995], analytical detection limits of the 21 identified elements decrease with increasing atomic number and are lowest for transition elements (Tables 1 and 2). In the fine fraction, eight elements contribute $\sim 95\%$ of the total aerosol load of the air being transported to Mount Kenya (Table 1). They are Si (48.9%), S (17.6%), Na (12.7%), Cl (4.8%), K (4.1%), Ca (2.6%), Fe (2.2%), and P (2.1%). Si has a median concentration of $0.481 \mu\text{g m}^{-3}$, and S has a median concentration of $0.157 \mu\text{g m}^{-3}$. All other aerosol components have elemental median concentrations an order of magnitude lower. Episodes in which highest maximum concentrations are observed are associated with high Si and S fluxes. Other high flux episodes are linked to high concentrations of K, Na, Cl, Ca, and Fe, all associated with particular sources and modes of transport.

In the coarse aerosol fraction, 11 elements contribute to $>94\%$ of the total elemental mass (Table 2). They are Si (23.2%), Na (16.4%), Pb (12.0%), Fe (8.5%), Ca (8.0%), Al (6.7%), As (6.6%), P (6.3%), Cl (4.2%), Sr (1.7%), and Ti (1.1%). Highest median concentrations are Si ($0.342 \mu\text{g m}^{-3}$), Fe ($0.118 \mu\text{g m}^{-3}$), Ca ($0.082 \mu\text{g m}^{-3}$), and Al ($0.074 \mu\text{g m}^{-3}$), all crustal constituents carried in aeolian dust. The maximum concentration of Ca observed is $0.759 \mu\text{g m}^{-3}$, that of Na is $0.712 \mu\text{g m}^{-3}$ (associated mainly with marine source sand), and that of Pb is $0.041 \mu\text{g m}^{-3}$ (mainly from local vehicular emissions).

Silicon is the most common constituent of both fine- and coarse-fraction midtropospheric dust reaching Mount Kenya. Sulfur is significant only in the fine fraction, and Pb is significant only in the coarse fraction. Sodium constitutes a significant portion of the aerosol loading in both fine and coarse fractions (13% and 16%, respectively), but it must be used with caution in interpreting the sources of aerosols impacting on Mount Kenya, since self-absorption of X rays may take place during PIXE analysis [Asking *et al.*, 1987].

For most elements, concentration levels are lower than those reported for remote sites in industrialized countries, for example, S = $0.315 \mu\text{g m}^{-3}$ in the western United States [cf. Kretzschmar *et al.*, 1977; Lanefors *et al.*, 1977; Moyers *et al.*, 1977; Adams *et al.*, 1980; Priest *et al.*, 1981]. The concentrations are also lower than elsewhere in Africa, for example, S $\sim 1.2 \pm 0.3 \mu\text{g m}^{-3}$ from the high veld region of South Africa [Piketh *et al.*, 1999]. They are nearly 3 times lower than those observed in rural north China, for example, S = $0.05 \mu\text{g m}^{-3}$, associated with equatorward aerosol

Table 1. Fine-Fraction Elemental Detection Limit, Percentage of Weight Fraction, and Median and Maximum Concentration of the Elements Present in the Sample on Mount Kenya at 4420 m Above Mean Sea Level During the August to September 1997 Period

Elements	Detection Limit, $\mu\text{g m}^{-3}$	Percentage of Detected Mass, $\mu\text{g m}^{-2}$	Cumulative Percentage	Median Concentration, $\mu\text{g m}^{-3}$	Maximum Concentration, $\mu\text{g m}^{-3}$
Si	0.021	48.9	48.9	0.481	1.100
S	0.017	17.6	66.5	0.157	1.056
Na	0.065	12.7	79.2	<0.067	0.191
Cl	0.016	4.8	84.0	0.046	0.165
K	0.012	4.1	88.1	0.030	0.25
Ca	0.010	2.6	90.7	0.014	0.121
Fe	0.003	2.2	92.9	0.019	0.110
P	0.022	2.1	95.0	<0.022	0.037
V	0.005	0.9	95.9	<0.005	0.013
Ti	0.006	0.8	96.7	<0.007	0.019
Pb	0.008	0.8	97.5	<0.008	0.023
Cr	0.004	0.7	98.2	<0.004	0.011
Br	0.005	0.5	98.7	<0.005	0.015
Se	0.003	0.4	99.1	<0.003	0.008
Mn	0.003	0.2	99.3	<0.003	0.008
Cu	0.002	0.2	99.5	<0.002	0.006
Zn	0.002	0.2	99.7	<0.002	0.006
As	0.003	0.2	99.9	<0.002	0.008
Ni	0.002	0.1	100	<0.002	0.005

transport from Siberia and Mongolia [Winchester *et al.*, 1981], and those reported at the geographic South Pole, for example, $S = 0.049 \pm 0.01 \mu\text{g m}^{-3}$ [Maenhaut *et al.*, 1979].

3.2. Aerosol Enrichment

The ratio of the measured atmospheric concentration of a given element to that of a reference element, in this case Si or Fe, and relative to the ratio of that element concentration to the reference element in a source of interest, crustal or marine, is defined here as the enrichment factor. If the factor is close to unity, this denotes a crustal source for aerosols [cf. Maenhaut *et al.*, 1996]. A standard crustal aerosol composition for the East Africa region, of a type advocated by Lawson and Winchester [1979], would have been ideal for the characterization of Mount Kenya aerosols. No such data are available; in its absence the Mason and Moore [1982]

global average crustal composition has been used instead. Enrichment factors with respect to Fe are lower than those referenced against Si by a factor of around 0.5 for most elements.

In the fine fraction, Mount Kenya aerosols are significantly enriched (Figure 1a). Highest enrichments are observed for Br, Pb, S, and Cl. Most other elements are significantly enhanced. Only K, Ca, and Fe are depleted (having a ratio of less than unity). Similar enrichments have been reported for South Pole aerosols [Maenhaut *et al.*, 1979], the North Atlantic Ocean area [Duce *et al.*, 1975], Bermuda Duce and Hoffman, 1976], Hawaii [Hoffman *et al.*, 1972], Jungfrauoch, Switzerland, and northern Norway [Rahn, 1972, 1976]. In the coarse fraction most noncrustal elements are enriched by factors greater than 10. Only K, Si, and Al approximate the reference standards. In both fine and coarse fractions, elements with high enrichment factors may be attributed

Table 2. Coarse-Fraction Elemental Detection Limit, Percentage of Weight Fraction, and Median and Maximum Concentration of the Elements Present in the Sample on Mount Kenya at 4420 m Above Mean Sea Level During the August to September 1997 Period

Elements	Detection Limit, $\mu\text{g m}^{-3}$	Percentage of Detected Mass, $\mu\text{g m}^{-2}$	Cumulative Percentage	Median Concentration, $\mu\text{g m}^{-3}$	Maximum Concentration, $\mu\text{g m}^{-3}$
Si	0.018	23.2	23.2	0.342	1.368
Na	0.053	16.4	39.6	<0.065	0.712
Pb	0.005	12.0	51.6	<0.005	0.041
Fe	0.002	8.5	60.1	0.118	0.509
Ca	0.008	8.0	68.1	0.082	0.759
Al	0.023	6.7	74.1	0.074	0.403
As	0.002	6.6	81.4	<0.002	0.026
P	0.018	6.3	87.7	0.036	0.122
Cl	0.012	4.2	91.9	0.048	0.322
K	0.008	2.4	94.3	0.082	0.759
Sr	0.005	1.7	96.0	<0.005	0.022
Ti	0.005	1.1	97.1	0.015	0.061
Zn	0.001	1.0	98.1	0.010	0.065
Cu	0.001	0.6	98.7	<0.001	0.097
Mn	0.002	0.4	99.1	0.004	0.050
V	0.0048	0.3	99.4	<0.005	0.010
Cr	0.003	0.2	99.6	<0.003	0.008
Ni	0.002	0.2	99.8	<0.002	0.007
Br	0.003	0.2	100.0	<0.003	0.013

to a variety of anthropogenic sources [Maenhaut *et al.*, 1979; Arimoto *et al.*, 1987]. The bulk of aerosols passing over Mount Kenya appear to contain a significant anthropogenic component. This is in addition to local and remotely derived surface aeolian mineral dust, hereafter referred as aeolian dust.

Fine- to coarse-fraction ratios reveal that most of the 18 aerosol elements detected on Mount Kenya may be transported to the site from afar as fine dust (Figure 2). Missing from the ratio analysis is S. This is because it is present only in the fine fraction and is only associated with long-range transport. The fine-to-coarse ratios of K, Ca, and Fe are similar to those obtained over southern Africa [Maenhaut *et al.*, 1996]. The low ratios of fine-to-coarse Fe, Zn, and Ca indicate the local origin of the elements. When occurring in significant concentrations in the fine fraction, as happens in periodic episodes, Fe could be associated with long-range transport.

The principal component analysis is used here to identify the number of contributing sources and the contribution of each source to the measured concentration at Mount Kenya [cf. Henry *et al.*, 1984]. To adequately fingerprint each major source using this method, many source samples would be required. Clearly, such an effort was beyond the scope of our project. It is, however, useful to identify the most important ones. Consequently, we utilized a Statistical Package for the Social Sciences (SPSS) Standard Version (Release 8.0.0, December 1997) software to determine the

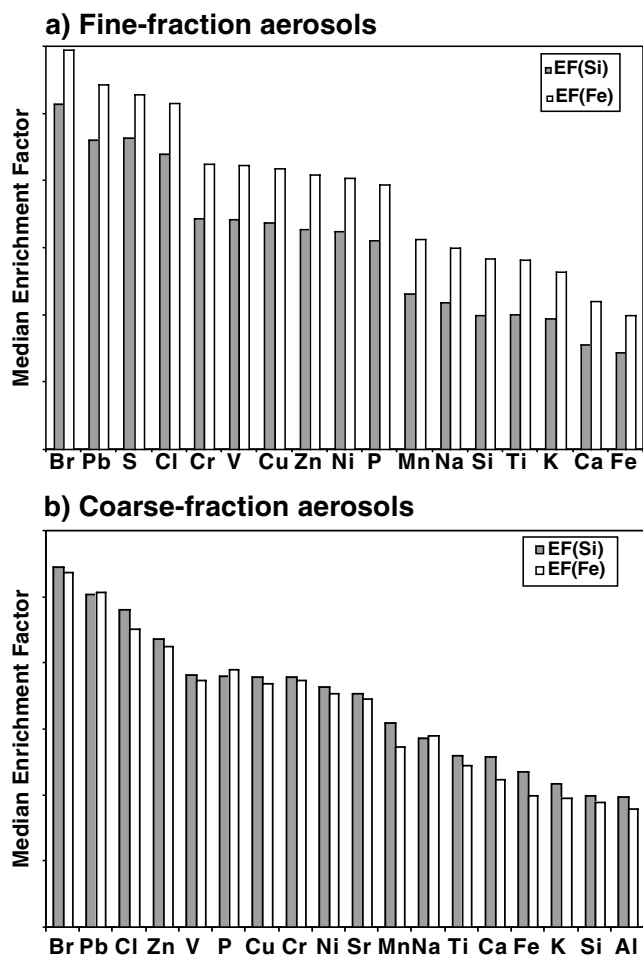


Figure 1. Median crustal enrichment factors for Mount Kenya aerosols relative to average crustal rock [Mason and Moore, 1982] using Si and Fe as reference elements: (a) for fine-fraction aerosols and (b) for coarse-fraction aerosols.

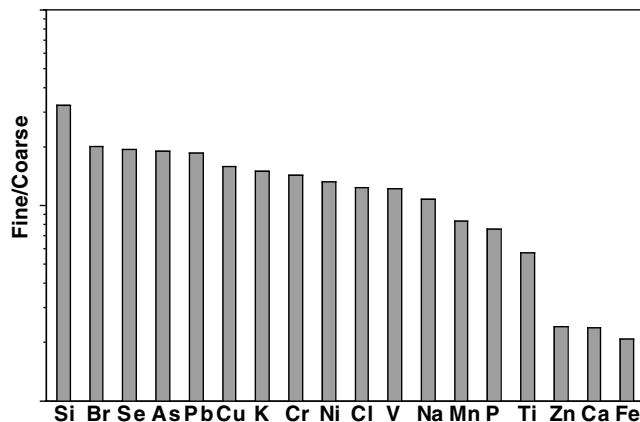


Figure 2. Ratios of mean fine-fraction concentrations to mean coarse-fraction concentrations for 18 important constituent elements of Mount Kenya aerosols.

principal components, with eigenvalue ≥ 1 and then rotated using the varimax procedure with Kaiser normalization [cf. Thurston, 1981, Thurston and Spengler, 1985]. The results show that aeolian aerosols, in the coarse fraction, constitute 58% of material transported to and from Mount Kenya. There is a large unexplained variance that may be accounted for by contribution from plants and marine sources. From the fine mode fraction, three sources can be delineated: anthropogenic mixed within mineral dust, explaining 48% of the total variance; pyrogenic, including biomass burning (18%); and marine (11%). The extent to which motor vehicle emissions may impact on the site is assessed from the ratio Br/Pb. Kenya uses Tel-B[®] as an antiknock agent which should produce a ratio of 0.77 in the atmosphere [Maenhaut and Akilimali, 1987]. The observed ratio at Mount Kenya varied between 0.48 and 1.16. Some, but not all, vehicular emission products were local and derived from sources in townships surrounding the mountain and were transported up the mountain in local up-valley winds [Gatebe, 1999]. The largest source area for vehicle emissions in Kenya is Nairobi [Gatebe *et al.*, 1994]. Emission products reaching the observation site from such urban sources would be carried by transport in the winds of the general circulation over the region.

3.3. Long-Range Transport of Trace Elements to Mount Kenya

Fine-fraction aerosols are commonly accepted to be good indicators of long-range transport [Li and Winchester, 1990; Thurston and Spengler, 1985; Junge, 1979]. In general, coarse aerosols tend to have local origins [Gatebe, 1999; Li and Winchester, 1990; Maenhaut *et al.*, 1996]. An exception is coarse-fraction Cl, which has been shown to be a clear indicator of transport from ocean areas [Maenhaut *et al.*, 1996]. It is used as such here.

In the absence of strong gradient winds, concentrations of coarse aerosols originating close to Mount Kenya show clear diurnal variability, as illustrated by Si (Figure 3a). The diurnal signal is the consequence of local thermotopographic winds induced by the mountain. During the day with warm upslope valley winds, concentrations increase, whereas at night they decrease during stable conditions associated with cool katabatic flow in downslope mountain winds. Diurnal variability is much less evident in fine-fraction long-range transport. With gradient winds strong enough to mask thermotopographically induced local winds, the diurnal signal weakens or disappears, and long-range transport effects become discernible. These manifest themselves in aperiodic episodes that are related to large-scale flow fields and emission sources.

During the day, local upslope flow may transport aerosols derived in the vicinity of Mount Kenya (e.g., in Nairobi) to the site. At night, reversed downslope flow will prevent this happening. Obtaining samples in the stable conditions on this

mountain gives the best estimation of background aerosol loading associated with transport from afar. To study long-range transport to and from Mount Kenya, a 36-hour moving average has been applied to the observed nocturnal aerosol concentrations. The

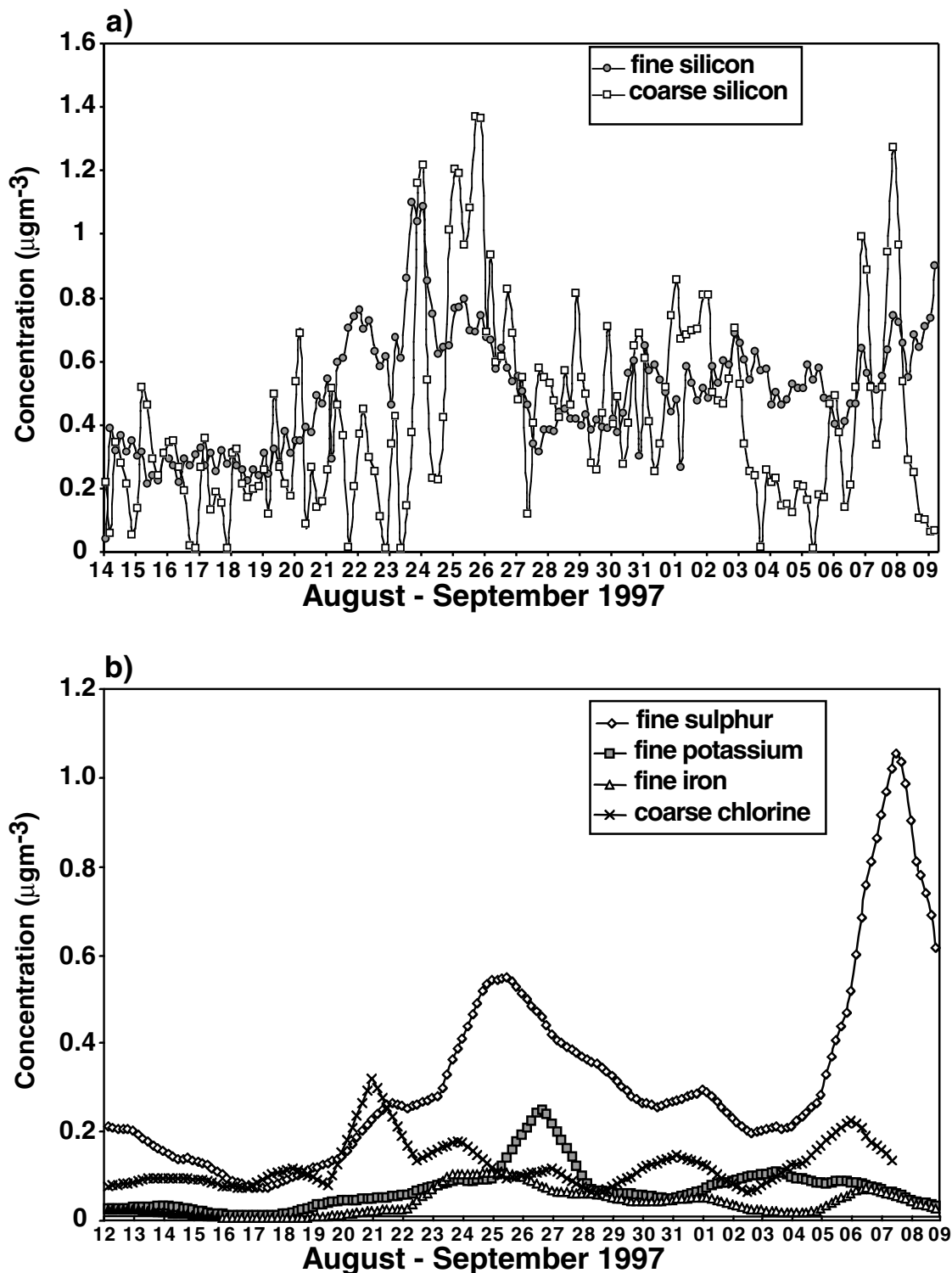


Figure 3. Time series of elements in Mount Kenya aerosols for the August–September 1997 case study. (a) Fine- and coarse-fraction Si. The diurnal effect of local winds on the coarse fraction is clear; the fine fraction transported from afar is largely immune to the diurnal cycle. (b) Episodes of high-concentration transport of fine S, K, and Fe and coarse Cl to Mount Kenya during August–September 1997.

episodic nature of such transport is clearly discernible from Figure 3b.

Four aerosol elements, S, Si, K, and Fe, together constitute about $\sim 73\%$ of the fine-fraction elemental aerosol loading in the long-range transport to Mount Kenya. They have been used, together with coarse Cl, to demonstrate distinctive regional and interhemispheric modes of transport to and from the Kenyan region of equatorial Africa for case studies between August 12 and September 9, 1997.

Different atmospheric transport modes greatly affect concentrations of S recorded on Mount Kenya (Figure 4). The transport modes are derived from a trajectory analysis [Gatebe *et al.*, 1999]. Long-range transport of aerosols containing negligible quantities of S is associated with the continuous recirculation of sulfur-free air over equatorial Africa or with northern hemisphere airflow from, for example, Egypt and the Arabian Peninsula. In contrast, high concentrations of background S in well-mixed air reaching Mount Kenya are observed following recirculation and transport

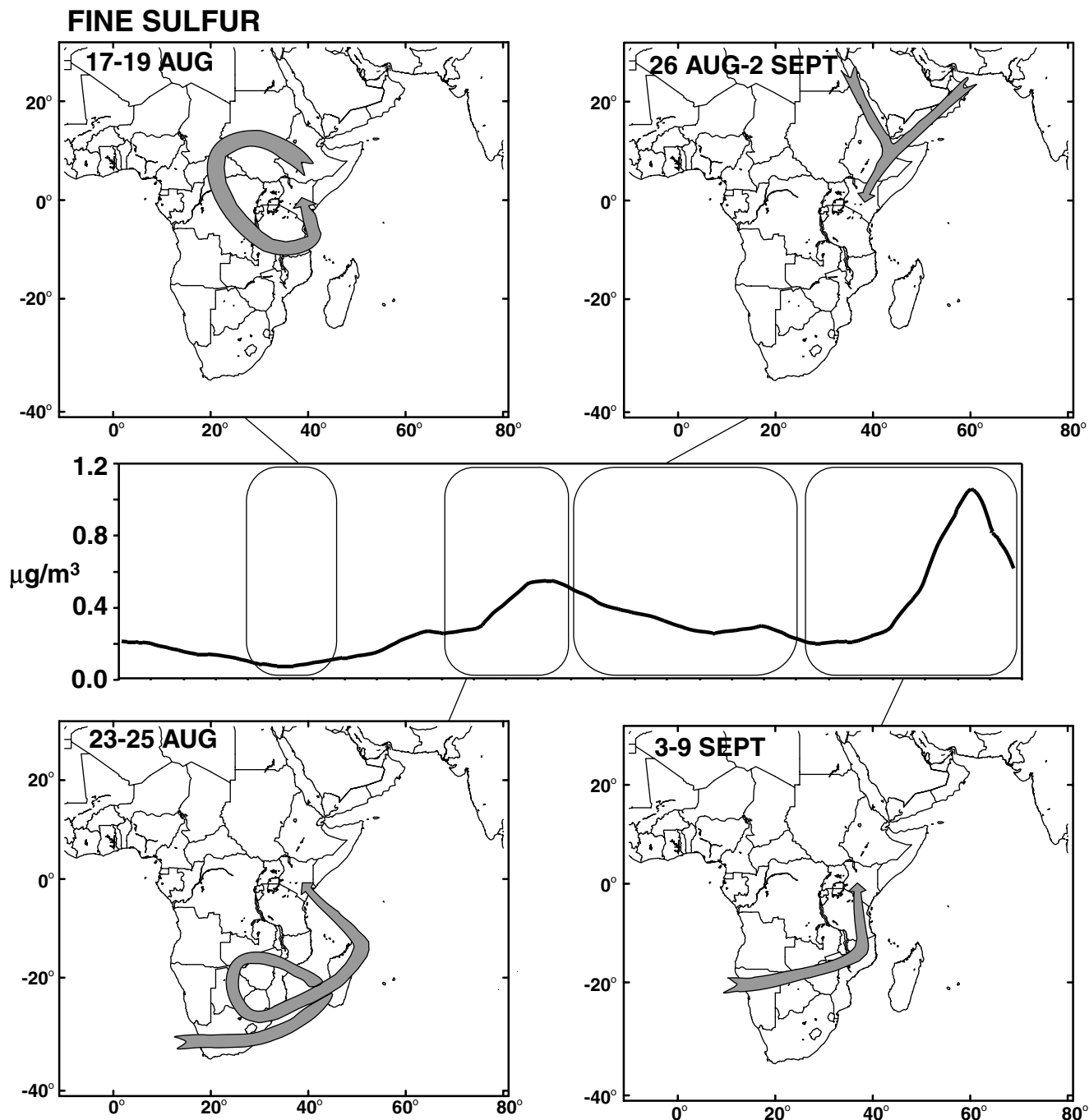


Figure 4. Transport pathways defined by clustered 500-hPa back trajectories indicating transport to Mount Kenya between August 17 and September 9, 1997. Periods for analysis have been chosen to explain distinctive episodes of fine-fraction S transport to the observation site at 4420 m on the mountain.

over the industrial heartland of South Africa ($\sim 0.6 \mu\text{g m}^{-3}$ between August 23 and 25) and transport over the Zambia/Congo copper belt region ($1.1 \mu\text{g m}^{-3}$ between September 3 and 9). Over northeastern South Africa and Zambia/Congo, copper belt region sulfur emissions are the highest in Africa [Zunckel *et al.*, 2000]. In the August episode the 6400-km pathway from South Africa to Mount Kenya was over Mozambique, Madagascar, and the Indian Ocean before recurving inland to reach the equator in Kenya in 5 days. The transport to Mount Kenya from northern Zambia during the September episode was continental and occurred over a pathway ~ 3600 km long, and the aerosol was only 3 days in transit. This explains the higher concentrations in the second episode. Average August background particulate S loading over the South African high veld is $1.2 \mu\text{g m}^{-3}$ [Piketh *et al.*, 1999]. Assuming, in the absence of simultaneous measurements over Mount Kenya and South Africa, average conditions prevailed over South Africa during the August peak episode at Mount Kenya, a dilution by a factor of about 2 in sulfur loading by the time the air mass reached Mount Kenya is suggested. With both the transport from South Africa and northern Zambia the material reaching Mount Kenya at an altitude of 4420 m may have been transported thence in the haze layer normally capped by the ~ 500 -hPa absolutely stable layer that is such a persistent feature of the winter atmosphere over southern Africa [Cosijn and Tyson, 1996]. Transport from South Africa of the kind illustrated by the August episode is not frequent; it occurs on about 5% of days in a year [Freiman *et al.*, 1999].

The fine-fraction S transported to Kenya during the August episode was associated with Si and Fe (Figure 5). Fine-fraction Si is associated with aeolian dust derived from dry land surfaces [Maenhaut *et al.*, 1996]. Similarly, Fe is usually associated with dust from crustal material [Priest *et al.*, 1981; Maenhaut *et al.*, 1996]. Over South Africa, episodes of fine S and fine Fe observed at sites remote from emission sources are frequently coeval, having proximate sources in the industrial areas of northeastern South Africa [Piketh *et al.*, 1999]. The combination of fine S and Fe in the plume reaching Mount Kenya suggests the industrial origin of both constituents and argues against the fine-fraction S having

been acquired during transport over the ocean or from local sources in Kenya. After reaching Mount Kenya and stagnating over Kenya for a day or two during near-calm conditions, transport thereafter was to the Northern Hemisphere toward the South Asian subcontinent. In contrast, during the September episode, having reached Mount Kenya from northern Zambia, the transport plume from Mount Kenya bifurcated into two halves. One stream (49%) moved over Uganda, the Congo, and the Cameroon and into the Atlantic Ocean. The second (49%) recirculated to the south and east before exiting to the Indian Ocean north of Madagascar. Little interhemispheric exchange of aerosols occurred on this occasion.

The August high-concentration Cl episode (peak value of $0.3 \mu\text{g m}^{-3}$) was associated with coarse-fraction Cl and the direct transport of marine air from the central Indian Ocean (Figure 6a). In the same plume, fine-fraction Si was detected in significant quantities. Clearly, this could only have originated as recirculation of previously derived material from land areas. The finding that so-called clean air from the central Indian Ocean area may contain relatively high concentrations of silicon is significant. Transport of the Cl and Si plume from Mount Kenya contained an important interhemispheric component. The onward transport of marine chlorine was mainly to the Atlantic along the equatorial trough (70%) with a secondary pathway recirculating over Rwanda, Burundi, the Congo, and Tanzania back to the Indian Ocean.

Potassium is usually taken as a signature of biomass burning [Crutzen and Andreae, 1990]. From August 26–28, K concentrations recorded on Mount Kenya were higher than at other times. It would appear that the source of the K was burning in the horn of Africa region (Figure 6b). No evidence is at hand to suggest biomass-burning products reaching Kenya from the usual Angola, Zambia, Zimbabwe, Malawi, and Tanzania areas of the southern Africa region identified as the primary source of intensive fire activities from August to October [Cahoon *et al.*, 1992; Andreae, 1993]. On this occasion, interhemispheric transport from Mount Kenya was to the Indian Ocean.

Although our data were for a limited time period, we have shown transport corridors depicting the nature of interhemispheric transport of aerosols, which is quite consistent with the 5-year air

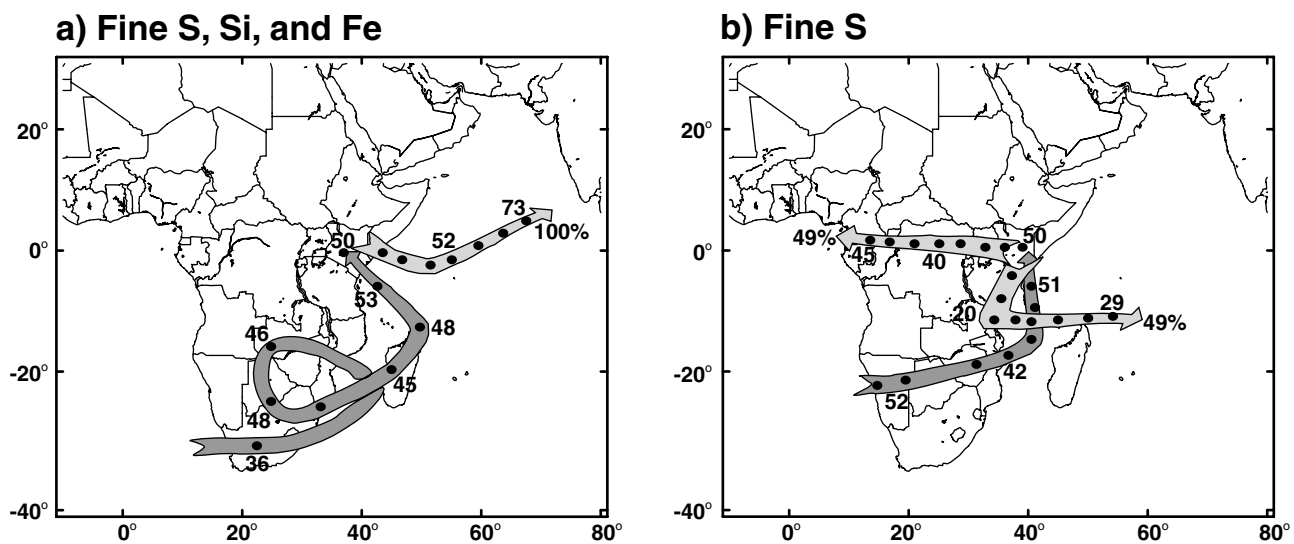


Figure 5. Transport to and from Mount Kenya, as defined by 500-hPa backward and forward trajectory pathways, of (a) fine-fraction S, Si, and Fe from southern Africa on August 23–25, 1997, and (b) fine-fraction S from southern Africa on September 3–9, 1997. Daily heights of the transport plume are for centroid trajectories within the trajectory cluster defining the plume and are given as tenths of hectopascals (i.e., 73 denotes 730 hPa). The dots represent daily intervals.

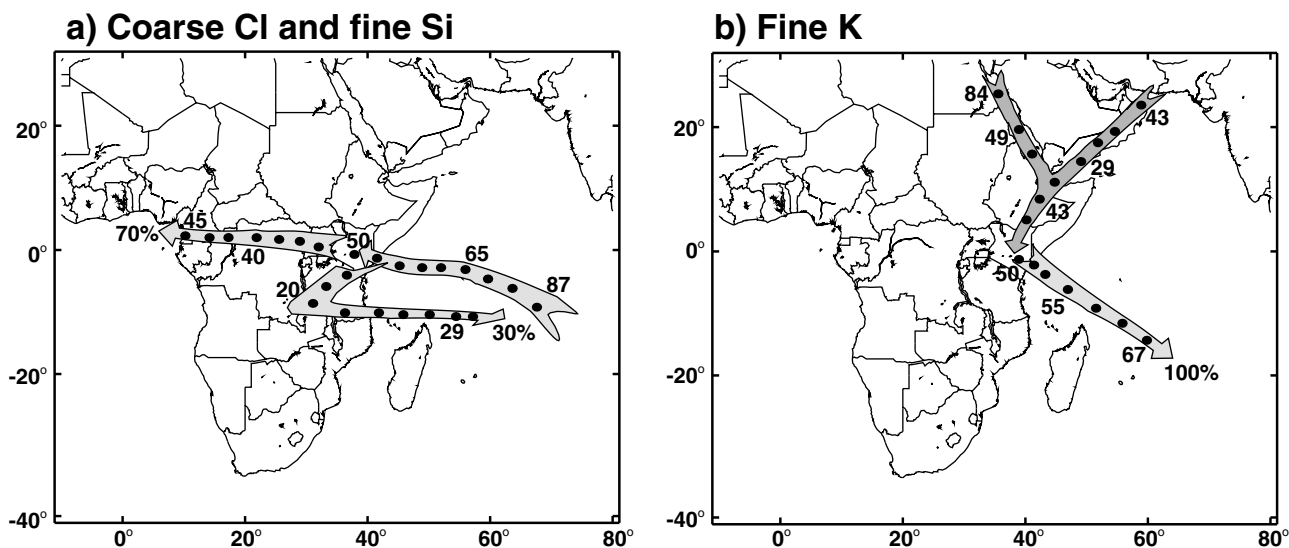


Figure 6. Transport to and from Mount Kenya, as defined by 500 hPa backward and forward trajectory pathways, of (a) coarse-fraction Cl and fine-fraction Si from the central Indian Ocean on August 19–22, 1997, and (b) fine-fraction K from, for example, Egypt and the Arabian Peninsula on August 26 to September 2, 1997. Daily heights of the transport plume are for centroid trajectories within the trajectory cluster defining the plume and are given as tenths of hectopascals (i.e., 67 denotes 670 hPa). The dots represent daily intervals.

transport climatology developed for the site [Gatebe *et al.*, 1999]. However, it does not appear possible to reach definitive conclusions.

4. Summary and Conclusions

Mount Kenya is an ideal high-altitude, remote, rural site for the monitoring of midtropospheric air quality in an equatorial region. Situated almost on the equator, observations taken at an altitude of 4420 m of aerosols transported over adjacent Africa and beyond reveal that in both the fine and coarse fractions, measured background concentrations of constituent elements are 3 factors lower than those reported in industrialized countries and elsewhere in Africa. However, they are 3 factors higher than concentrations reported from the South Pole. Most of the fine aerosols over Mount Kenya are enriched in heavy metals and halides with respect to average crustal rock. The enrichment represents transport to the mountain. This transport is both local and long range. Most of the former is associated with the coarse fraction of the constituent aerosols and nearby natural and anthropogenic sources. Local sources contribute most to midtropospheric aerosol during the day when convection and upslope winds on the surface of Mount Kenya are most active. At night the stable atmosphere and downslope winds minimize upward transport and allow long-range transport products to be measured more effectively. Consequently, long-range transports have only been considered for nocturnal data.

Trajectory analysis reveals that minimum S fluxes are observed at Mount Kenya in association with recirculation of sulfur-free air over equatorial and adjacent tropical Africa. Maximum sulfur concentrations are associated with transport of air passing over the industrial heartland of South Africa or over the Zambian/Congo copper belt where S emissions are known to be high. Transport from these regions in southern Africa to Kenya occurs along pathways ranging in length from 3500 to 6500 km and at the top of the haze layer capped by the ~500-hPa stable layer. Transport of fine S from South Africa is associated with anthropogenic Fe in addition to other usual dust components like Si and is of infrequent occurrence. Work done in South Africa suggests that on about 5%

of all days, transport of this kind occurs between South Africa and Kenya.

The transport of marine aerosols from the Indian Ocean to Mount Kenya, manifest during episodes of high concentration, coarse-fraction Cl, may be accompanied by simultaneous transport of fine-fraction Si back to land from mid-oceanic regions. Such a combination of aerosols can only result from large-scale recirculation of Si from Africa back to Africa, unless the Si originated from the Indian subcontinent. So-called clean air masses from the central Indian Ocean may contain relatively high concentrations of silicon. Despite observations being made during the period of maximum biomass burning to the south of Kenya, no K derived from burning in that region appears to have been transported to Mount Kenya. Instead, the little K observed at Mount Kenya in a single episode was transported there from the horn of Africa region.

Interhemispheric transport of aerosols has been observed to occur with both transport from southern Africa to the north and that from the Northern Hemisphere to the south. Much of the material reaching Mount Kenya is transported thence along the equatorial trough over adjacent countries toward the Atlantic Ocean. At the same time, recirculation of aerosols over central Africa and the Indian Ocean frequently occurs in transport en route to Kenya and as it leaves the country. All these transport patterns are modulated by seasonal changes in atmospheric circulation as the Sun moves north and south of the equator. Interhemispheric transport of aerosols across equatorial eastern Africa seems to be a feature of the climate of the region and cannot be ignored, particularly in the late austral winter.

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