SAFARI 2000 Physical and Chemical Properties of Aerosols, Dry Season 2000

Abstract

This data set contains particle size distributions and particle compositions for studying aerosol particles produced by savanna burning in southern Africa. We used analytical transmission electron microscopy (TEM), including energy-dispersive X-ray spectrometry (EDS) and electron energy-loss spectroscopy (EELS), to study aerosol particles from several smoke and haze samples and from a set of cloud samples. These aerosol particle samples were collected using the University of Washington Convair CV-580 research aircraft (Pósfai et al., 2003) during the dry season, August-September 2000.

Individual aerosol particles in smoke plumes from biomass fires and in regional hazes in southern Africa were studied using analytical transmission electron microscopy, which allowed detailed characterization of carbonaceous particle types in smoke and determination of changes in particle properties and concentrations during smoke aging. Based on composition, morphology, and microstructure, three distinct types of carbonaceous particles were present in the smoke: organic particles with inorganic (K-salt) inclusions; 'tar ball' particles; and soot. The relative number concentrations of organic particles were largest in young smoke, whereas tar balls were dominant in a slightly aged (~1 hour) smoke from a smoldering fire. Flaming fires emitted relatively more soot particles than smoldering fires, but soot was a minor constituent of all studied plumes. Further aging caused the accumulation of sulfate on organic and soot particles, as indicated by the large number of internally mixed organic/sulfate and soot/sulfate particles in the regional haze. Externally mixed ammonium sulfate particles dominated in the boundary layer hazes, whereas organic/sulfate particles were the most abundant type in the upper hazes. Apparently, elevated haze layers were more strongly affected by biomass smoke than those within the boundary layer. Based on size distributions and the observed patterns of internal mixing, we hypothesize that organic and soot particles are the cloud-nucleating constituents of biomass smoke aerosols. Sea-salt particles dominated in the samples taken in stratus clouds over the Atlantic Ocean, off the coast of Namibia, whereas a distinct haze layer above the clouds consisted of aged biomass smoke particles.

Background Information

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Data File Information

There are two types of particle data here: aerosol composition data and particle size distribution data.

Compositon Data Files

These data files contain the atomic compositions of individual aerosol particles, as generated from Energy-Dispersive X-ray Spectra (EDS). These include:

- X-ray counts and derived atomic percents for C, O, Si, S, Cl, and K for each analyzed particle in 8 samples (ASCII comma-delimted files)
- Triangular plots, containing relative concentrations of selected elements, as determined by EDS analyses (GIF images)

The file names are "**composition_XXXXX.csv**" where **XXXXX** is the sample number (the first 2 numbers are the flight number, see the **Data Samples Information Table** for details).

Column	Description
C	Atomic % Carbon
O	Atomic % Oxygen
Si	Atomic % Silicon
S	Atomic % Sulfur
Cl	Atomic % Chlorine
K	Atomic % Potassium

Size Distribution Data Files

These data files contain size distributions of particle types, as measured on Transmission Electron Microscope (TEM) images, sorted by sample. These include:

- Size distribution of particle types, as measured on TEM images (ASCII comma-delimted files). There are three different types of files (for the different particle types): organic; sulfate; and tarball.
- data plots (GIF images)

The file names are "**particle-type_XXXXX.csv**" where **particle-type** is "**organic**", "**sulfate**", or "**tarball**", and **XXXXX** is the sample number (the first 2 numbers are the flight number, see the **Data Samples Information Table** for details).

Column	Description
diameter (nm)	Diameter of the particle (nm)
particle number	The number of particles of this type of this size

TEM_images_particle_types.pdf

The file "TEM_images_particle_types.pdf" contains more information on aerosol particle types and pictures of those particles.

Data Collection Information

During the S2K Dry Season Campaign (Aug-Sep 2000), aerosol particles were collected from the University of Washington Convair CV-580 research aircraft (see picture at right), and the individual particles were analyzed using transmission electron microscopy (TEM). There are data for the following flights:

- Flight 1815, 17 Aug. 2000, Kruger National Park, South Africa
- Flight 1816, 18 Aug. 2000, Madikwe Game Reserve, South Africa/ Botswana
- Flight 1825, 31 Aug. 2000, West of Beira, Mozambique
- Flight 1826, 01 Sept. 2000, Kaoma, Zambia
- Flight 1837, 13 Sept. 2000, Walvis Bay, off Namibian coast

We examined smoke from both flaming and smoldering fires in order to assess the differences in the properties of individual particles from both types of emissions. Smoke samples immediately above biomass fires were studied from four research flights that sampled fire emissions in the Kruger National Park, South Africa; in the Madikwe Game Reserve, South Africa/Botswana; west of Beira, Mozambique; and near Kaoma, Zambia. Samples were also collected from smoke plumes 32 and 16 km downwind from the fire sources in the Madikwe Game Reserve and near Beira. Even though these "aged" smoke particles were still fairly young (~ 1 hr old), significant changes occurred in them compared to the particles in the smoke just above the fires.


The University of Washington CV-580 aircraft

Persistent, stratified haze layers that seriously degrade visibility are striking features during the dry season in southern Africa. Samples collected from layers of this regional haze provided an opportunity to study the particle types that occur in the hazes, and how their compositions and sizes compare with particles in young smoke. We analyzed haze samples from the four flights listed above, and from a sample collected above the Atlantic Ocean, off Walvis Bay, Namibia. In addition, haze aerosol particles associated with marine stratus clouds were analyzed; a list of samples is given in the table below.

Data Samples Information Table

UW Flight No.	Date (mmdd) (2000)	Location	Type of Sample	Time (UTC)	Sample #	Impactor Stage #	Type of Data Provided					σ (1/μ)	Comments
							EDS comp.	Organic SD	Tarball SD	Sulfate SD	Rel. conc.		
1815	0817	Kruger National Park, South Africa	haze	08:29-09:25	15-H2					x	x	1.88×10^{-5}	In boundary layer, at 4000 ft
					15-H3					x	x		
			smoke	10:25-10:34	15-SS2			x	x		x	1×10^{-4}	Young, smoldering and flaming fire
					15-SS3			x	x		x		
			smoke	11:28-11:31	15-SF2							2.3×10^{-3}	Young, flaming fire
					15-SF3		x	x	x		x		
1816	0818	Madikwe Game Reserve, South Africa/Botswana	haze	08:56-09:14	16-H1	1						2×10^{-5}	Background haze, upwind of prescribed fire, at 6,000 ft
					16-H2	2							
					16-H3	3	x						
			smoke	10:19-10:23 and 11:03-11:10	16-S3	3	x						20 miles downwind of prescribed fire
1825	0831	West of Beria, Mozambique	haze	09:42-10:15	25-H1	1			x			5.56x 10^{-5}	In boundary layer, en route to Mozambique
					25-H2	2			x				

					25-H3	3							coast, at 9,000 ft
			smoke	11:40-11:47	25-SY2	2	x	x			x	4.2x 10^{-4}	Young smoke of smoldering fire
					25-SY3	3	x	x			x		
			smoke	12:26-12:40	25-SA2	2	x	x	x		x	1.7x 10^{-4}	10 miles downwind of the same smoldering fire
					25-SA3	3	x	x	x		x		
1826	0901	Kaoma, Zambia	haze	06:29-07:35	26-H3	3	x					2-5x 10^{-5}	Upper haze layer at 12,000 ft, en route to Zambia
			smoke	09:25-09:33	26-S3	3	x						Young smoke
1837	0913	Walvis Bay, off Namib-ian coast	cloud	09:05-10:44	37-C2	2	x						Below, in, and above marine stratus clouds
					37-C3	3	x						
			haze	11:32-12:12	37-H3	3	x						Main, upper haze layer, at 13,000 ft

Aerosol Particle Types

Biomass burning produces mainly carbonaceous particles. Since both elemental and organic carbon (C) occur in biomass smoke, the broad group of carbonaceous particles can include different particle types that have highly variable atmospheric effects. Whereas elemental C is thought to only occur in soot (or black carbon, BC), organic compounds may occur in any smoke particles, including soot. Single-particle studies have been scarce on biomass smoke particles, and so little information is available on the individual-particle level or on carbonaceous aerosols in general. Since carbonaceous particles are the least accessible among the major types of tropospheric aerosol particles for single-particle electron-beam methods, the lack of data is understandable.

The mass of BC ranges from 10% to 30% of the total aerosol in particulate emissions from savanna fires. Apart from C, important quantities of potassium (K), sulfur (S), chlorine (Cl), and silicon (Si) are emitted from biomass burning. Although emission factors have been measured for several types of vegetation, details of the distributions of these elements on individual particles is not known.

Analytical TEM is a powerful tool for characterizing individual aerosol particles because it provides simultaneous morphological, compositional, and structural information. However, it has limitations. First, TEM methods are labor intensive and require the continuous presence of the operator; therefore, the number of analyzed particles is relatively small and statistics are poor. Second, in the vacuum of the TEM and under the electron beam some compounds may be vaporized. We performed experiments with volatile organic acids ($C_6H_{10}O_4$ and $C_7H_6O_4$) and found that such particles immediately sublimate when the electron beam hits them, even when a very low-intensity beam is used. Thus, only the more refractory organic substances will remain and be observable in the TEM samples. Nevertheless, we found that analytical TEM is still extremely useful for characterizing biomass smoke and haze particles and is a unique source of information on particle coatings, agglomeration, and possible atmospheric reactions.

The strategy used in this study was to identify major particle groups based on morphology, composition, and behavior

in the electron beam. We term the three most important particle types in biomass smoke plumes as: (1) 'organic particles with inorganic inclusions'; (2) 'tar balls' (carbon-rich, spherical particles); and (3) 'soot'. In the haze samples, the most abundant particles were ammonium sulfates, either externally or internally mixed with carbonaceous particles. Sea-salt particles were present in the samples collected near or over the ocean, and characteristic biogenic particles occurred in most haze samples.

For more information on aerosol particle types and pictures of those particles, see the file "TEM_images_particle_types.pdf".

Particle Type	Morphology	Composition	Structure	Mixing
Organic particle with inorganic inclusion	Irregular, particles are spread on the lacey support film	Carbonaceous with inorganic K-salt inclusions	Amorphous	Externally mixed in young smoke, internally mixed with sulfate and soot in the haze
Tar ball	Spherical	Carbonaceous with minor O, in cases with minor S, K, Si, Cl	Amorphous	Externally mixed
Soot	Agglomerates of 20 to 50-nm spherules	Carbonaceous with minor O, in cases with minor S, K, Si	Turbostratic graphitic layers form the individual onion-like spherules	Externally mixed in young smoke, internally mixed with sulfate in haze
Sulfate	Spherical to irregular, decomposes in the electron beam	Ammonium sulfate	Crystalline in the vacuum of the TEM	Both externally and internally mixed with organic and soot particles in the haze

Experimental Procedure

We used a three-stage impactor (model MPS-3, California Measurements, Inc.) with nominal cuts at 2 and 0.3 µm (first stage d > 2, second stage 2 > d > 0.3, and third stage d < 0.3 µm); we studied mainly the second- and third-stage samples. In some sample sets, there were only minor differences in the size distributions and compositions between particles from the two lower stages; in such cases data from the two stages were combined. Particles were collected on copper TEM grids. For most samples, we used TEM grids that were covered with a lacey Formvar supporting film. The particles attach to this film, and many extend over the holes of the lace, which is excellent for EDS or EELS analyses because spectra can be obtained with little or no background signal. Low backgrounds are especially important in analyzing carbon and oxygen on a supporting film that also contains these elements. During the flight to Namibia (UW flight number 1837), we used TEM grids covered with continuous Formvar films. The samples were stored in grid boxes in a desiccator and part of the time in air until studied in the TEM.

TEM was performed using mainly a Philips CM20 TEM operated at 200 kV accelerating voltage and equipped with an ultrathin-window Noran Voyager energy-dispersive X-ray detector. Electron energy-loss images were obtained using a 125 kV Hitachi 7100 TEM with an attached Gatan Imaging Filter that acts as both a spectrometer and an imaging device. Additionally, a 200 kV JEOL 2000FX microscope was used for imaging and EDS analyses. Images of the particles were obtained at magnifications ranging from 5000 to 500,000.

The sizes of particles were measured on digitized electron micrographs by fitting ellipses to the particle outlines and taking the average of the short and long axes of the best fitting ellipse as the particle diameter. Particle number size distributions were derived for tar balls, sulfates, and organic particles. The shapes of soot particles are highly irregular and therefore the size distributions of soot were not determined. The upper limits of the resulting size distributions may be affected by the inhomogeneous distribution of particles on the grids: larger particles pile up in the center, where particle overlap makes size measurements impossible.

Relative Concentrations of Particle Types

Almost every aerosol particle collected from the biomass smoke plumes was carbonaceous, that is, their major element was C. Most particles in the haze samples also contained C since even the ammonium sulfate particles were

coated with carbonaceous films. The relative concentrations of other elements showed greater variability than C, so we used the element compositions (particularly the concentrations of K, S, Cl, and Si), the typical morphologies, and the behavior of particles in the electron beam to distinguish particle types. In the following we will describe the main particle groups, then discuss the changes in relative particle number concentrations and compositions in various types of samples.

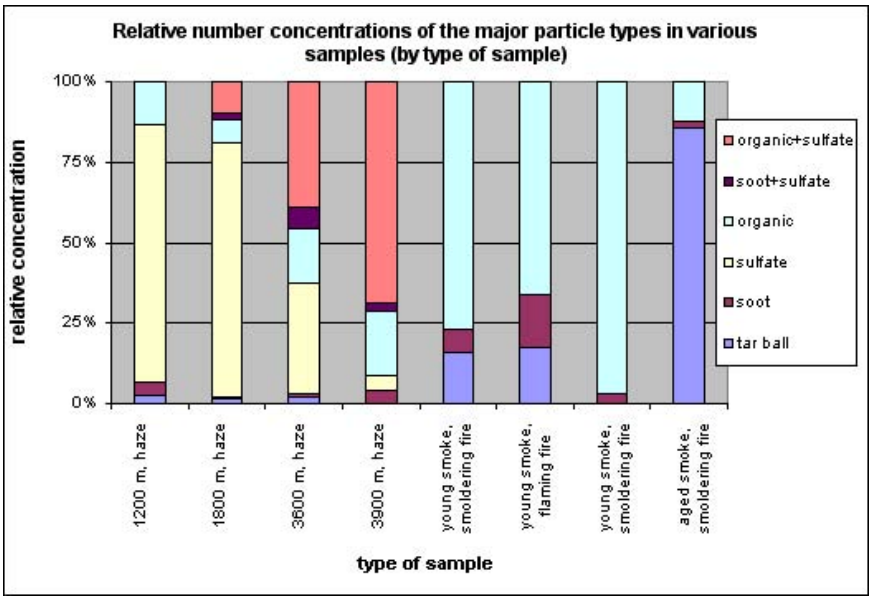
The following table contains some of the results of the data analysis, i.e., the relative concentrations of the different particle types within the different aerosol samples taken.

Data Samples Information Table: Relative Concentrations of Particle Types

Flight	Sample	tar ball	soot	sulfate	organic	
1815	15-H2	5	5	110	18	
	15-H3	2	7	126	21	
	sum	**7**	**12**	**236**	**39**	**294**
1815	15-SS2	50	21	0	124	
	15-SS3	22	13	0	230	
	sum	**72**	**34**	**0**	**354**	**460**
1815	15-SF3	44	41	0	165	
	sum	**44**	**41**	**0**	**165**	**250**
1825	25-SY2	0	5	0	112	
	25-SY3	0	2	0	96	
	sum	**0**	**7**	**0**	**208**	**215**
1825	25-SA2	169	2	0	24	
	25-SA3	179	7	0	25	
	sum	**348**	**9**	**0**	**49**	**406**

Flight	Sample	tar ball	soot	sulfate	organic	soot + sulfate	organic + sulfate	
1816	16-H3	3	2	184	16	5	23	
	sum	**3**	**2**	**184**	**16**	**5**	**23**	**233**
1826	26-H3	4	3	73	37	14	84	
	sum	**4**	**3**	**73**	**37**	**14**	**84**	**215**
1837	37-H3	0	8	9	40	5	135	
	sum	**0**	**8**	**9**	**40**	**5**	**135**	**197**

Sample Type	Sample	tar ball	soot	sulfate	organic	soot + sulfate	organic + sulfate	
1200 m, haze	15-H2,3	7	12	236	39	0	0	
1800 m, haze	16-H3	3	2	184	16	5	23	
3600 m, haze	26-H3	4	3	73	37	14	84	
3900 m, haze	37-H3	0	8	9	40	5	135	
young smoke, smoldering fire	15-SS2,3	72	34	0	354	0	0	
young smoke, flaming fire	15-SF3	44	41	0	165	0	0	
young smoke, smoldering fire	25-SY2,3	0	7	0	208	0	0	
aged smoke, smoldering fire	25-SA2,3	348	9	0	49	0	0	
	sum	**478**	**116**	**502**	**908**	**24**	**242**	**2270**



Additional Sources of Information

For more information on aerosol particle types, and pictures of those particles, see the file "TEM_images_particle_types.pdf".

References

Pósfai, M., R. Simonics, J. Li, P. V. Hobbs, and P. R. Buseck. 2003. Individual aerosol particles from biomass burning in southern Africa: 1. Compositions and size distributions of carbonaceous particles, J. Geophys. Res., 108(D13), 8483, doi:10.1029/2002JD002291.

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