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Contributions of biomass/biofuel burning to organic aerosols and particulate matter in Tanzania, East Africa, based on analyses of ionic species, organic and elemental carbon, levoglucosan and mannosan

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Abstract. Atmospheric aerosol samples of PM_{2.5} and PM₁₀ were collected at a rural site in Tanzania, East Africa, in 2011 during wet and dry seasons and were analysed for carbonaceous components, levoglucosan, mannosan and watersoluble inorganic ions. The contributions of biomass/biofuel burning to the organic carbon (OC) and particulate matter (PM) mass were estimated to be 46-52% and 8-13%, respectively. The mean mass concentrations of PM2.5 and PM_{10} were $28\pm6\,\mu g\,m^{-3}$ and $47\pm8\,\mu g\,m^{-3}$ in wet season, and $39 \pm 10 \,\mu\text{g}\,\text{m}^{-3}$ and $61 \pm 19 \,\mu\text{g}\,\text{m}^{-3}$ in dry season, respectively. Total carbon (TC) accounted for 16-19% of the $PM_{2.5}$ mass and 13–15% of the PM_{10} mass. On average, 86 to 89% of TC in PM2.5 and 87 to 90% of TC in PM_{10} was OC, of which 67–72% and 63% was found to be water-soluble organic carbon (WSOC) in PM2 5 and PM₁₀, respectively. We found that concentrations of levoglucosan and mannosan (specific organic tracers of pyrolysis of cellulose) well correlated with non-sea-salt potassium (nss-K⁺) ($r^2 = 0.56-0.75$), OC ($r^2 = 0.75-0.96$) and WSOC $(r^2 = 0.52-0.78)$. The K⁺/OC ratios varied from 0.06 to 0.36 in $PM_{2.5}$ and from 0.03 to 0.36 in PM_{10} with slightly higher ratios in dry season. Mean percent ratios of levoglucosan and mannosan to OC were found to be 3-4 % for PM2.5 and PM_{10} in both seasons. We found lower levoglucosan / K⁺ ratios and higher K⁺/EC (elemental carbon) ratios in the biomass-burning aerosols from Tanzania than those reported from other regions. This feature is consistent with the high levels of potassium reported in the soils of Morogoro, Tanzania, suggesting an importance of direct emission of potassium by soil resuspension although K^+ is present mostly in fine particles. It is also likely that biomass burning of vegetation of Tanzania emits high levels of potassium that may be enriched in plant tissues. The present study demonstrates that emissions from mixed biomass- and biofuel-burning activities largely influence the air quality in Tanzania.

1 Introduction

Africa is facing air pollution problems with particulate matter (PM) due to its growing economy. Atmospheric aerosols that are derived from both natural and anthropogenic sources have been reported around the world (Medeiros et al., 2006; Nyanganyura et al., 2007; Harrison and Yin, 2008; Engling et al., 2011; Pavuluri et al., 2011). Biomass burning which is associated with forest, grassland, domestic and open fires has drawn global concern for its effects on human health, visibility and global climate. In areas with intensive land use changes or agricultural practices, field burning of biomass and agricultural wastes is also an important source of aerosol particles and trace gases in the atmosphere (Andreae and Merlet, 2001; Koe et al., 2001; Streets et al., 2003). The major types of biomass burning in Africa include forest and savanna fires (Cachier et al., 1991, 1995; Lacaux et al., 1993; Liu et al., 2000). However, agricultural field burning is another type of biomass burning which may be a regionally important source of atmospheric aerosols during the dry season (Lacaux et al., 1993).

Carbonaceous materials, the major components of atmospheric aerosols, consist of elemental carbon and a complex mixture of numerous organic compounds, which can account for 20 to 50% of PM2.5 mass in urban and rural locations (Querol et al., 2009). On the other hand, a large portion of water-soluble organic species have been well recognized to be emitted from biofuel and/or biomass burning (Mayol-Bracero et al., 2002). Levoglucosan (1,6-Anhydro- β -D-glucopyranose), water-soluble potassium and even organic carbon are known to be the typical components of biomass-burning aerosol (Cachier et al., 1995; Simoneit et al., 1999). Potassium has been used as a conventional biomass-burning tracer (Cachier et al., 1991), but its contribution from sea-salt aerosol may be an important source (Riley and Chester, 1971), whereas levoglucosan, which is produced by the pyrolysis of cellulose, has been used as a unique molecular tracer of biomass-burning aerosols (Simoneit et al., 1999: Fraser and Lakshmanan, 2000: Puxbaum et al., 2007). In addition, the ratio of levoglucosan to mannosan has been proposed as an indicator of different biomass fuel types (Schmidl et al., 2008; Engling et al., 2009).

Tanzania as a whole does not suffer from serious air pollution problems yet, but localized pollution does occur due to its growing population, and agricultural and industrial activities. Unfortunately, compared with the chemical characterization performed for atmospheric aerosols in other countries around the world, such studies in Africa and in particular Tanzania have not been conducted for organic aerosols (Cachier et al., 1991; Maenhaut et al., 1996; Gao et al., 2003). In addition, biomass- and biofuel-burning activities in Tanzania are common phenomena. However, the studies of atmospheric aerosols have not been reported in relation to specific tracers such as levoglucosan (Simoneit et al., 1999). Therefore, the objective of this study is to investigate the contribution of biomass- and biofuel-burning activities to the organic carbon (OC) and PM mass at a rural background site in Morogoro, Tanzania. Here, we discuss the seasonal variations in the chemical characteristics of carbonaceous components, levoglucosan and mannosan in PM2.5 and PM10 and estimate the contributions of biomass/biofuel burning to organic aerosols and PM mass in rural Tanzania. The study on levoglucosan and mannosan in aerosols has been conducted here for the first time in Tanzania.



Fig. 1. Map showing the location of the sampling site of Morogoro, Tanzania.

2 Experimental

2.1 Sampling site and aerosol collection

Intensive aerosol sampling campaigns were carried out at a rural background site (300 000 inhabitants) in Morogoro $(06^{\circ}47'40''8 \text{ S}, 37^{\circ}37'44''5 \text{ E}; altitude 504 \text{ m a.s.l})$ during the wet (30 May to 13 June 2011) and dry seasons (28 July to 8 August 2011). Morogoro is located at about 200 km west of the coast of the western Indian Ocean where the city of Dar es Salaam, a business capital of Tanzania, is located (Fig. 1). The major industry of Morogoro is agriculture without any modern industries. Possible main sources of local aerosols include field burning of crop residue and waste, emissions from livestock (dairy cattle or farm) and domestic/forest fires. Aerosol samples were collected at 2.7 m above ground level using PM2.5 and PM10 low volume samplers (Gent type, flow rate $17 L \min^{-1}$) in parallel (Maenhaut et al., 1994; Hopke et al., 1997). The two samplers were equipped with quartz fibre filters (Pallflex 2500QAT-UP, 47 mm), which were pre-baked at 450 °C for 4 h in a furnace to eliminate adsorbed organics before use. The samplers were set up at a meteorological synoptic station located within the Solomon Mahlangu Campus of Sokoine University of Agriculture (SUA). The site and its large radius were covered by grass (vegetation) that may reduce immediate emission of local soil dust to interfere with the sampling.

The sample collection was conducted in approximately 24 h intervals, and the filters were exchanged at 7:30 a.m. LT A total of 21 sets of samples and 2 field blanks (air-exposure time of 30 s) were collected for each sampler and used in this study. Before sampling, the filters were placed in a preheated glass jar with a Teflon-lined screw cap. After sampling, the filter samples were folded in half face to face in a clean glass vial with a Teflon-lined screw cap and stored in a freezer at -20 °C, then transported to the atmospheric chemistry laboratory at the Institute of Low Temperature Sciences (ILTS), Hokkaido University (Japan), and kept frozen at -20 °C prior to analysis. All the procedures were strictly quality-controlled to avoid any possible contamination of the samples.

2.2.1 PM mass measurement

For the determination of particulate mass, filters were weighed before and after sampling with a high-sensitivity analytical microbalance (Sartorius Model M2P, detection limit 1 μ g). Before weighing, the filters were conditioned at room temperature (20 °C) and relative humidity of 50 % for 24 h.

2.2.2 Carbonaceous aerosol components

The concentrations of organic carbon and elemental carbon (EC) were determined with a field-type carbon analyser (Sunset Laboratory Inc., USA) using thermal/optical transmission method (Birch and Cary, 1996) following the Interagency Monitoring Protected Visual Environments (IM-PROVE) thermal evolution protocol. A 1.54 cm² portion of each quartz fibre filter was placed in a quartz tube inside the thermal desorption chamber of the analyser and heated stepwise as described in detail by Wang et al. (2005). Briefly, helium (He) gas is applied in the first ramp and is switched to helium/oxygen mixture in the second ramp. A non-dispersive infrared detector measured the evolved carbon dioxide at each temperature step, and the transmittance of light (red 660 nm) through the filter punch was used for determining the OC/EC split. The "artefact-free" particulate OC data were obtained in a tandem filter set-up as the difference between the OC on the front filter and that on the back filter (Viana et al., 2006). It is known that both positive (i.e. adsorption of volatile organic carbon that is assumed to prevail) and negative artefacts may occur for particulate OC when collecting aerosol samples with quartz fibre filters (Mader et al., 2003).

To determine water-soluble organic carbon (WSOC), a punch of 2.59 cm² from each quartz fibre filter was extracted with 15 mL organic-free ultrapure water in a 50 mL glass vial under ultrasonication for 15 min. The extracts were then filtrated using a syringe filter (Millex-GV, 0.22 μ m) and analysed for WSOC using a total organic carbon (TOC) analyser (Model TOC-Vcsh, Shimadzu, Kyoto, Japan) (Miyazaki et al., 2011). Before the measurement of WSOC, the extract was acidified with 1.2 M HCl and purged with pure air in order to remove dissolved inorganic carbon and volatile organics. The triplicate analyses of laboratory standards showed that the analytical uncertainties were within 5 %.

All carbonaceous components in the reported data for the front filters have been corrected using the blank filters. The mean back to front filter ratio for OC was less than 10% of that on the front filter during the campaigns. EC was not detected on the back filter, whereas WSOC was negligible. The detection limits calculated as three times the standard deviation (3σ) of OC and total carbon (TC) values in the field blanks were 0.12–0.45 µgC m⁻³ for OC and TC, and 0.02–0.14 µgC m⁻³ for WSOC.

2.2.3 Levoglucosan and mannosan

For the determination of levoglucosan and mannosan (specific biomass-burning tracers derived from the pyrolysis of cellulose and hemicellulose; Simoneit et al., 1999), the filter samples were analysed by the method described in Fu and Kawamura (2011). Briefly, a portion of each filter (1.54 cm^2) was extracted with dichloromethane/methanol (2:1; v/v)under ultrasonication. The extracts were concentrated using a rotary evaporator under a vacuum, and then sugar compounds were derived to TMS ethers with 50 µL of N,O-bis-(trimethylsilyl)trifluoroacetamide (BSTFA) containing 1% trimethylsilyl chloride and 10 µL of pyridine at 70 °C for 3 h. The derivatives were diluted by addition of $140 \,\mu\text{L}$ of *n*hexane containing 1.43 ng μ L⁻¹ of the internal standard (C₁₃ *n*-alkane) and then determined with a gas chromatographmass spectrometer (GC/MS). The detection limit of levoglucosan and mannosan was 0.06 and 0.01 ng m⁻³, respectively. Levoglucosan and mannosan (as TMS derivatives) were identified by comparison of mass spectra with that of the authentic standard. Fragment ions of m/z 217 and 204 were used for their quantification using calibration curves by analysing aliquots of a stock standard solution of levoglucosan and mannosan. The recoveries of levoglucosan and mannosan were 85% and 91%, respectively. Detailed analytical procedures can be found elsewhere (Fu et al., 2008; Mochida et al., 2010).

2.2.4 Water-soluble inorganic ions

An ion chromatograph (761 Compact IC, Metrohm, Switzerland) was used for the determination of the major watersoluble anions $(SO_4^{2-}, NO_3^{-}, Cl^- \text{ and } CH_3SO_3^-)$ and cations $(NH_4^+, Na^+, K^+, Mg^{2+} \text{ and } Ca^{2+})$. A 2.59 cm² punch of each quartz fibre filter was extracted twice with 5 mL ultrapure water (resistivity of $> 18.2 \text{ M}\Omega \text{cm}$, Sartorius arium 611 UV) under ultrasonication for 15 min. The extracts were filtered through a syringe filter (Millex-GV, 0.22 µm pore size, Millipore) and injected to IC for the determination of inorganic ions. Anions were measured using a SI-90 4E Shodex column (Showa Denko, Tokyo, Japan) and an eluent of 1.8 mM Na₂CO₃ plus 1.7 mM NaHCO₃. For cation analysis, we used a Metrosep C2-150 (Metrohm) column with 4 mM tartaric acid + 1 mM dipicolinic acid as the eluent. The injection loop volume was 200 µL for both cation and anion measurements. All ions were quantified against a standard calibration curve. The analytical errors in duplicate analysis of the authentic standards were within 7 %. Detection limit ranged from 0.006 to $0.02 \text{ ng } \text{L}^{-1}$ for anions and 0.01 to $0.02 \text{ ng } \text{L}^{-1}$ for cations. The reported concentrations for the PM mass, carbonaceous and ionic species are all corrected for field blanks. The non-sea-salt (nss)- SO_4^{2-} and nss-K⁺ were calculated using Na⁺ as a reference tracer for sea salt.

2.3 Meteorology

The climate of Tanzania is characterized by a warm and wet season (March-May) and a dry and somewhat cool/dry season (July-September). The two seasons are distinguished by the passage of the inter-tropical convergence zone, with the northeasterly (NE) East African monsoon between November and March, and the southeasterly (SE) East African monsoon between April and October (Trewartha, 1961; Bargman, 1970). Morogoro, where the sampling site was located, experiences a humid, tropical, savanna climate with an annual average rainfall of 870 mm (range: 610-1180 mm) (TMA, 2011). During the two-week wet season campaign, there were only 4 days without rain. However, few aerosol samples were met with a very weak rain. In contrast, there were 2 rainy days out of 14 during the dry season. The ambient temperature varied from 22.9 to 29.1 °C (average 26 °C) in wet season campaign and from 21.1 to 28.2 °C (average 24.6 °C) in dry season campaign. The daily average relative humidity ranged from 65 to 96 % (av. 81 ± 0.7 %) in the morning hours and from 41 to 60% (av. $50 \pm 0.6\%$) in the afternoon. During the entire sampling period (May through August), the equatorial belt on the western coast of the Indian Ocean experienced mainly one type of wind pattern southeasterly monsoons.

3 Results and discussion

3.1 PM mass and inorganic ion concentrations

Table 1 presents concentrations of water-soluble inorganic ions and PM2.5 and PM10 masses in aerosols collected during the 2011 campaigns in the wet and dry seasons at the rural site in Morogoro. Concentrations of inorganic ions in PM_{2.5} and PM₁₀ are comparable between two seasons although higher concentrations were observed in dry season. During both seasons K⁺ was the most abundant cation in $PM_{2.5}$. Na⁺ and Ca²⁺ were dominant ions in PM_{10} , whereas SO_4^{2-} was mainly associated with PM₁₀. The mean concentrations and associated standard deviation of the PM2.5 mass were $28 \pm 6 \,\mu g \, m^{-3}$ in wet season and $39 \pm 10 \,\mu g \, m^{-3}$ in dry season. Those of the PM_{10} mass were $47\pm8\,\mu g\,m^{-3}$ in wet season and $61 \pm 19 \,\mu g \, m^{-3}$ in dry season. The mean concentrations of PM2.5 mass and PM10 mass were higher in the dry than the wet season. The seasonal variations of the aerosol levels could be attributed to changes in source strengths and seasonal meteorology such as precipitation and mixing height. However, there were small differences in meteorological conditions especially in the temperature during the sampling days. In wet season, wet scavenging by rainfall could be efficient in removing the PM. During the dry season, unfavourable meteorological conditions (e.g. no rain and frequent inversion) and the enhanced emission from biomass



Fig. 2. Mean concentration ratios (%) of selected aerosol components or species in $PM_{2.5}$ to those in PM_{10} during the 2011 wet and dry season campaigns in Morogoro.

burning (i.e. primary sources) could be responsible for the higher concentrations of PM.

The mean mass concentration ratios of PM2.5 / PM10 were calculated on the basis of the data for PM2.5 and PM10 samples taken in parallel and then averaged over all samples from the campaign to be $60 \pm 17\%$ (range: 39–93%) in wet season and $67 \pm 20\%$ (range: 41% to 84%) in dry season (Fig. 2). The results indicate that most of the PM_{10} mass was present in the fine size fraction (PM_{2.5}) in both seasons. This may be due to a larger contribution from pyrogenic aerosols and photochemically produced organic aerosols, which are most likely enriched in fine $(PM_{2.5})$ particles. In contrast, the contribution from soil dust may be small, which is known to be mostly associated with PM₁₀ particles. However, our present $PM_{2.5} / PM_{10}$ ratios are substantially higher than the values obtained in a previous study (Mkoma et al., 2010a, b) and similar to those reported in rural and urban sites around the world (Gu et al., 2010; Ram and Sarin, 2011).

In previous studies conducted in 2005 during the wet and dry seasons and 2006 during the wet season at the main campus of Sokoine University of Agriculture in Morogoro (about 10 km from the current sampling site), mean PM_{10} mass concentrations were obtained to be $28 \pm 13 \,\mu g \, m^{-3}$, $49 \pm 12 \,\mu\text{g}\,\text{m}^{-3}$ and $12 \pm 13 \,\mu\text{g}\,\text{m}^{-3}$, respectively (Mkoma et al., 2009a, b; Mkoma et al., 2010a). These values are lower than those for the PM₁₀ mass concentrations obtained in the current study (Table 1) possibly due to variations in the emission from biomass burning and their source strengths as well as meteorological parameters such as precipitation. Compared to the ambient air quality standards set by the Tanzania Bureau of Standards (TBS) for PM_{10} (60 to 90 µg m⁻³), none of the PM₁₀ samples exceeded the TBS limit during the sampling periods (TBS, 2006). Currently, there is no ambient PM2.5 standard in Tanzania. The European Union (EU) daily limit concentration for PM_{10} mass is 50 µg m⁻³, which cannot be exceeded more than 35 days per year, and the EU yearly average limit concentration is $40 \,\mu g \,m^{-3}$ (EU Directive 1999/30/EC, 1999). Nearly two thirds of our PM₁₀ mass data were higher than the EU daily limit, and the average concentration for the entire sampling period is larger than the **Table 1.** Mean concentrations and concentration ranges of the PM mass, carbonaceous components, levoglucosan, mannosan and inorganic ions in $PM_{2.5}$ and PM_{10} in Morogoro during the wet and dry seasons, 2011.

Component/species		PM ₂		PM ₁₀				
	Wet season $(n = 11)$		Dry season $(n = 10)$		Wet season $(n = 11)$		Dry season $(n = 10)$	
	Range	Mean \pm SD	Range	Mean \pm SD	Range	Mean \pm SD	Range	Mean \pm SD
PM (μ g m ⁻³) and carbonaceous components (μ gC m ⁻³)								
PM	17–38	28 ± 6	30–61	39 ± 10	36–57	47 ± 8	37–99	61 ± 19
TC	2.6–7	4 ± 1.4	4.6–10	7 ± 2	3–10	6 ± 2	4–16	9 ± 4
OC	2-6	3.9 ± 1.3	3.4–8	6 ± 2	3-8	5.6 ± 1	4-14	8 ± 3
EC	0.2 - 1.1	0.5 ± 0.3	0.5–1.4	1 ± 0.3	0.2–2	0.7 ± 0.4	0.2–4	1.3 ± 1
WSOC	1.4–5	3 ± 1	2.5–5	3.9 ± 0.9	2-4	3.5 ± 0.6	2.8–7	5 ± 1
Levoglucosan (ng m $^{-3}$)	38-308	146 ± 85	138–408	253 ± 77	49–377	209 ± 95	99–659	308 ± 154
Mannosan (ng m ⁻³)	3–25	13 ± 7	12-35	24 ± 7	5–34	20 ± 9	10–59	30 ± 15
Inorganic ions $(ng m^{-3})$								
$\rm NH_4^+$	41-458	210 ± 122	325–2344	928 ± 669	76–628	207 ± 180	97–1400	654 ± 432
NO_3^-	43-136	64 ± 27	114–264	180 ± 55	93–248	172 ± 56	172–774	440 ± 209
SO_4^{2-}	72–376	214 ± 75	59.9–560	261 ± 189	183–511	294 ± 92	321-1135	585 ± 280
$nss-SO_4^{2-}$	29-328	133 ± 97	139–439	244 ± 125	16–324	74 ± 102	85-409	280 ± 139
Cl ⁻	39–99	62 ± 22	35-175	80 ± 50	73–602	183 ± 148	47–656	328 ± 200
MSA ⁻	8-35	16 ± 7	11–26	18 ± 4	8-21	13 ± 4	13–50	28 ± 12
Na ⁺	126-568	320 ± 117	378–937	621 ± 154	529-2913	1145 ± 681	386–4349	2243 ± 1279
Mg ²⁺	36-263	94 ± 86	12-146	79 ± 61	65–580	239 ± 150	59–985	460 ± 334
K ⁺	167–741	393 ± 169	715–2710	1538 ± 702	170–1683	632 ± 406	922–3788	1909 ± 1089
nss-K ⁺	156–734	382 ± 170	691–2688	1516 ± 703	149–1657	591 ± 408	851-3669	1829 ± 1053
Ca ²⁺	39–99	62 ± 22	111-608	297 ± 153	594–1948	1272 ± 416	532-3529	1702 ± 1074

EU yearly limit. Nevertheless, the PM mass data from this rural site in Tanzania are comparable to the few available data sets from rural southern Africa (Nyanganyura et al., 2007). Our concentrations of PM_{10} (Table 1) are also comparable to or are lower than those reported from other sites in Europe and Asia (Van Dingenen et al., 2004; Gu et al., 2010; Maenhaut et al., 2011; Ram and Sarin, 2011).

3.2 Carbonaceous components

3.2.1 Total carbon, organic carbon and elemental carbon

As seen from Table 1, the higher mean concentrations of TC, OC and EC were obtained in the dry season, whereas lower mean concentrations were observed in the wet season. This seasonal trend is consistent with that of the PM mass, which can be explained by the variations in source strengths and partly by meteorological conditions. The mean mass ratios showed that TC accounted for 16% and 19% of the PM_{2.5} mass in wet and dry seasons, whereas TC accounted for 13% and 15% of the PM₁₀ mass, respectively. Among the carbonaceous components, OC was the most abundant, accounting for 89% of TC in PM_{2.5} in wet season and 86% in dry season. Abundances of OC in PM₁₀ mass were 12% in wet season and 13% in dry season. The EC/TC ratios for wet and dry seasons were 0.11 \pm 0.07 and 0.14 \pm 0.05 for PM_{2.5} and 0.10 \pm 0.03 and 0.13 \pm 0.05 for

 PM_{10} , respectively. These EC/TC ratios could be representative of a large set of emission measurements from diverse types of biomass burning, including biofuel use (Andreae and Merlet, 2001). However, our ratios are generally lower than the EC/TC ratios (0.13–0.24) reported from the sites impacted by biomass-burning activities (Ram and Sarin, 2010; Engling et al., 2011).

Figure 3 shows the temporal variation of OC and EC concentrations and OC/EC ratios for $PM_{2.5}$ and PM_{10} aerosols. The OC and EC concentrations exhibited similar trends with small fluctuations in both sizes during the sampling period. The average OC/EC ratio for PM2.5 and PM10 was 10 during wet season, whereas it became lower (7 and 8, respectively) during the dry season. Higher values of OC were observed on 29 July through 2 August 2011 possibly due to the enhanced source strength. Low OC/EC ratios observed in this study might have been caused by an increased emission of EC compared to OC and decreased precipitation especially in the dry season. The somewhat high EC concentrations in dry season are likely due to the contributions from the charcoal-making process and domestic use as well as burning of agricultural wastes. However, we presume that different biomass-burning activities could have resulted into a mixture of aerosols at our sampling site.

The relationship between OC and EC and the mass ratio of OC to EC have been used to identify origins, emission and transformation characteristics of carbonaceous aerosols (Turpin and Huntzicker, 1991). As shown in Fig. 4a, the



Fig. 3. Temporal variations in the concentrations of OC and EC and OC/EC ratios at the Morogoro site during the wet and dry seasons in (a) $PM_{2.5}$ and (b) PM_{10} .

correlation coefficient (r^2) of OC and EC is higher in dry season than in wet season, indicating that during the dry season the OC and EC in PM_{2.5} had a co-genetic source. In wet season the impacts of local emission (use of fuel wood and charcoal) on organic aerosols could be more significant than the long-range transport of aerosols. In PM₁₀, the correlation coefficients of OC and EC were somewhat comparable between the two seasons, suggesting that similar emission and transport processes affect OC and EC in this fraction (Fig. 4b). Emissions from wood fuel and agricultural waste burning as well as forest fires are possible sources of the carbonaceous components, together with enhanced biogenic volatile organic compounds released from vegetation followed by photochemical oxidation.

3.2.2 Water-soluble organic carbon

Water-soluble organic carbon (WSOC) is a major fraction in atmospheric aerosols that contributes to the number density of cloud condensation nuclei. The mean concentrations of WSOC in PM_{2.5} and PM₁₀ aerosols were 3.0 ± 1.0 and $3.5 \pm 0.6 \,\mu\text{gC}\,\text{m}^{-3}$ in wet season and 3.9 ± 0.9 and $5 \pm 1 \,\mu\text{gC}\,\text{m}^{-3}$ in dry season, respectively (Table 1). Substantially larger average PM_{2.5} / PM₁₀ ratios for WSOC (>80%) were obtained, which are much higher than those for OC (Fig. 2). This result indicates that WSOC is largely enriched in fine particles. Main contributors to WSOC are probably biomass-burning particles and secondary organic aerosol, because both are known to be preferentially associated with the PM_{2.5} size fraction (e.g. Fuzzi et al., 2007).

In PM_{2.5}, fractions of WSOC in OC (WSOC/OC) were 72 % in wet season and 67 % in dry season, whereas those of PM₁₀ were a little smaller (63 %) in both seasons. However, considering that biomass-burning aerosol is highly watersoluble (e.g. Kundu et al., 2010), one would expect larger WSOC/OC ratios in dry season than in wet season. It is



Fig. 4. Relations between OC and EC in (a) $PM_{2.5}$ and (b) PM_{10} during the wet and dry seasons in Morogoro.

surprising that WSOC/OC ratio in PM_{10} is similar for both seasons, whereas in PM2.5 the production of WSOC was slightly more favourable during the wet season. This result may suggest that short-chain organic compounds with polar functional group (e.g. dicarboxylic acids) may contribute more to WSOC during wet season via aqueous-phase oxidations of various organic precursors (Carlton et al., 2007; Miyazaki et al., 2009). Short-chain dicarboxylic acids and related compounds are generally produced through photochemical oxidation of various precursors, and are abundantly present in the PM2.5 contributing to the WSOC fraction (Kawamura et al., 2005, 2007, 2010). PM₁₀, however, contains high molecular weight organic compounds that are water-insoluble. High molecular weight lipids including dicarboxylic acids are emitted to the atmosphere by soil re-suspension (Kawamura and Gagosian, 1990) and fungal spore emission (e.g. humic-like substances and proteins) (Wang et al., 2009, 2011; Mkoma and Kawamura, 2013).

Time series of WSOC and WSOC/OC in $PM_{2.5}$ and PM_{10} during wet and dry seasons in Morogoro are shown in Fig. 5. In $PM_{2.5}$, the WSOC and WSOC/OC do not show any clear seasonal trend at times during the sampling period, being in contrast to PM_{10} , which showed high WSOC in dry season. Precipitation events may have scavenged WSOC during wet season, or WSOC-enriched particles may have acted as cloud condensation nuclei. During those days with higher WSOC, elevated levels of levoglucosan and mannosan were observed (Figs. 6 and 7), suggesting a possible biomass-burning contribution to WSOC. Other studies reported that WSOC may possibly originate from biogenic aerosols and/or wood burning (Andreae and Merlet, 2001; Maenhaut and Claeys, 2007), and the aging of organic aerosols could cause enhanced WSOC/OC ratios (Aggarwal and Kawamura, 2009).

3.3 Levoglucosan and mannosan concentrations and ratios of levoglucosan to mannosan and K⁺

Concentrations of levoglucosan (LG) during the campaigns ranged from 38 to 408 ng m^{-3} in PM_{2.5} and from 49 to 659 ng m⁻³ in PM₁₀, whereas mannosan (MN), an isomer of levoglucosan, ranged from 3 to 35 ng m⁻³ in PM_{2.5} and from



Fig. 5. Temporal variations of concentrations of WSOC and WSOC/OC ratios at Morogoro during the wet and dry seasons for $PM_{2.5}$ and PM_{10} .



Fig. 6. Temporal variations of levoglucosan in $PM_{2.5}$ and PM_{10} at Morogoro during the wet and dry seasons.

5 to 59 ng m^{-3} in PM₁₀ (Table 1). Higher concentrations were obtained during the dry season than during the wet season. Time series concentrations for the two biomass-burning tracers in PM2.5 and PM10 aerosols are shown in Figs. 6 and 7, respectively. The two tracers also strongly correlated $(r^2 = 0.91 - 0.98)$ in both PM_{2.5} and PM₁₀ during wet and dry seasons (Fig. 8). The highest concentrations of levoglucosan and mannosan in PM2.5 were observed from 29 July to 3 August 2011, whereas in PM_{10} higher values are from 1 to 3 August 2011. Field burning of agriculture residue (e.g. corn straw) after the harvest and enhanced biomass burning (e.g. wild and forest fires) are common in Tanzania and eastern and southern Africa during the dry season (July-September), which are likely responsible for the elevated concentrations of levoglucosan and mannosan as observed between 29 July and 3 August.

The mean $PM_{2.5}/PM_{10}$ concentration ratios for LG and MN were >90% and >89%, respectively, in the samples collected during dry season (Fig. 2). This indicates that



Fig. 7. Temporal variations of mannosan in $PM_{2.5}$ and PM_{10} at Morogoro during the wet and dry seasons.



Fig. 8. Correlation between levoglucosan and mannosan in (a) $PM_{2.5}$ and (b) PM_{10} during the wet and dry seasons at Morogoro.

biomass-burning products are enriched in the fine aerosol fraction. Other studies also found similar results using other tracers or techniques (Maenhaut et al., 1996; Liu et al., 2000). During the wet and dry seasons, LG and MN were found to account for, on average, 3-6% of WSOC and 3-4% of OC in both PM_{2.5} and PM₁₀. These results suggest that the studied aerosols are significantly derived from biomass-burning activities in both wet and dry seasons, which have previously been thought to be different between the two seasons (Simoneit et al., 1999; Puxbaum et al., 2007; Stone et al., 2010; Zhang et al., 2012).

Table 2 shows the ratios of biomass-burning tracers: levoglucosan (LG), mannosan (MN) and water-soluble K^+ . The concentrations of those tracers and OC are found in Table 1. The LG/MN ratios were found to be fairly high, ranging from 10 to 13 in $PM_{2.5}$ and from 9 to 13 in PM_{10} (Table 2). Those high ratios suggest that the biomass burning in Tanzania is associated with the burning of hard wood and crop residue because the LG / MN ratios can be used to distinguish the biomass categories with lower ratios for soft wood (3-5) and higher ratios (>10) for hard wood and crop residues (e.g. Engling et al., 2009). The observed LG/MN ratios in this study are lower than those (>100 and 21.7) reported by Engling et al. (2006) and Iinuma et al. (2007), respectively, from Africa savannah grass. But our ratios are comparable to or lower than those observed at rural and background sites in Asia where fuel wood (as main energy source) and biomass-

Season/size	LG/MN		K ⁺ /OC		LG/K ⁺		LG/OC ratio (%)		LG/PM ratio (%)	
	Min–Max	$Mean\pm SD$	Min–Max	$Mean\pm SD$	Min–Max	$Mean \pm SD$	Min-Max	$Mean \pm SD$	Min–Max	$Mean\pm SD$
PM _{2.5} size fraction										
Wet	11.0-13	11 ± 0.8	0.06-0.27	0.11 ± 0.06	0.82-0.57	0.37 ± 0.1	0.8–10	4 ± 3	0.32-0.85	0.59 ± 0.2
Dry	10.0-13	11 ± 1.1	0.14-0.36	0.25 ± 0.7	0.12-0.35	0.18 ± 0.7	3.0-5.0	4 ± 0.6	0.32-1.05	0.69 ± 0.3
PM ₁₀ size fraction										
Wet	9.0-12	10 ± 0.8	0.03-0.20	0.11 ± 0.04	0.16-0.87	0.38 ± 0.2	1.6–5.0	3 ± 1	0.26-0.78	0.46 ± 0.2
Dry	9.0–13	10 ± 1.1	0.14-0.36	0.23 ± 0.06	0.95-0.24	0.17 ± 0.5	2.0-5.0	4 ± 0.6	0.20-0.67	0.49 ± 0.1

Table 2. Mean ratios and ranges of levoglucosan to mannosan (LG/MN), potassium to organic carbon (K^+ /OC), levoglucosan to potassium (LG/K⁺) and levoglucosan to PM mass (LG/PM) in PM_{2.5} and PM₁₀ during the wet and dry seasons.

burning smoke from agricultural residues has a greater influence (Fu et al., 2008; Zhang et al., 2012). On the other hand, our LG/MN ratio of $PM_{2.5}$ samples is higher than those reported from residential burning and forest fires in Maine, United States of America (Medeiros et al., 2006); Aveiro, Portugal (Pio et al., 2008); and Rondônia, Brazil (Decesari et al., 2006).

We calculated the levoglucosan to K^+ (LG/K⁺) ratios using the emission factors reported by Andreae and Merlet (2001) from tropical and sub-tropical sites impacted by the burning of savanna and grassland vegetation, biofuel, and agriculture residues to be 0.82, 6.4 and 0.63, respectively. Gao et al. (2003) observed a LG/K⁺ ratio of about 1.0, whereas linuma et al. (2007) measured a ratio of 20 for savannah grass in Africa. The reported values from other studies are somewhat higher than LG/K⁺ ratios of 0.17–0.38 from this study in Morogoro. The lower LG/K⁺ ratios in Morogoro suggest that our sampling site in Tanzania may be influenced by emissions of K⁺ from the burning of plants in Tanzania and also from the soils that contain a high abundance of K⁺ in Morogoro, as discussed below.

3.4 Relations between biomass-burning indicators

The concentrations (Table 1) and temporal trends of levoglucosan, mannosan (Figs. 6, 7) and nss-K⁺ (Fig. 9) coincide with each other during the sampling period in Morogoro, indicating that they all can be used as biomass burning tracers in Tanzania. Biomass burning sources are generally characterized by high K⁺ contents (Cachier et al., 1991; Andreae and Merlet, 2001). In this study, K⁺ was mostly associated with PM_{2.5} particles, suggesting that biomass burning is an important source, especially in the dry season, when 79 % of K⁺ in PM₁₀ was present in the fine (PM_{2.5}) size fraction (Fig. 2). Fine K⁺ is a well-known biomass contributor in tropical aerosols (Maenhaut et al., 1996).

 K^+/OC ratios could be an indicator to distinguish biomass burning from other OC sources. The K^+/OC ratios in Morogoro varied from 0.06 to 0.36 in PM_{2.5} and from 0.03 to 0.36 in PM₁₀ aerosols. These ratios are comparable to or higher than K^+/OC ratios reported for savannah burning (0.08–0.10) (Echalar et al., 1995) as well as tropical forest (0.06), burning of biofuel (0.01) and charcoal (0.08)



Fig. 9. Temporal variations of $nss\mathcal{K}^+$ and $nss\mathcal{K}^+\mathcal{OC}$ ratios in $PM_{2.5}$ and PM_{10} during the wet and dry seasons.

and agricultural residue burning (0.04-0.13) calculated from emission factors (Andreae and Merlet, 2001). It is of interest to note that the higher end of the K^+ / OC ratios in Morogoro is much greater than those reported from other areas, a point to be discussed later. As expected, slightly higher K⁺ / OC ratios were observed in July-August (dry season), about a factor of 2 higher than the ratios in wet season (Fig. 9), suggesting that influence of biomass burning on ambient aerosols is more significant in dry season. The similar levels of K^+ / OC ratios in $PM_{2.5}$ and PM_{10} (Fig. 9) suggest that K^+ and OC are emitted from similar sources (e.g. biomass burning and biogenic emission) in both fractions. In spite of the different concentration levels between PM2.5 and PM10, the two fractions exhibit almost the same sensitivity to biomass burning. A good correlation was obtained between nss-K⁺ and OC especially in dry season for both $PM_{2.5}$ ($r^2 = 0.67$) and PM_{10} ($r^2 = 0.74$), suggesting that OC is mostly derived from biomass burning (Engling et al., 2006; Herckes et al., 2006; Engling et al., 2011).

The mean nss-K⁺/EC ratios for the wet and dry seasons were 0.9 ± 0.3 and 1.6 ± 0.5 in PM_{2.5} and 1.0 ± 0.5 and 1.8 ± 1 in PM₁₀, respectively. Interestingly, these ratios are higher than those calculated from emission factors reported by Andreae and Merlet (2001) for tropical forest (average (0.4) and the burning of agricultural residues (average (0.2)). Further, the ratios are much higher than those reported in Christchurch, New Zealand (average 0.05), where wood and coal burning were considered as major sources of domestic heating (Wang et al., 2005). The higher nss-K⁺/EC ratios obtained in the biomass- and biofuel-burning aerosols from Tanzania should be associated with the high levels of K⁺ in the soils of Tanzania (Wickama and Mowo, 2001; Joachim et al., 2008). The plants growing in Tanzania may contain a high abundance of potassium due to the high levels of this element in the soils in Morogoro, Tanzania, where mica and feldspar are abundant (Wickama and Mowo, 2001; Holmborn, 2003). The mineralogy of soils where plants grow may determine the high levels of K^+/EC ratios in the Tanzanian aerosols. Biomass burning of plant matter and agriculture residue may release larger quantities of K⁺-rich particles to the atmosphere in Tanzania. Because PM₁₀ samples also showed high nss- K^+ / EC ratios, direct emission of K^+ from soils may contribute to the coarse aerosols. However, it should be minimal because K⁺ is largely enriched in the fine particles as mentioned above.

Nss-SO₄²⁻ to EC ratios in PM_{2.5} were 0.3 ± 0.2 (range: 0.1–0.8) in wet season and 0.3 ± 0.1 (range: 0.1–0.5) in dry season. Nss-SO₄²⁻/TC ratios in PM_{2.5} ranged from 0.01 to 0.05 with an average of 0.03 in wet season and 0.04 in dry season. These values are comparable to SO₄²⁻/TC (0.03 ± 0.01) ratios reported in Brazilian biomass smoke (Ferek et al., 1998). Furthermore, our SO₄²⁻/OC and NO₃⁻/OC ratios are comparable to those reported from individual fires in Brazil (Ferek et al., 1998). Therefore, these comparisons suggest that Morogoro aerosols are mostly influenced by different biomass-burning sources (Ferek et al., 1998).

The correlations of biomass marker species (levoglucosan and mannosan) with nss-K⁺, WSOC and OC were also examined to evaluate the source strength of biomass/biofuel burning. During the dry season, levoglucosan and mannosan showed a good correlation with nss-K⁺ in both PM_{2.5} and PM_{10} (Fig. 10a, b), suggesting that biomass burning contributed to the particulate K in the dry season. Levoglucosan (and mannosan) also positively correlates with WSOC and OC in PM_{2.5} especially in dry season (Fig. 10c, e) (figure for mannosan not shown). Strong relations were found for WSOC and OC with levoglucosan (and mannosan) in PM_{10} especially in the dry season (Fig. 10d, f). These results suggest that emissions from biomass burning significantly contribute to WSOC and OC during the campaigns. A positive correlation ($r^2 = 0.54$) was found between WSOC and nss-K⁺ in PM_{2.5} during the dry season. In the wet season, **Fig. 10.** The relationship between levoglucosan and nss-K⁺, OC and WSOC in PM_{2.5} (**a**, **c**, **e**) and PM₁₀ (**b**, **d**, **f**) at Morogoro during the wet and dry seasons.

no correlation (PM_{2.5}) and poor correlation (PM₁₀) were observed between WSOC and nss-K⁺. These results again demonstrate that biomass burning is an important source of fine organic aerosols in rural Tanzania during the dry season.

Biomass combustion is common in both rural and urban areas in Tanzania and other African countries. Forest fires covering small areas and short duration are also common in Tanzania, with highest frequency in dry season. Recent studies on land cover and cropland distribution reported that larger areas of East Africa, where Tanzania is located, are covered with tropical grasses, deciduous tropical forest, shrubs and woodlands (Still et al., 2003; Rommerskirchen et al., 2006; Vogts et al., 2009). Most southern African countries are characterized by savanna ecosystems, and the main vegetation types in Madagascar and Mozambique (possible source regions of air masses) are tropical forest, woodlands and shrubs (Arino et al., 2008). Land use change, especially conversion of natural vegetation to cropland, can result in a range of environmental impacts including aerosol loading into the ambient atmosphere.

Field burning of agriculture residues and forest/wild fires occur in dry season (July to October) in southern Africa including Tanzania (Swap et al., 1996). Hence, it is very likely that contributions of biomass burning to ambient organic aerosols in Tanzania are important in dry season. Active fire spots were observed from MODIS satellite images during the dry season in June to end of October (Fig. 11) in Mada-gascar, Mozambique and Tanzania where air masses mostly travelled through the region to our sampling site in Mo-rogoro during our campaigns (http://earthobservatory.nasa.gov/GlobalMaps/view.php?d1=MOD14A1_M_FIRE).





Fig. 11. Biomass-burning active fire spots detected by MODIS satellite images (from NASA website) over southern and eastern Africa and Madagascar in July 2011.

3.5 Contributions of biomass/biofuel burning to OC and PM mass

Various approaches can be used for source identification; however, the use of simple tracers can be quite valuable. Here, we use levoglucosan to estimate the contributions from biomass- and biofuel-burning activities to the OC and PM mass. Levoglucosan is considered as a good indicator of biomass burning (Simoneit et al., 1999). It is reasonable to assume that it derives nearly exclusively from this source in Tanzania. Andreae and Merlet (2001) reviewed the emissions of trace gases and aerosols from biomass burning and provided emission factors for many pyrogenic species that are emitted from various types of biomass burning, including (1) savanna and grassland, (2) tropical forest, (3) extratropical forest, (4) biofuel burning, (5) charcoal making, (6) charcoal burning, and (7) agricultural residues. For all these burning activities, with the exception of charcoal making and charcoal burning, emission factors for particulate OC and levoglucosan are available. Although charcoal making and charcoal burning may be very important sources of OC at our site, their emission factor data are not available in Andreae and Merlet (2001), and thus these two factors were not considered in the following analysis.

The ratio of levoglucosan to OC (LG/OC) in aerosol could be used to indicate biomass-burning source contributions (e.g. Andreae and Merlet, 2001; Zhang et al., 2008). From the data given in Table 1 of Andreae and Merlet (2001), the calculated LG/OC mass percent ratios ranged from 8.0 to 8.2 % (av. 8.14 %) for the various types of biomass-burning emissions. The respective percentage of LG/PM_{2.5} ratios

ranged from 4.4 to 6.9 % (av. 5.38 %). In our study, the mean LG/OC ratios are 3–4 % in PM_{2.5} and PM₁₀ in both seasons, whereas the LG/PM ranged from 0.43 to 0.69 %; however, they are high in the dry season when concentrations of biomass-burning tracers are enhanced (Table 2). These values are comparable to or lower than those reported for biomass combustion studies from various sites (Andreae and Merlet, 2001; Mazzoleni et al., 2007; Zhang et al., 2008, 2012; Reche et al., 2012).

The percentage contributions of biomass- and biofuelburning activities to the OC and PM mass for the PM2.5 and PM_{10} during 2011 wet and dry season campaigns are given in Table 3. The calculations were done on a sample-by-sample basis, and the individual percentages were then averaged over all samples for each campaign. Here, we assumed that the contributions of emission factors in PM2.5 are the same as those in PM₁₀. A source apportionment calculation, thereby making use of the average LG/OC ratio of 8.14% derived from Andreae and Merlet (2001), indicated that on average 50 % (range: 26–68 %) of the OC in PM_{2.5} and 46 % (range: 30-67%) in PM₁₀ at our site originate from biomass- and biofuel-burning (i.e. nearly half of the OC in both size fractions and seasons). Other studies have reported the contribution of biomass/biofuel burning activities to OC to be 7-21 % in $PM_{2.5}$ and PM_{10} (Zhang et al., 2010; Sang et al., 2011; Mohr et al., 2012; Reche et al., 2012). Wood and agricultural waste burning (including corn straw that is common near our site), charcoal making, charcoal burning and biogenic organic matter could also be important contributors to the particulate OC.

Mean contributions of biomass/biofuel burning to PM mass during the sampling period, obtained by making use of the average LG / PM_{2.5} ratio of 5.38 % derived from Andreae and Merlet (2001), are 12 % (range: 5.9–19 %) for $PM_{2.5}$ and 9% (range: 3.7–15%) for PM_{10} (Table 3). The PM mass percentages in Morogoro (Tanzania) are similar to or somewhat higher than those (about 10%) reported for the PM_{2.5} and PM₁₀ mass in Europe during winter campaigns and Asia (Sang et al., 2011; Reche et al., 2012). At our site, contributions of organic matter (OM) to the PM2.5 mass (using OCto-OM conversion factor of 1.8) are 26% and 29% in the wet and dry seasons, respectively, whereas those for PM_{10} are 24 % and 26 %. The OM makes up about one quarter of the $PM_{2.5}$ and PM_{10} mass in both seasons, suggesting that soil dust (mainly from crustal matter) is a likely contributor to the PM in the current study. The OM percentages in PM10 in the present study are much lower than those (48% and 44 %) from the 2005 wet and dry season campaigns at Morogoro (Mkoma et al., 2009a, b). Other studies in rural European sites reported the contributions of biomass smoke to OM in the range of around 47–68 % (Puxbaum et al., 2007). Therefore, our results suggest that mixed biomass-burning activities from domestic use of biofuels (in rural and urban areas), which prevails throughout the year, and biomass

Size fraction	С	ontribution of I	BB to the OC	(%)	Contribution of BB to the PM mass (%)				
	Wet season		Dry season		Wet season		Dry season		
	Min–Max	Mean \pm SD	Min–Max	Mean \pm SD	Min–Max	Mean \pm SD	Min–Max	Mean \pm SD	
PM _{2.5}	26–68	47 ± 11	36–62	52 ± 7	6.0–16	11 ± 3	5.9–19	13 ± 5	
PM ₁₀	30–67	46 ± 10	30–58	46 ± 8	4.9–15	8.5 ± 3	3.7–12	9 ± 3	

Table 3. Mean percentage contribution to the OC and PM mass from biomass/biofuel burning (BB) for the $PM_{2.5}$ and PM_{10} during 2011 wet and dry season campaigns in Morogoro.

burning (enhanced in dry season) largely influence the air quality in Tanzania.

4 Summary and conclusions

In this study, the impacts of biomass/biofuel burning and other emission sources on atmospheric aerosols (PM2.5 and PM_{10}) have been investigated during the wet and dry seasons at a rural background site in Tanzania, East Africa. We found that concentrations of biomass/biofuel burning tracers (levoglucosan, mannosan and water-soluble nss-K⁺) were higher in the dry season. Levoglucosan and mannosan showed a good correlation with nss- K^+ in PM_{2.5} and PM₁₀ in particular during the dry season. Similarly, the biomassburning tracers positively correlated with WSOC and OC in $PM_{2.5}$ and PM_{10} especially in dry season. The K⁺/OC ratios varied from 0.06 to 0.36 in $PM_{2.5}$ and from 0.03 to 0.36 in PM₁₀ comparable to or higher than those reported for savannah burning, tropical forest, burning of biofuel, charcoal and agricultural residue (Echalar et al., 1995; Andreae and Merlet, 2001). We found that the K^+ / OC and K^+ / EC ratios are higher in our site than at other study sites. The higher ratios are associated with high levels of potassium in the soils of Morogoro, Tanzania. The mean LG/OC ratios were 3-4% in PM_{2.5} and PM₁₀ in both seasons. The average contributions of biomass/biofuel burning to OC were 47% in $PM_{2.5}$ and 46 % in PM_{10} during wet season and 52 % in PM_{2.5} and 46 % in PM₁₀ during dry season, whereas those to PM mass were 10% and 13% in PM_{2.5} and 8.5% and 9% in PM₁₀ during wet and dry seasons, respectively. This study suggests that mixed biomass- and biofuel-burning activities could have a significant impact on the air quality in the Tanzanian atmosphere. This study also provides, for the first time, the baseline data for biomass-burning tracers (i.e. levoglucosan and mannosan) for the eastern and southern African region, where biomass- and biofuel-burning activities have a significant impact on regional and global climate.

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