



Chemical characterization and source apportionment of fine particulate matter in Eastern Africa using aerosol mass spectrometry.

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Abstract.

Ambient air pollution poses a significant threat to public health, particularly in low and middle-income countries, where detailed data on particulate matter (PM) mass and composition are scarce. We conducted a year-long study on PM composition and sources in Eastern Africa (Kigali, Rwanda). The annual mean concentration of PM₁ was 31 µg/m³, with slightly higher concentrations during the dry season. Organic aerosols (OA) contributed 73% of the observed PM₁ mass, black carbon (BC) 16%, nitrate 6%, sulfate and ammonium 2% each, and chlorine 1%. BC is approximately 60% due to fossil fuel and 40% from biomass burning emissions. Tracer ions detected by the mass spectrometer suggest that photochemistry plays a significant role in the formation of secondary OA during the daytime (6:00 am to 6:00 pm), while primary OA dominates in the morning and evening due to increased anthropogenic activity and shallower boundary layer height. PM₁ in Kigali is primarily composed of Oxygenated Organic Aerosols (OOA, 45%), Hydrocarbon-like OA (HOA, 32%), and Biomass Burning OA (BBOA, 23%). Secondary organic aerosol (SOA) accounted for 47% and 41% of PM₁ during the wet and dry seasons, respectively, while primary OA (POA: BBOA + HOA) contributed 53% and 59%. This suggests that seasonal changes in



PM₁ mass in Kigali are primarily driven by deposition rather than shifts in emissions, chemical processing, or source strengths.

1. Introduction

Air pollution exposure has severe impacts on public health. Air pollution is classified as the second leading risk factor for deaths in Africa(Health Effects Institute. 2022). The adverse effects of air pollution manifest as cardiovascular diseases, respiratory diseases, and cerebrovascular diseases(Brito et al. 2018a; Health Effects Institute. 2022; WHO 2025, n.d.). In 2019, ambient air pollution was estimated to cause 4.2 million premature deaths globally, with 89% occurring in low- and middle-income countries (LMICs), including 1.1 million deaths in Africa. (UN Environment 2019; WHO 2025, n.d.) Studies in the Global South, including Eastern Africa, have revealed that particulate matter (PM), defined as solid or liquid particles suspended in the air, is the air pollutant most responsible for human health impacts and a leading risk factor across sub-Saharan Africa (SSA)(Health Effects Institute 2022). PM mass concentrations in Africa often reach unhealthy levels compared to the World Health Organization's (WHO) air quality guidelines, posing significant health risks to public health and the environment (Gaita et al. 2014a; Langley Dewitt et al. 2019; Onyango et al. 2024a; Singh et al. 2021a, 2021b). Rapid population growth, urbanization, energy consumption, and motorization have been reported as drivers of air pollution in LMICs (Abera et al. 2021; Brito et al. 2018b; Health Effects Institute. 2022; REMA 2018a).

Eastern Africa and the Global South, in general, face a noticeable scarcity of air pollution measurements(AirNow 2025; openaq 2025). This hinders the understanding of air pollution impacts. Developing effective regulatory strategies requires information on air pollutant sources, which in turn require measurements of PM composition. The data gaps are even larger for PM composition measurements than for PM mass concentrations(Dhammapala 2019).

The few published studies on air pollution in Eastern Africa showed that PM concentrations exceed the WHO guidelines(Gahungu and Kubwimana 2022a; Kalisa, Kuuire, and Adams 2023; McFarlane et al. 2021; Ndamuzi et al. 2024a; Onyango et al. 2024a; REMA 2018a) and higher concentrations of PM_{2.5} (particulate matter smaller than 2.5 microns in diameter) are present in urban areas(Health Effects Institute. 2022). PM_{2.5} concentrations in eastern African cities are 3 to >10 times higher than the 5 µg/m³



55 air quality guidelines (AQG) set forth by the World Health Organization. Gahungu and Kubwimana(Gahungu and Kubwimana 2022b) used low-cost sensors and found that the daily mean concentration of $PM_{2.5}$ in Kigali, Rwanda, was approximately $30 \mu g/m^3$ in the rainy season and $55 \mu g/m^3$ in the dry season. Kalisa et al. measured hourly average $PM_{2.5}$ concentrations of $25 \mu g/m^3$ in the city center of Kigali(Kalisa et al. 2018). Onyango et al., using US State Department $PM_{2.5}$ monitor data and
60 filter collection, reported that the mean $PM_{2.5}$ in Kampala, Uganda is $59.4 \mu g/m^3$ (Onyango et al. 2024a). McFarland et al. reported mean $PM_{2.5}$ concentrations of $43.5 \mu g/m^3$ in the central African city of Kinshasa, Democratic Republic of Congo (DRC). (McFarlane et al. 2021). Ndamuzi et al., using low-cost sensors coupled with mathematical models, reported an annual mean $PM_{2.5}$ of $35.0 \mu g/m^3$ in Bujumbura, Burundi (Ndamuzi et al. 2024a).

65 There is limited information on PM composition and sources in Eastern Africa. Studies on $PM_{2.5}$ chemical speciation and source apportionment in Eastern Africa identified traffic emissions (36%) and biomass/secondary aerosols (28%) as the main sources of $PM_{2.5}$ in Mbarara, Uganda(Onyango et al. 2024b)while mineral dust and traffic emissions together account for 74% of $PM_{2.5}$ in Nairobi, Kenya(Gaita et al. 2014b). The chemical composition and sources of $PM_{2.5}$ in other Eastern African cities
70 are still unexplored, and without detailed data on PM composition, source identification remains unknown.

Rwanda, an Eastern African country, is experiencing rapid population growth in its urban centers, particularly in its capital city, Kigali. The annual rate of urbanization is 4.1%, contributing to a high population density of 2,330 inhabitants per km^2 as of 2023(Kigali city 2024b). Notably, 85% of
75 households in Kigali rely on biomass burning for cooking, which includes wood, charcoal, and waste materials(MINEFRA 2018a). Additionally, 13% of households use petroleum-based fuels such as diesel, kerosene, and liquified petroleum gas (LPG), while 2% utilize electricity for cooking(MINEFRA 2018a; Rwanda Ministry of infrastructure 2018). In 2019, air pollution-related deaths in Rwanda were estimated at 9,286, and an estimated economic cost of \$349 million(Fisher et al. 2021), following a rise of 41%
80 from 2008 to 2019 (GBD 2024; Taghian et al. 2024).

To better understand PM chemical composition, concentration, and sources in Eastern Africa, we conducted long-term ground measurements of PM mass concentration and chemical composition using



an aerosol mass spectrometer at a central site in Kigali, Rwanda. This study uses a 12-month(April 2023 to May 2024) dataset that captures the four alternating rainy and dry seasons in Rwanda and Eastern Africa in general. These data represent the first mass spectrometry-based PM composition measurements in the region, offering high-time resolution insights. The findings not only enhance an understanding of PM composition but also provide a vital tool for policymakers to develop data-driven air pollution mitigation strategies and targeted interventions in this region.

2. Methods

2.1 Sampling location

Real-time PM₁ data were collected from April 2023 to May 2024 at a fixed monitoring station located at the University of Rwanda, College of Science and Technology (UR/CST) at latitude -1.9619, longitude 30.0645 in Nyarugenge District, Kigali City (Figure S1, Supporting Information).

Kigali is characterized by rolling hills, with steep slopes connecting a network of valleys and ridges at an altitude of approximately 1,500 meters above sea level(Rwanda Meteo 2025). The monitoring site is in the city center, on the third floor of a three-story building at UR/CST. The site is surrounded by a mix of commercial and residential neighborhoods, with busy roads in the city center in Nyarugenge District, making it representative of an urban background station.

Like many Eastern African cities, Kigali experiences four alternating seasons: the long dry season (June, July, and August), the short rainy season (September, October, and November), the short dry season (December, January, and February), and the long rainy season (March, April, and May). The annual average rainfall in Kigali is 1170 mm, the annual mean temperature is 22° C, and the annual maximum temperature is 33° C(Kigali city 2024b; World bank 2021).

Kigali City is experiencing rapid urbanization with a population of 1,745,555 inhabitants(Kigali City 2024a), leading to an increase in traffic and frequent traffic congestion. As of 2018, the vehicle fleet in Kigali was dominated by imported used vehicles, with 95.2% of the total fleet being older than 10 years. (UN Environment 2018; UN Environment 2019) These vehicles burn high-sulfur fuel, as the fuels used in most eastern African countries are 50 ppm sulfur for diesel and 100 ppm sulfur for gasoline(Environmental Compliance institute 2018; Miller and Jin 2018).



110 2.2 In-situ measurements of PM and BC

A quadrupole Aerosol Chemical Speciation Monitor (Q-ACSM) from Aerodyne Research was used to measure the ambient concentration and composition of non-refractory PM₁ (NR-PM₁) in real-time (Ng, Herndon, et al. 2011a). The ACSM uses aerosol mass spectrometry to speciate NR-PM₁ components, including organic aerosol (Org), sulfate (SO₄), nitrate (NO₃), ammonium (NH₄), and chloride (Chl).
115 During sampling, ambient air is sampled through an inlet line equipped with a 1 µm cyclone to remove larger particles. The sample air is drawn into the ACSM through a ½-inch stainless steel tube at a flow rate of 3 L/min using an isokinetic sampling line. The particles are then focused by aerodynamic lenses before impacting on the vaporizer heated at ~ 620° C to generate gas phase molecules that are then ionized by the filament. The ionized particles are then detected by the quadrupole mass spectrometer with unit
120 mass resolution.

As described by Ng et al., (Ng, Herndon, et al. 2011a) aerosol mass concentrations are calculated from the difference between the sample mode and filter mode of the instrument, with each scan comprising 28 filter samples and 28 ambient samples, scanning over 32 seconds each to generate a 30-minute time resolution data points for our analysis.

125 The instrument's aerodynamic lens was aligned, and the flow calibration was performed before starting the data collection in April 2023. Similarly, the relative ionization efficiency (RIE) calibration (Ng, Herndon, et al. 2011a) was conducted at the beginning of the data collection period, during sampling, and again after 12 months of sampling, which covers the data duration. The RIE values and ammonium nitrate (NH₄NO₃) response factors (RF) were applied to their respective sampling period. During all RIE
130 calibrations, the response factor and RIE for both NO₃ and SO₄ remained stable within a 10% variation across the four calibrations conducted during the campaign.

Ambient black carbon (BC) mass concentration was measured via light absorption at 880 nm with a Magee AE33 aethalometer. The AE33 automatically corrects for filter loading effects using a dual-spot sample collection method; hence, the filter loading effect compensation was not performed during this
135 analysis (Magee scientific 2015). BC source apportionment to distinguish BC from biomass burning and BC from fossil fuel burning was performed using the method described by Sandradewi et al. (Sandradewi



et al. 2008), This approach assumes that the total optical absorption at 950 and 470 nm is the sum of biomass burning and fossil fuel burning fractions.

PM_{2.5} mass concentrations were obtained from a Teledyne T640 PM_{2.5} mass monitor (Teledyne 2016) installed at the U.S. embassy in Kigali. The embassy is located approximately 10 km from the University of Rwanda sampling site. Temperature, relative humidity, wind speed, wind direction, and rainfall data were obtained from a nearby automatic meteorological station owned by Rwanda Meteo located at -1.955793 latitude and 30.05641 longitude for the same period of time.

145 2.3 Data analysis

ACSM data were processed offline using ACSM local v1.6.2.1 in Igor Pro (WaveMetrics 2024). Given that the instrument is equipped with a standard vaporizer, the particle Collection Efficiency (CE) was adjusted to 0.5, as recommended in the literature (Ng, Herndon, et al. 2011b). The CE accounts for particle loss associated with shape-related collection losses at the vaporizer due to the inefficient focusing of non-spherical particles, particle losses resulting from bounce effects of larger particles in the ionization region, and aerodynamic inlet losses as a function of particle diameter (Huffman et al. 2005; Liu et al. 2007; Matthew, Middlebrook, and Onasch 2008; Ng, Herndon, et al. 2011a). The RIE values for NH₄ and SO₄, along with the Response Factor (RF) for NO₃, were automatically loaded into the instrument's data acquisition software and applied to the raw data after each calibration. These factors were also used during post-processing to ensure that the correction factors were consistently applied, ensuring accuracy in the final data analysis.

To ensure the quality of the calibration and instrument performance, the total NR-PM₁ was compared to the PM_{2.5} data obtained from the USA embassy T-640 (see Figure S3 in the supplemental information). Positive matrix factorization (PMF2.exe) (Paatero and Tapper 1994) and the PMF evaluation tool (PET v.3.06) was used to perform the source apportionment of the organic aerosol for the whole dataset. The PMF-derived source profiles were systematically selected based on the approach explained by Zhang et al. (Zhang et al. 2011)



3. Results and discussion

165 3.1 NR-PM₁, BC, and PM_{2.5} mass concentration, chemical composition, and seasonal variation.

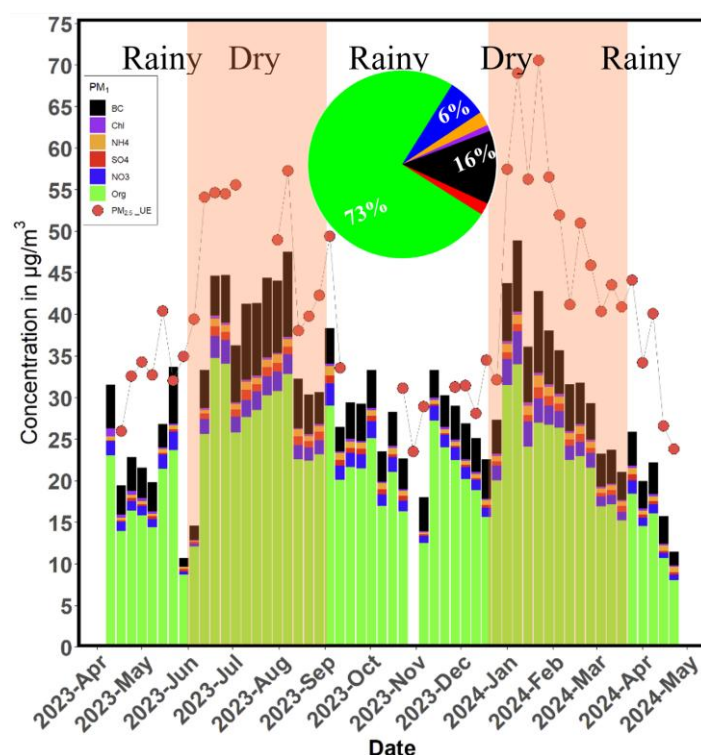
Figure1 shows the temporal variations in weekly mean concentrations of PM₁ components, including organic aerosols (Org), nitrate (NO₃), sulfate (SO₄), ammonium (NH₄), chloride (Cl), and black carbon (BC). There is a clear seasonal variation in both PM₁ (NR-PM₁ + BC) and PM_{2.5} concentrations, with higher concentrations during the dry seasons (mean PM₁ = 37 µg m⁻³) than the rainy seasons (mean PM₁ = 28 µg m⁻³). The annual mean concentration in PM₁ was 31 µg m⁻³ whereas PM_{2.5} at the U.S. Embassy in Kigali (denoted as PM_{2.5}_UE) was 41 µg m⁻³. The measured annual mean BC concentration was 4.5 µg/m³ with a seasonal mean concentration of 5.9 µg/m³ in the dry seasons and 3.8 µg/m³ in the rainy seasons.

Concurrent measurements of PM₁ and PM_{2.5} in Kigali showed a strong correlation ($R^2 = 0.91$) with a slope of 0.65 (See Figure S3 in supporting information). Some of the mass differences between PM₁ and PM_{2.5} can likely be attributed to the impact of dust, which cannot be detected by the ACSM. In addition, site characteristics may play a role, as the US embassy site is classified as an urban near-road site compared to UR/CST site classified as an urban background. The annual mean concentration of PM_{2.5} of 41 µg m⁻³ is comparable with the mean PM_{2.5} concentration of 42.6 µg m⁻³ reported by Gahungu et al. in the same city (Gahungu and Kubwimana 2022b). However, it was higher than the daily mean concentration of PM_{2.5} of 25 µg m⁻³ reported by Kalisa et al. for a 3-month data span in both dry and wet seasons (Kalisa et al. 2018).

Organic aerosols (OA) dominate the PM₁ composition, accounting for 73% of the total mass loading, followed by black carbon (BC) at 16%. Inorganic species collectively contributed 11% of the total PM₁, distributed as follows: NO₃ at 6%, SO₄ at 2%, NH₄ at 2%, and Cl at 1%. The compositions are similar to the findings from a remote area in the same country by Kirago et al., 2022 (Kirago et al. 2022) (mean of all seasons): OM at 72%, BC at 4.5%, SO₄ at 12.6%, NO₃ at 4%, and NH₄ at 5.2%. However, SO₄ and NH₄ fractions were 2.6 times lower, and the BC fraction was 4.5 times higher at this urban background site compared to the remote site.



- 190 The high contribution of OA to PM_{10} mass was also recorded in previous studies in African cities. For example, aircraft measurements in South Africa showed that OA contributed 53% of PM_{10} mass, though there was a larger contribution of sulfate (27%) (Brito et al. 2018b). Additionally, filter-based measurements in Dar es Salaam, Tanzania, showed that OA accounted for 68% and inorganic species accounted for 24% of PM_{10} mass (Mkoma et al. 2009).
- 195 OA dominates the PM_{10} composition across both rainy and dry seasons, with a relatively stable fractional contribution: 74% on average during the dry season (mean of JJA and DJF) and 71% during the rainy season (mean of MAM and SON). A similar seasonal composition was also recorded at a remote site in the same country as 72 % and 69% as the mean of the dry and rainy seasons respectively (Kirago et al. 2022)



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Figure 1: Measured PM_{10} and $PM_{2.5}$ in Kigali, Rwanda. Each stacked bar represents the weekly mean concentrations of NR- PM_{10} components (SO₄, Org, NO₃, NH₄, CHl) and Black Carbon (BC). Red dots indicate $PM_{2.5}$ concentrations measured at the U.S. Embassy, located about 10 km from the ACSM sampling site. The pie chart depicts the overall mean fractional contribution of the total PM_{10} mass



loading (NR-PM₁ + BC). The shaded areas highlight the dry seasons in Rwanda, providing context for seasonal variations in particulate
205 matter.

3.2 Diurnal variations in PM₁ mass and composition

The campaign average diurnal variation in PM₁ components (NR-PM₁ and BC) is presented in Figure 2; seasonal diurnal patterns are shown in Figure S4 in the supporting information. The diurnal patterns show
210 the interplay of emissions and atmospheric processes on pollutant concentrations. These include traffic emissions from busy roads near the sampling site, with 85% of vehicles in Kigali being over 20 years old (Anon 2018; MINEFRA 2018b; Niyibizi et al. 2015; UN Environment 2019). Additionally, emissions from solid fuels contribute significantly, as biomass energy accounts for approximately 85% of the total energy used for cooking in Kigali (MINEFRA 2015; Rwanda Ministry of infrastructure 2018; UN
215 Environment 2019), including emissions from restaurants in the downtown area and densely populated residential areas near the station. Diurnal variability is also influenced by meteorology, including changes in the planetary boundary layer height (PBLH) that is expected to be shallower during night and morning time (6:00 pm to 7:00 am) and expand during the daytime and hence controls the particle mixing volume and concentration.

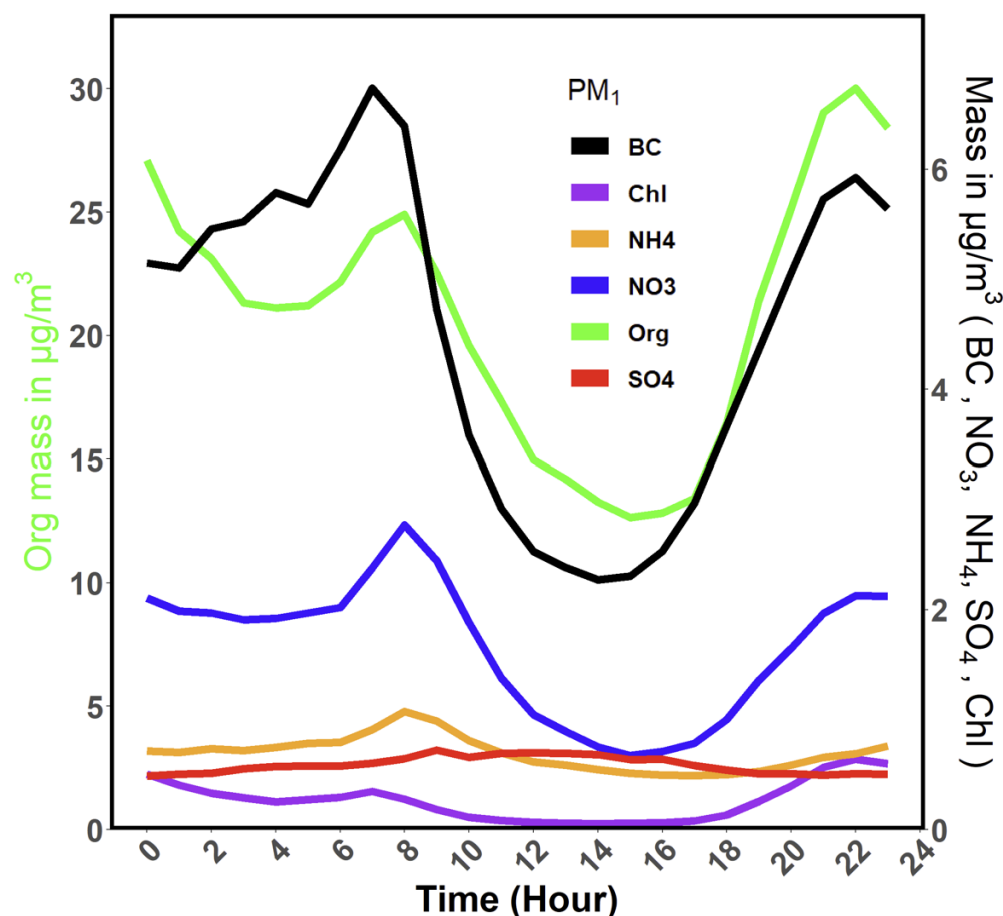
220 OA, BC, and NO₃ all exhibit an early morning peak (6:00 am to 9:00 am), a midday decrease, and a second evening peak (6:00 pm to 10:00 pm). The morning peak coincides with increased anthropogenic activities, likely associated with rush hour traffic and cooking activities in the morning. The midday dips in particle concentrations can be attributed to enhanced atmospheric mixing and reduced anthropogenic activities. Daytime increases in the planetary boundary layer height increase the mixing volume and hence
225 reduce the particle concentration during the daytime (Subramanian et al. 2020a). Additionally, higher wind speeds during the daytime contribute to particle dilution (Figure S2 in the Supporting Information). The evening peak is likely due to a combination of decreasing boundary layer height after sunset (typically around 6 pm) and the evening rush hour.

The intra-day variations in concentrations of OA, BC, and NO₃ are large. OA concentrations are ~20 –
230 25 µg m⁻³ from midnight to 10 am but fall to ~12 µg m⁻³ in mid-afternoon. Similarly, BC concentrations change from ~7 µg m⁻³ to ~2.5 µg m⁻³ over the same period.



The shape of the diurnal patterns offers insight into potential emissions sources and aerosol processing. The morning peak in BC occurs at 7:00 am. This is one hour earlier than the corresponding morning peak in both OA and NO₃. There are a few possible reasons for the earlier peak in BC. One is that the anthropogenic source mix changes throughout the morning, with the relative impacts of cooking and traffic changing over the period from ~6:00– 10:00 am. A second possibility is that the slight delay in the OA and NO₃ peaks reflects the importance of secondary OA and nitrate formation, and the one-hour delay in their peaks relative to BC is the result of the time needed for chemical conversion from precursor gases to PM components.

OA, BC, and NO₃ all have contributions from local emissions and, therefore, exhibit strong intra-day patterns associated with anthropogenic activities. SO₄, on the other hand, has a much flatter diurnal profile. There is no morning increase in sulfate, though there is a slight midday increase from 10 am to 6 pm. This suggests that SO₄ concentrations in Kigali are regional, reflecting the lack of local SO₄ emissions or SO₂ sources that would contribute to daytime SO₄ formation. For example, there are no coal-fired power plants in Rwanda or nearby areas of neighbouring countries (Global Energy Monitor 2025). Back trajectory analysis (Figure S12) showed that the emissions from the recent activity of the stratovolcano Nyiragongo, which erupted in June 2021 and is located at 1.52°S latitude and 29.25°E longitude in the Democratic Republic of Congo (DRC), did not move toward Kigali (Smithsonian Institution 2025).



250 Figure 2: Campaign average diurnal profile of NR-PM₁ composition (Chl, NH₄, NO₃, SO₄, Org, and BC) with OA plotted on the left y-axis and the other species on the right y-axis.

3.3 Average mass spectrum and contribution of tracer ions

Figure S7 in the Supporting Information shows the campaign-average mass spectrum (MS) measured by the ACSM. The most abundant ions in the mass spectrum are m/z 18 (H_2O^+) and 44 (CO_2^+). M/z 44 is commonly used as a tracer for aged OA (Ng, Canagaratna, et al. 2011a). It accounts for 10.5% of the measured organic aerosol mass whereas m/z 18 is for particle water contents.

Other prominent tracer ions in the mass spectrum include m/z 43, which has contributions from both C_3H_7^+ , associated with fresh emissions, and $\text{C}_2\text{H}_3\text{O}^+$, linked to secondary organic aerosol. M/z 43 contributed 7% of the OA mass in Kigali. Additional tracer ions such as tracers for urban fresh traffic



emissions (e.g., m/z 57 ($C_4H_9^+$)) and fresh biomass burning emissions (m/z 60, ($C_2H_4O_2^+$))(Atabakhsh et al. 2023; Christodoulou et al. 2023; Cubison et al. 2011; Werden et al. 2023) together contribute approximately 3.5% of the average PM_{10} mass loading.

Inorganic species also contribute significantly to PM_{10} , with notable nitrate peaks at m/z 30 and m/z 46, sulfate at m/z 48 and m/z 64, and ammonium at m/z 16 and m/z 17. Small peaks at m/z 35 and m/z 36 indicate chloride makes a minimal contribution (see Figure S7 in the supporting information).

NO_3 accounts for 6% of the total PM_{10} mass and consists of both organic and inorganic nitrate species. The ratio of m/z 46 to m/z 30 is often used to quantify the relative amounts of inorganic and organic nitrate using equation 1(Budisulistiorini et al. 2013; Lanz et al. 2007; Ng, Canagaratna, et al. 2011b).

$$f_{orgNO_3} = \frac{(R_{ambient} - R_{NH_4NO_3})(1 + R_{orgNO_3})}{(R_{orgNO_3} - R_{NH_4NO_3})(1 + R_{ambient})} \quad (1)$$

Here, f_{orgNO_3} is the mass fraction of the total ACSM nitrate that is organic, and R is the ratio of m/z 46 to m/z 30 for ambient data, NH_4NO_3 , and organic nitrates. $R_{NH_4NO_3}$ can be measured for each instrument during periodic ionization efficiency calibrations. R_{orgNO_3} can be determined from atomizing laboratory standards of organic nitrates, or from literature values. For example, Brito et al.(Bruto et al. 2018a) used $R_{orgNO_3} = 0.1$.

An alternative approach, proposed by Day et al(Day et al. 2022), uses a ratio-of-ratios. This method amends equation 1 using $R_{NH_4NO_3}/R_{orgNO_3}$. The advantage of this approach is that it accounts for instrument-to-instrument variations in both $R_{NH_4NO_3}$ and R_{orgNO_3} , instead relying on a constant ratio of ratios for most organic nitrates ($R_{NH_4NO_3}/R_{orgNO_3} = 2.75$).

The mean $R_{ambient}$ for our data is 0.25 ± 0.6 , with hourly values ranging from 0.02 to 1.11. Figure S5 in the supporting information shows the results of calculating f_{orgNO_3} both by assuming $R_{orgNO_3} = 0.1$ and using the ratio-of-ratios approach. The former attributes the ambient nitrate as 61% organic, whereas the ratio-of-ratios approach yields f_{orgNO_3} near 100%. Therefore, both approaches indicate that organic nitrates make up a significant fraction of the observed nitrate. The presence of a slight peak in NH_4 during the



morning rush hour (Figure 2) suggests that at least a portion of the nitrate is inorganic, and therefore the f_{OrgNO_3} from the ratio-of-ratios approach might be an overestimation.

The diurnal profile of four OA tracer ions (m/z 43, 44, 57, and 60) are shown in Figure 3 -. The mass concentrations of all four ions show a similar diurnal pattern as the total OA mass (shown in Figure 2):
290 there is a morning increase in concentration, followed by an afternoon minimum, and concentrations rise again in the evening. While all four of these ions show the same general pattern, the slight differences in their behavior provide insights into PM sources and chemical processing in Kigali.

M/z 60 is a tracer for biomass burning emissions. The morning maximum concentration occurs from 6-7 am and is 1.5 times higher than the minimum concentration observed from 2-4 am. Many people in Kigali
295 (85%) use biomass fuel for cooking(MINEFRA 2015). This morning's increase in m/z 60 is likely from cooking activities before the start of the workday.

M/z 57 is commonly used as a tracer for fresh fossil fuel combustion emissions, especially from traffic. The morning peak in m/z 57 mass concentration ($0.6 \mu\text{g}/\text{m}^3$) occurs at 7:00 am, with slightly lower concentrations at 8:00 am. This mirrors the diurnal pattern of BC (Fig. 2), which also has a morning
300 maximum at 7 am ($6.8 \mu\text{g}/\text{m}^3$). The high concentrations of both BC and m/z 57 suggest that the morning rush hour is most intense from 7-8 am. The morning peaks in BC and m/z 57 occur slightly after the morning peak in m/z 60 ($0.3 \mu\text{g}/\text{m}^3$); this suggests that while both cooking and traffic are important sources in the morning hours, cooking activity starts slightly before traffic volume increases.

M/z 44 is a tracer for oxygenated secondary organic aerosol (SOA). M/z 44 also has a morning peak. The
305 morning peak in m/z 44 ($2.3 \mu\text{g}/\text{m}^3$) occurs at 8:00-9:00 am, after the earlier morning peaks in m/z 60 and m/z 57. This is suggestive that the morning increase in m/z 44 is due to the rapid formation of SOA and that the vehicle and biomass-burning emissions associated with anthropogenic activities are important sources of SOA precursors. Traffic emissions are a well-known source of SOA precursor vapors(Gaita et al. 2014b; Ndamuzi et al. 2024b; Onyango et al. 2024b; REMA 2018b; Subramanian et al. 2020a; UN
310 Environment 2019; West et al. 2020) this is especially true for older vehicles, poorly maintained, or otherwise higher emitting than new vehicles with modern emission control systems(Anon 2018).

M/z 43 is a marker for both primary emissions (C_3H_7^+) and SOA ($\text{C}_2\text{H}_3\text{O}^+$). Our quadrupole ACSM has unit mass resolution and, therefore, cannot separate these isobaric ions. The diurnal pattern of m/z 43



reflects its dual nature as a marker of primary and secondary OA in Q-ACM. The concentration of m/z 43 rises rapidly from 6-7 am, coincident with the morning increase in the concentration of m/z 57. This initial increase in m/z 43 concentration is likely associated with fresh vehicle emissions. There is a further increase in m/z 43 concentration from 8-9 am, coincident with the rapid increase in m/z 44 concentration. This further increase in m/z 43 is likely due to SOA formation from vehicles and biomass burning emissions.

The temporal patterns of the four marker ions shown in Figure 3 provide insight into sources and atmospheric processes. They all decrease during the afternoon. As with the PM components shown in Figure 2, this decrease is likely due to a combination of changing anthropogenic emissions (e.g., less traffic outside of the morning rush hour), increasing boundary layer height, and faster winds. Concentration rises again in the evening due to a combination of a sinking boundary layer after sunset and evening traffic and cooking activities.

While concentrations of all four tracer ions decrease during the day, the decrease in m/z 44 is much smaller than for the other ions. For example, the ratio of the morning peak (7:00 am) to the afternoon trough (2:00 pm) in m/z 57 is ~ 3.3 , and the ratio in m/z 44 concentration (8:00 am versus 4:00 pm) is ~ 1.3 . This is likely due to the sustained formation of SOA throughout the day. Days in Kigali are consistently sunny (Figure S2 in the Supporting Information), and there is likely a large pool of reactive organic gases available to form SOA. Thus, the impact of boundary-layer driven dilution is larger for ions associated purely with primary emissions (e.g., m/z 57) than for ions associated with SOA.

Concentrations of m/z 43, 57, and 60 are all higher in the evening hours ($\sim 8:00$ - $11:00$ pm) than during the morning rush hour. The pattern of traffic and cooking emissions is also reversed in the evening compared to the morning. The late-day increase in the concentrations of m/z 57 and 43 precedes the increase in m/z 60. This is consistent with expected activity patterns, as people travel home from work before cooking at night.

The mass fractions of each tracer ion, shown in Figure 3b, offer additional insight into the competition between emissions, chemistry, and dilution. The diurnal variation of the mass fraction of m/z 57 in OA (f_{57}) is similar to the trend in m/z 57 mass. There are morning and evening peaks, with a slightly higher peak in the evening and a midday trough. The ratio of the peaks to the trough is smaller for f_{57} than m/z



57 mass (1.5 and 3.3 for f_{57} and m/z 57, respectively). This reflects the large role that boundary layer dilution plays on mass concentrations and suggests that traffic activity, and hence emissions, remain high throughout the day.

345 The diurnal pattern in f_{44} shows a different trend. It rapidly increases after 7 am and peaks at $f_{44} = 0.14$ at 2:00 pm. The diurnal trend in f_{44} thus indicates that there is rapid formation of SOA, especially in the hours immediately following the high emissions associated with the morning rush hour. After 2 pm, f_{44} gradually decreases as solar intensity diminishes until sunset at 6:00 pm. Taken together, Figs 3a and 3b suggest that there is strong and sustained SOA production throughout the day and that SOA production
350 is fast enough to at least partly counteract the large and rapid boundary layer-driven dilution that occurs during midday.

The diurnal profile of the normalized contribution of m/z 43 (f_{43}) remains relatively flat throughout the day. This is a result of the combined role of m/z 43 as an indicator for primary and secondary OA. $C_3H_7^+$ is emitted during the morning rush hour and its concentration decreases with the expansion of the
355 boundary layer and oxidation chemistry. $C_2H_3O^+$ increases due to SOA formation through photochemistry. These opposing behaviors result in a relatively constant f_{43} throughout the day.

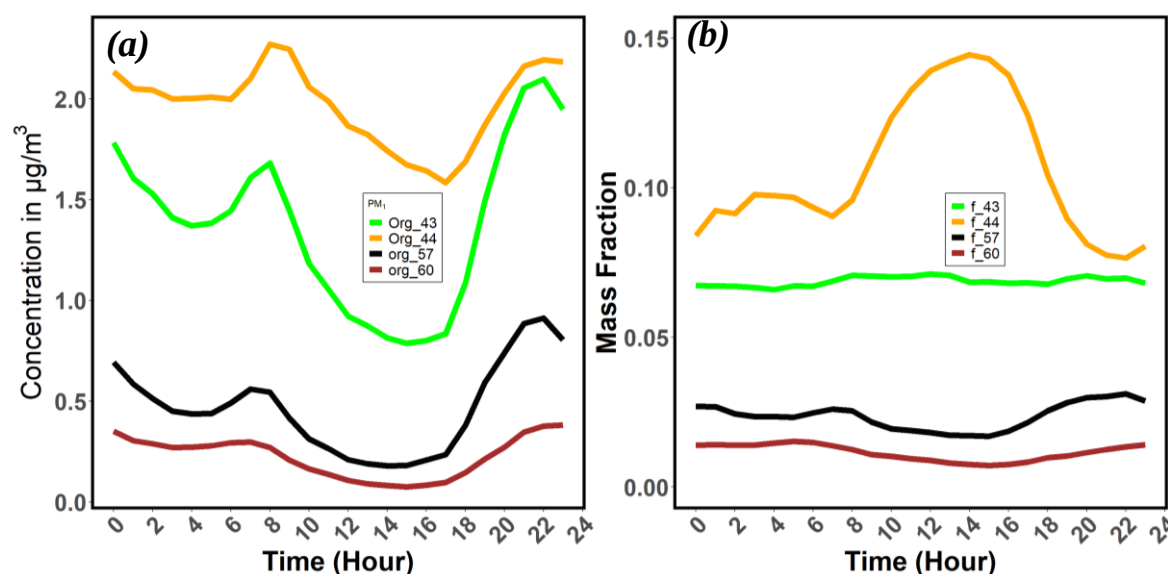


Figure 3: Diurnal variation of individual tracer ions in terms of (a) absolute mass concentrations and (b) mass fraction.



3.4 PM Source apportionment

360 We performed PMF source apportionment of the OA-only mass spectrum using PMF2.exe and PMF Evaluation Tool version 3.08. The number of factors and source profiles were systematically selected and validated following the recommendations of Ulbrich et al. (Zhang et al. 2011). The most interpretable solutions were chosen based on Q/Q_{Expected} (Q : sum of squared scaled residuals), changes in the total sum of squared scaled residuals with an increasing number of factors, normalized fractional contributions of individual source profiles, and factor spectra comparisons with standard AMS mass spectra (Zhang et al. 2011). Section IV of the Supporting Information shows more details on the PMF analysis, including a comparison of three versus four-factor solutions.

Figure 4(a) shows the mass spectral profiles of deconvolved organic PM_{10} source apportionment using PMF. Three factors, Oxygenated Organic Aerosol (OOA), Hydrocarbon-like Organic Aerosol (HOA), and Biomass-Burning Organic Aerosol (BBOA) were identified. The mass spectrum of HOA is characterized by the pronounced hydrocarbon ion series of $\text{C}_n\text{H}_{2n+1}$ and $\text{C}_n\text{H}_{2n-1}$ (He et al. 2010). Important peaks in this mass spectrum include m/z 41, 43, 55, 57, 69, and 71. In the BBOA mass spectrum, the most prominent peak is at m/z 29, with other significant peaks at m/z 60, 55, and 43. The OOA mass spectrum is characterized by the large abundance of m/z 44.

375 The HOA, BBOA, and OOA factor profiles show strong similarity to published profiles from ambient deconvolved spectra in other cities, indicating consistency with existing data. When compared to the reference mass spectra, all three factors exhibited strong correlations. The HOA factor had an R^2 value of 0.88 and a slope of 0.85, while the BBOA factor achieved an R^2 of 0.85 and a slope of 0.94. Similarly, the OOA factor profile showed an exceptionally strong correlation with the reference spectra, with an R^2 value of 0.99 (see Figure S10 in the supplementary information document).

PMF results indicate that there are large contributions from both primary and secondary OA. Over the course of the year, the mean contributions of each factor are 45% OOA, 32% HOA, and 23% BBOA. Figure 4b further shows that there is little seasonal variation in source strengths. During the dry season, the mean composition is 47% OOA, 23% BBOA, and 30% HOA, versus 41% OOA, 28% BBOA, and 31% HOA during the wet season. This suggests that the changes in PM_{10} mass between the dry and rainy

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seasons (Figure 1) are due more to changes in particle loss (e.g., via wet deposition) rather than shifts in emissions, chemical processing, or source strengths.

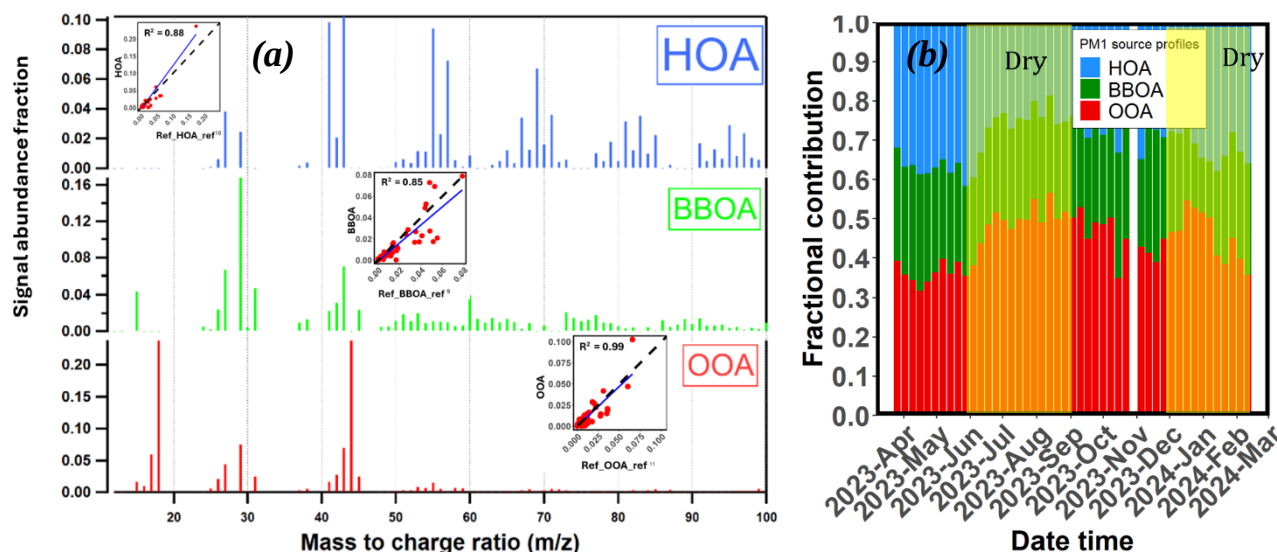


Figure 4: PMF factor profiles (a). The inset scatter plots show the comparison of the source profiles with reference source profiles, and (b) show the seasonal variations in source fractional contributions. In (b), each bar represents the mean relative fractional contribution of the three sources for each week. The yellow shading indicates the two dry seasons.

The diurnal pattern of the source strengths is shown in Figure 5. As expected, they mirror the trends in tracer ions shown in Figure 3. The OOA factor peaked during the daytime, mirroring f_{44} , underscoring the important role of photochemistry and secondary organic aerosol production at the site. OOA concentrations are less impacted by dilution than HOA and BBOA due to strong daytime SOA production.

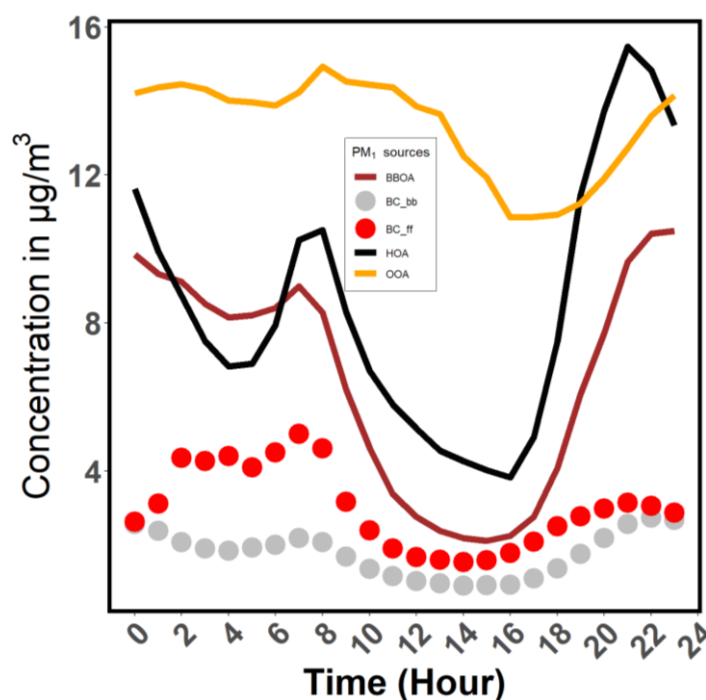


Figure 5: The lines show the diurnal concentrations of OOA, HOA, and BBOA determined from PMF source apportionment of ACSM data. The dots indicate diurnal trends of BC attributable to fossil fuel emissions (red) and biomass burning (grey).

Both BBOA and HOA factors show morning and evening peaks, which mirror anthropogenic activities. The morning peak in BBOA occurs before the morning peak in HOA associated with rush hour traffic. In the afternoon, the trend is reversed, with HOA concentrations increasing and reaching an evening peak before BBOA. For both BBOA and HOA, the evening peak in concentrations is larger than the morning peak. Throughout most of the day, concentrations of HOA are smaller than concentrations of OOA. However, during the evening peak from ~6:00-10:00 pm, HOA concentrations are larger than OOA concentrations.

BC contributes ~16% of the PM₁ mass. Using the algorithm developed by Sandradewi et al. (Sandradewi et al. 2008), BC was decomposed into BC from fossil fuel combustion (BC_{ff}) and BC from biomass burning (BC_{bb}) fractions. The annual mean BC measured in Kigali consists of 59% BC_{ff} and 41% BC_{bb}. Figure S6 in the supporting information shows that there is minimal seasonal variation in BC source apportionment. Across all seasons, BC_{ff} contributed 55–63% of the total BC mass loading, with BC_{bb} contributing 37–45% of observed BC mass. These results are contrary to the BC composition



recorded at the Rwanda Climate Observatory (RCO), which is located in a remote area. There, BC was 8-13% BC_{ff} and 87-92% BC_{bb}(Andersson et al. 2020a; Kirago et al. 2022), this difference may be
415 attributed to the site characteristics where RCO is a remote site compared to the measurement at this urban site that is likely to be influenced by the anthropogenic emissions from cooking and traffic.

The diurnal patterns of both BC_{ff} and BC_{bb} are shown alongside the PMF analysis in Figure 5. Overall, the diurnal patterns of BC_{ff} and BC_{bb} are similar to the diurnal patterns in HOA and BBOA,
420 respectively. The BC_{ff} component, represented by the red markers, exhibits pronounced peaks during the morning (6:00–9:00 am) and evening (5:00–9:00 pm) hours, corresponding to increased vehicular emissions during rush hours under the influence of a shallower planetary boundary layer. In contrast, the BC_{bb} component, depicted by the grey markers, shows relatively consistent levels throughout the day, with slight increases observed during the late evening (8:00–10:00 PM) and early morning (6:00–10:00
425 AM) hours.

There was a positive correlation between hourly BC and BBOA ($R^2 = 0.86$), BC_{bb} and BBOA ($R^2 = 0.86$), and BC and m/z 60 ($R^2 = 0.94$). Similarly, the HOA factor showed a linear relationship with BC ($R^2 = 0.73$), BC_{ff} ($R^2 = 0.61$), and m/z 57 ($R^2 = 0.95$), (see S10 and S11 in the supplement information) indicating significant contributions from fossil fuel combustion and biomass burning to the total PM mass
430 at the site.

While there is limited data on air pollution sources in other East African countries, our results are consistent with the past literature. In Mbarara, Uganda, a neighboring country of Rwanda, elemental PM_{2.5} composition analysis and PMF revealed large contributions of traffic emissions and biomass burning, contributing 36% and 28% of the total PM_{2.5} mass, respectively (Onyango et al. 2024b). In Nairobi,
435 Kenya, Gauta et al. (Gaita et al. 2014b) reported that traffic-related emissions accounted for 39% of the total PM_{2.5} mass.

In Western African countries, primary organic aerosols (POA) are also major contributors to PM_{2.5} mass. For instance, in Bamako, Mali, POA contributes 60–65% of the total PM_{2.5} mass. Similarly, in Dakar, Senegal, traffic emissions (including motor vehicle tailpipe emissions and resuspended road dust) account
440 for 45–49% of the total PM_{2.5} mass loading (Doumbia et al. 2023; UN Environment 2018, 2019).



Different countries in Eastern Africa are implementing various measures to reduce PM emissions. For example, in the transportation sector, Rwanda has adopted electric mobility policies to reduce traffic-related PM emissions (Republic of Rwanda 2021). A 2021 strategic paper on electric mobility adaptation by the Ministry of Infrastructure (Republic of Rwanda 2021) outlines import and excise duty exemptions on electric vehicles (EVs), spare parts, investments in charging infrastructure, and access to high-occupancy vehicle lanes for EVs. Additionally, this strategy applies to a carbon tax on high-emission vehicles (old petrol-powered vehicles) to discourage the use and importation of older and polluting vehicles. This strategy introduced tax exemptions on LPG to improve the affordability and adoption of EVs (Republic of Rwanda 2021). To enhance compliance, the government of Rwanda has established motor vehicle inspection centers, requiring commercial vehicles to undergo biannual inspections and personal vehicles to be inspected annually. This initiative ensures that older, high-PM-emitting vehicles are monitored and regulated. Research by Subramanian et al. (Subramanian et al. 2020b) shows that Rwanda's car-free initiatives, such as car-free zones and car-free days, can successfully reduce $PM_{2.5}$ concentrations by 10-12 $\mu g/m^3$ and black carbon (BC) by 1 $\mu g/m^3$, demonstrating the effectiveness of vehicle emissions reductions in PM concentrations.

In the household energy sector, Rwanda is actively working to reduce PM emissions from biomass cooking by lowering biomass energy use from 85% to 44% (Republic of Rwanda 2022). The 2022 ministerial guidelines on clean cooking technologies promote the adoption of LPG, biogas, liquid fuels, and improved clean-burning cookstoves to reduce indoor and outdoor PM emissions from biomass combustion hence the large-scale distribution of low-emission cookstoves to rural communities further supports this transition (Republic of Rwanda 2022).

For industrial PM emissions, the eastern African community developed different air emission Standards for instance EAS 750:2010, EAC 751:2010, and EAC 1047:2022, which regulate air ambient, cement factory, and tailpipe emissions respectively (Rwanda Standard Board 2010). This effort is expected to improve air quality and reduce the impact of air pollution on public health, the environment, and climate change.



4. Conclusion

The measurements presented here show that PM₁ in Kigali is impacted by high primary emissions of BC and OA, along with strong secondary production of OA and nitrate. This is consistent with Rwanda being a nation with a relatively old and higher-emitting vehicle fleet, high usage of solid biomass fuels for cooking, and high solar insolation.

The transport sector in Rwanda is dominated by old vehicles imported from other countries. Rwanda's registered motor vehicles, including two-wheeled taxis ("motos") but excluding security organs and government vehicles, totaled 191,015 in 2017 and 264,524 in 2020. Of these, only 15% were new vehicles, while the remaining 85% were imported used vehicles, 95% of which were manufactured before 2005 (Niyibizi et al. 2015; REMA 2018b; Republic of Rwanda 2021; UN Environment 2018, 2019). While we do not have specific emissions measurements for the vehicle fleet, the ambient data presented here suggest that vehicles in Rwanda are high emitters of both BC and OA. Additionally, these vehicle emissions contribute precursor vapors for SOA production.

In Rwanda, as in most Eastern African countries, biomass (e.g., charcoal, wood, and biogas derived from waste materials) accounts for more than 80% of total energy consumption (MINEFRA 2018b). Our analyses show that biomass burning, even in urban Kigali, contributes 23% of OA and 41% of BC. Some of the OA and BC attributable to biomass burning are emitted locally, as evidenced by the morning and evening peaks in BBOA and BC_{bb}. However, the morning and evening enhancements in BBOA and BC_{bb} are relatively small, and there are also high concentrations of BBOA and BC_{bb} overnight (e.g., from 12:00 am to 4:00 am). Anderson et al. (2020) and Kirago et al. (2022) measured a mean concentration of ~0.5 µg m⁻³ at the remote Rwanda Climate Observatory; this BC was 91% attributed to biomass burning. If we assume that the Rwanda Climate Observatory measurements represent a regional concentration of biomass-related BC, then about one-third of the biomass-burning BC in Kigali is from regional sources.

There are modest seasonal variations in PM₁ mass concentrations, with about 10% higher concentrations of both PM₁ and PM_{2.5} during the dry season. Source apportionment analysis of both OA and BC shows



495 smaller variations in source strengths across seasons. This suggests that the main reason for higher PM concentrations in the dry seasons is a reduction in wet deposition, rather than increased source strengths. However, there remains the possibility that there exist season-dependent shifts in emissions sources. For example, the rainy seasons have a higher fraction of BC from biomass-burning sources than the dry seasons. This may be due to biomass burning for agricultural residue disposal or because of transported emissions from open burning in the northern and southern regions of Rwanda (Andersson et al. 2020b).

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While regional cooperation frameworks exist, weak enforcement remains a major challenge in addressing transboundary PM pollution. Strengthening compliance requires the establishment of a legally binding regional agreement with clear PM reduction targets and penalties for non-compliance. Prioritizing a joint regional PM monitoring network would enable real-time tracking of PM sources, supporting evidence-based enforcement. Additionally, empowering cross-border regulatory bodies to conduct independent audits, inspections, and enforcement actions is essential to ensure compliance with PM standards. Strengthening regional funding mechanisms for clean energy transitions and emission reduction projects targeting PM sources, such as biomass burning, industrial activities, and vehicle emissions, would provide critical financial support while fostering collaboration among governments, industries, and environmental agencies across the region.

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5. Data availability.

All data presented in this work can be obtained by directly contacting the corresponding author at apresto@andrew.cmu.edu upon request. Data will also be posted to a publicly available server (<https://kilthub.cmu.edu/>) upon final acceptance.

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6. Competing interests.

The authors declare that they have no conflict of interest.



7. Author contribution

The experimental design was done by AAP, ALR, and TH. Data collection was carried out by AAP and
520 TH. TH performed the data analysis and compiled the instrumental data. TH, AAP, and ALR wrote the
paper, with all authors contributing significantly to the interpretation of the results, discussions, and
finalization of the paper.

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