Research article

Air pollution in Kigali, Rwanda: spatial and temporal variability, source contributions, and the impact of car-free Sundays

R Subramanian[1,2,7,*], Abdou Safari Kagabo[3], Valérien Baharane[3], Sandrine Guhirwa[3], Claver Sindayigaya[3], Carl Malings[1,2,8], Nathan J Williams[4,7], Egide Kalisa[5], Haofan Li[1], Peter Adams[1], Allen L Robinson[1], H. Langley DeWitt[6], Jimmy Gasore[3,7], and Paulina Jaramillo[4,7]

[1]Center for Atmospheric Particle Studies, Carnegie Mellon University, Pittsburgh, PA 15218, USA
[2]Observatoire Sciences de l'Univers-Enveloppes Fluides de la Ville à l'Exobiologie, CNRS UMS3563, France
[3]Department of Physics, University of Rwanda, Kigali, Rwanda
[4]Department of Engineering and Public Policy, Carnegie Mellon University, Pittsburgh, PA, USA
[5]Institute for Applied Ecology New Zealand, School of Science, Auckland University of Technology, Auckland 1142, New Zealand
[6]Center for Global Change Science, Massachusetts Institute of Technology, Cambridge, MA, USA
[7]Kigali Collaborative Research Center, Kigali, Rwanda
[8]NASA Postdoctoral Program Fellow, Goddard Space Flight Center, Greenbelt, Maryland, MD 20771, USA
*Corresponding author, E-mail: subu@cmu.edu subu@lisa.u-pec.fr

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Abstract

Ambient air pollution, particularly fine particulate mass ($PM_{2.5}$) and ozone (O_3), is associated with premature human mortality and other health effects, but monitoring is scarce to non-existent in large parts of Africa. Lower-cost real-time affordable multi-pollutant (RAMP) monitors and a black carbon (BC) monitor were deployed in Kigali, Rwanda to fill the air quality data gap here. $PM_{2.5}$ data were corrected using data from a coincident, short-term campaign that used standard filter-based gravimetry, while gas data were verified by collocation with reference carbon monoxide (CO) and O_3 monitors at the Rwanda Climate Observatory at Mt Mugogo, Rwanda. Over March 2017-July 2018, the ambient average $PM_{2.5}$ in Kigali was 52 µg/m^3, significantly higher than World Health Organization (WHO) Interim Target 1. Study average BC was 4 µg/m^3, comparable to mid-sized urban areas in India and China and significantly higher than BC in cities in developed countries. Spatial variability across various urban background sites in Kigali appears to be limited, while $PM_{2.5}$ at Mt Mugogo is moderately correlated with $PM_{2.5}$ in Kigali. A sharp diurnal profile is observed in both $PM_{2.5}$ and BC, with the Absorption Angstrom Exponent (AAE) indicating that the morning peak is associated with rush-hour traffic-related air pollution (TRAP) while the late evening peak can be attributed to both traffic and domestic biofuel use. $PM_{2.5}$ in the dry seasons is about two times $PM_{2.5}$ during the following wet seasons while BC is 40-60% higher. Local sources contribute at least half the ambient $PM_{2.5}$ during wet seasons and one-fourth during dry seasons. Traffic restrictions on some Sundays appear to reduce $PM_{2.5}$ and BC by 10-12 µg/m^3 and 1 µg/m^3 respectively, but this needs further investigation. Dry season ozone in Kigali can exceed WHO guidelines. These lower-cost monitors can play an important role in the continued monitoring essential to track the effectiveness of pollution-control policies recently implemented in Rwanda.

Keywords

fine particulate matter, ozone, black carbon, sub-Saharan Africa, urban air pollution, vehicular emissions, biofuel emissions, low-cost sensors

Introduction

Ambient air pollution, especially fine particulate mass ($PM_{2.5}$) and ozone (O_3), has been associated with premature human mortality (Dockery et al., 1993; Jerrett et al., 2009; Laden et al., 2006). The World Health Organization (WHO) estimates that in 2016, ambient air pollution caused about three thousand deaths in Rwanda (Brauer et al., 2012; WHO, 2018). However, such estimates can be uncertain because exposure is inferred from satellite estimates. There has been no long-term ground-based monitoring in major cities like Kigali to validate estimated

exposures. The situation is similar in many other sub-Saharan African countries (Kalisa et al., 2019; Petkova et al., 2013). This lack of monitoring due to resource limitations also hampers scientific understanding of the sources contributing to air pollution in these countries, which is essential to formulating effective environmental management policies.

In Rwanda, biomass use accounts for 85% of energy consumption, in the form of wood and charcoal (MININFRA, 2018). Wood is used in rural households and charcoal in urban households. Petroleum in the form of transportation fuel, liquified petroleum gas (LPG) for household cooking, and electricity generation accounts for 13% of national energy consumption (MININFRA, 2018). As of 2018, 77% of on-road vehicles (excluding motorcycles) were manufactured before 2005 (Duhuze, 2018). As a result, air pollution in Kigali can be significantly higher at roadside locations than at urban background locations (Kalisa et al., 2018). Rwandan air quality is also influenced by regional forest fires and seasonal weather patterns (DeWitt et al., 2019). Rwanda's electricity generation (218 MW) is composed of 45% hydropower, diesel and heavy fuel oil at 27%, methane (14%), peat (7%), and solar (6%). Diesel is used to fuel peaking power stations and for backup generation during power outages, which can be significant contributors to ambient air pollution (Farquharson et al., 2018; Subramanian et al., 2018). Some of these sources were identified by Henninger (2013) using scanning electron microscopy of filter samples.

To more accurately quantify source contributions (a requirement for effective air quality management), source apportionment is often conducted with chemical mass balance (CMB) or receptor modeling (e.g. positive matrix factorization) using organic molecular markers (Shrivastava et al., 2007; Subramanian et al., 2007) or aerosol mass spectrometry (AMS) (Zhang et al., 2011). However, such studies require extensive sample collection and offline analysis or long-term deployment of expensive equipment, especially if sources can vary between seasons. Pikridas et al. (2013) find that an observation-based method (OBM), which uses the temporal pattern of pollution measured with even a low-cost PM monitor, closely replicates the regional/urban divide based on $PM_{2.5}$ composition measurements inside and upwind of Patras, Greece. Diamantopoulou et al. (2016) use regional air quality modeling to simulate observations and find the OBM analysis of the pseudo-observations comparable to the model-computed regional/local divide.

Traffic restrictions such as car-free days, low-emission zones (LEZ), and "odd/even" policies have been used in cities worldwide to reduce air pollution. The evidence that such policies reduce human exposure is mixed. No effect was observed on air quality in Mestre-Venice (Masiol et al., 2014). London's LEZ saw decreases in nitrogen dioxide (NO_2), but no significant changes in $PM_{2.5}$ or PM_{10} (Mudway et al., 2019). Extensive traffic control measures in Beijing during the 2008 Olympics reduced median black carbon (BC) concentrations by as much as 50% (Wang et al., 2009). The Rwandan government, to promote social welfare, has recently implemented "car-free" Sundays, wherein major roads are blocked off and people take part in group exercises on car-free streets starting at 7 AM and ending either at 10 AM or at noon. In 2017, the first Sunday of each month was designated "car-free", while in 2018 that was expanded to the first and third Sundays of each month. Additionally, to curb the use of older imported cars and harmonize duty structures with other East African countries, in 2017 the Rwandan government increased duties on cars, with the increase depending on the vehicle age (RRA, 2017). The vehicle import rate dropped by 20% in the first half of 2017, likely connected to the higher import duties (Ngabonziza, 2017). Monitoring is required to quantify the impact of these policies on air quality in Kigali.

Here, we show how low-cost sensors can improve scientific understanding of air quality in resource-challenged countries. The high time resolution of the RAMPs and BC monitor enables an examination of diurnal patterns in each season, which is not possible with integrated daily filter samples. We apply OBM to the RAMP and BC datasets to get a preliminary estimate of the regional and local contribution to Kigali air pollution in each season. Measurements at multiple "urban background" sites within Kigali allow us to examine intra-urban variability in air pollution at non-roadside locations across Kigali, unlike the urban background/roadside comparison by Kalisa et al. (2018). Long-term monitoring with RAMP monitors allows examination of the seasonal variability in ambient air pollution across multiple dry and wet seasons. Furthermore, we evaluate the impact of the "car-free Sunday" policy on air pollution. The multi-wavelength aerosol light absorption from the BC monitor helps

Table 1: *Site descriptions and deployment periods for the measurements reported in this study.*

Site	UR-CST	CMU-Africa	Gacuriro	Belle Vue	RCO
Type	Urban	Urban	Urban	Urban	Rural
Latitude	-1.96279	-1.94455	-1.9219	-1.92563	-1.58625
Longitude	30.06473	30.08961	30.09389	30.0924	29.56568
Instrument and deployment period					
RAMP #140			March-June 2017		July-Dec 2017
RAMP #145		March-June 2017		July-Dec 2017	
RAMP #152	July 17-July 18				
BC-1054	July 17-April 18				

us qualitatively identify contributions from biomass burning and fossil fuel combustion. Comparison of urban and rural air pollution provides insight into the impact of urbanization on air quality. We end with recommendations on ways to implement and improve such studies in Rwanda and other countries in the Global South.

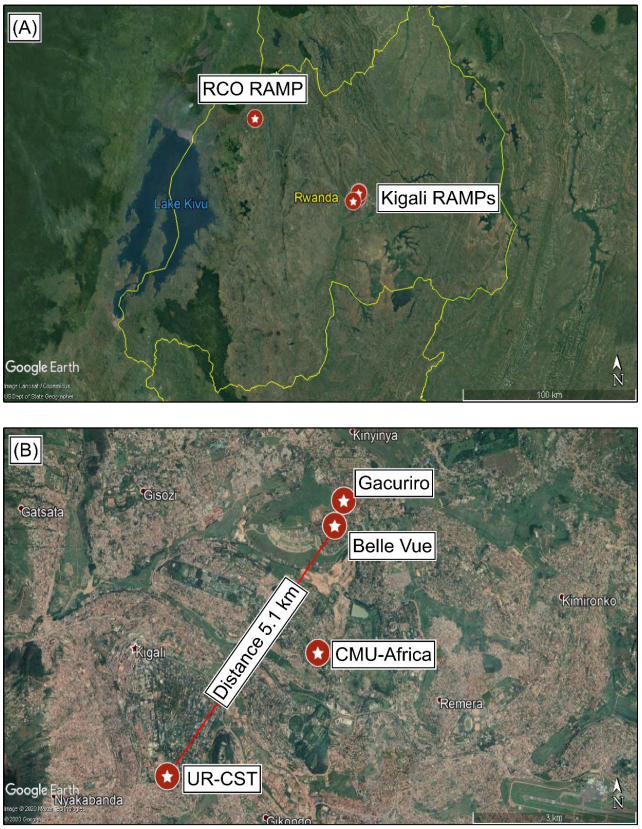

Figure 1: *Measurement sites in Rwanda (a) and specifically in Kigali (b) where RAMPs were deployed for varying periods over 2017-2018. Maps generated using Google Earth Pro.*

Methodology

Sampling locations

Starting in March 2017, RAMP monitors were deployed in Kigali at multiple locations (Figure 1) as summarized in Table 1. Kigali terrain is composed of hills and valleys; the RAMPs in this study were all located at hilltop sites. The Gacuriro and Belle Vue Estate locations are both residential neighborhoods about 0.5 km apart. The Carnegie Mellon University (CMU-Africa) site is about 2.5 km from Gacuriro, located in a commercial building complex near a major road and six stories above ground (this was the campus till late 2019). RAMP #152 and a Met-One 10-wavelength BC monitor (BC-1054) were deployed in July 2017 on the roof of the University of Rwanda's five-story College of Science & Technology building (UR-CST), where Kalisa et al. (2018) had collected filter-based samples for their urban background location over April-June 2017. This manuscript focuses on RAMP data collected at UR-CST between July 2017-July 2018. The BC data ends in April 2018 as the pump malfunctioned, which made the instrument inoperable. The Gacuriro and Belle Vue sites are about 5 km from the UR-CST site.

The RAMPs and gas sensor calibration

The RAMPs (Figure S1) were manufactured by Sensevere (now owned by Sensit Technologies, Valparaiso, IN, USA) and cost about US$ 3,000 each at the time of purchase (base unit without an external PM sensor). The RAMP monitors and calibration methodologies are described in previous work (Malings et al., 2019, 2020; Subramanian et al., 2018; Zimmerman et al., 2018). Briefly, the RAMP uses passive Alphasense (UK) electrochemical sensors to measure CO, nitrogen dioxide (NO_2), O_3, and other gases. The raw signals of the RAMP electrochemical gas sensors (collected at 4 times per minute) are processed and averaged to provide hourly ambient concentrations using generalized RAMP (gRAMP) calibration models (Malings et al., 2019) developed in Pittsburgh, Pennsylvania, USA. The gRAMP calibration models are based on data from several RAMP monitors collocated with reference gas monitors at the CMU campus in Pittsburgh in 2017 and were shown to transfer better to other locations in Pittsburgh than calibration models developed for individual RAMPs. For CO, a quadratic regression (QR) gRAMP model is used and for O_3, a hybrid random forest/linear regression ("hybrid-RF") gRAMP model is used.

For local verification of CO and O_3, RAMP #140 was collocated with reference monitors for CO and CO_2 (Picarro G2401) and O_3 (Teledyne T400) at the Rwanda Climate Observatory (RCO) on the summit of Mt Mugogo (about 70 km from Kigali and 2590 m above sea level, DeWitt et al. (2019)) over July-December 2017. Due to instrument malfunctions, collocated measurements are available for only 60 days of this six-month period. "Mugogo" linear regression, QR, and hybrid-RF models are developed using a subset (four weeks) of the collocation data; as shown in Zimmerman et al. (2018), a four week period is sufficient to develop calibration models for these electrochemical sensors. The remaining collocation data (32 days) are set aside to provide an independent or unseen data set for testing model performance, a practice previously established by our group in Zimmerman et al. (2018) and Malings et al. (2019). The performance of the Mugogo and the Pittsburgh gRAMP models is shown in Table 2. For CO, the gRAMP models showed slightly worse correlation (Pearson r) and normalized mean absolute error (CvMAE) than the Mugogo-based calibration models. For O_3, the gRAMP model is comparable to the better-performing Mugogo QR and hybrid-RF models for r and CvMAE. RAMP-specific Mugogo models show lower bias than the gRAMP models. During the dry season, most of the data from the gRAMP ozone model are within ±30% of the reference monitor data at RCO (Figure S2).

RAMP-specific models trained on one RAMP may not transfer as well to other RAMPs as the gRAMP models (Malings et al., 2019). As most of the Kigali O_3 data is from RAMPs not collocated at Mugogo, the results presented here for ambient CO and O_3 are based on the gRAMP models. As we do not have local verification for NO, NO_2, or SO_2, these data are not presented here.

Table 2: *Summary statistics showing the performance of Pittsburgh-based gRAMP and RAMP-specific Mugogo calibration models, both tested on 32 days of collocation at the Mugogo site. (These testing data were not used to build the Mugogo calibration models.)*

Model	CO			Ozone		
	Pearson r	CvMAE	Bias (ppb)	Pearson r	CvMAE	Bias (ppb)
Pittsburgh gRAMP	0.82	0.19	15.7	0.57	0.24	6.02
Mugogo linear	0.87	0.16	-4.22	0.16	0.24	1.10
Mugogo QR	0.92	0.13	-1.83	0.63	0.20	0.30
Mugogo hybrid-RF	0.90	0.16	-6.28	0.56	0.22	0.32

$PM_{2.5}$ measurements

$PM_{2.5}$ is measured using a Met-One neighborhood PM monitor (NPM) paired with each RAMP. The NPM is a nephelometer with a $PM_{2.5}$ cyclone, an inlet heater to reduce humidity effects, and a pump (flow rate 2 lpm). The $PM_{2.5}$ data are processed using methods developed based on collocations of over two dozen NPMs with US Environmental Protection Agency (EPA) federal equivalent method (FEM) beta attenuation monitors (BAM) at an urban background location and a source-impacted location in Pittsburgh (Malings et al., 2020). Either a physical (assumed composition and hygroscopic growth with scaling to BAM values) or an empirical approach (a quadratic regression with raw data, temperature, and RH as the variables) is used to convert as-reported NPM readings to "BAM-equivalent" $PM_{2.5}$ mass concentration (reported at 35% RH and 22 °C); both methods performed similarly in Pittsburgh. For Rwanda, we use the Pittsburgh-based physical approach, then apply a further localized correction to account for differences in aerosol size distribution and composition between Kigali and Pittsburgh. This localized correction of the "BAM-equivalent" $PM_{2.5}$ data is based on a comparison of the Gacuriro RAMP with 24-hour integrated filter-based measurements in April 2017 collected by Kalisa et al. (2018) at UR-CST about 5 km away (described in the SI). Briefly, the "BAM-equivalent" $PM_{2.5}$ data (corrected for hygroscopic growth but not for aerosol differences between Kigali and Pittsburgh) are strongly correlated (correlation coefficient, $r^2 = 0.77$) with the filter-based $PM_{2.5}$ for non-working days (weekends, holidays, and car-free Sundays). However, the comparison is more scattered for working days (weekdays that are not holidays), which might indicate some intraurban variability related to local activities such as traffic and industrial emissions that may be more prominent on working days. Overall, the working day "BAM-equivalent" RAMP $PM_{2.5}$ values are scaled up by 1.69 and the non-workday "BAM-equivalent" RAMP $PM_{2.5}$ data are scaled up by 1.39 (Figure S3).

BC mass and AAE measurements

The BC-1054 monitor (Met One Instruments, Inc.) deployed at the UR-CST site measures light attenuation by a filter sample at ten wavelengths between 370-950 nm. The as-reported BC mass concentrations from this monitor were processed using manufacturer-provided software (BC Load Correction 1.3.1), which corrects for known filter-loading artifacts (Kirchstetter and Novakov, 2007) using the algorithm developed by Virkkula et al. (2007). However, no correction is made for potential light absorption enhancement due to BC mixing state (Bond et al., 2006), which means the BC mass concentrations reported here may be overestimates. The Absorption Angstrom Exponent (AAE) was calculated based on all ten wavelengths as the negative slope of the relationship between the wavelength-dependent attenuation and the wavelength in a log-log space (Moosmüller et al., 2011). For fresh combustion aerosol mainly composed of black carbon and for particles small relative to the wavelength, AAE is expected to be near unity. A compilation of studies (Lack and Langridge, 2013) on fossil fuel emissions and urban pollution with BC as the dominant absorber yielded an average AAE of 1.1±0.3 (± one standard deviation). The AAE for light-absorbing organic compounds (brown carbon or BrC) associated with biomass burning or humic-like substances is higher, with estimates ranging from 2 to 6 (Kirchstetter et al., 2004; Sun et al., 2007). Hence, higher values of AAE can be used to qualitatively identify periods when non-BC aerosol components such as light-absorbing dust or biomass burning containing BrC are significant contributors to aerosol light absorption. We do not attempt to quantify the contributions of fossil fuel and biomass burning emissions here as there can be significant uncertainty associated with such methods, as described in DeWitt et al. (2019) and references therein.

Results and discussion

Intra-urban variability across Kigali

The UR-CST and Belle Vue Estate sites are about 5 km apart and have the largest paired data set across both dry (July-September) and wet (October-November) seasons. An orthogonal distance regression (ODR) fit of the paired hourly average $PM_{2.5}$ concentrations yields a slope 0.996±0.012 and effectively zero-intercept. An ordinary least-squares fit (not forced through zero) has a correlation (r^2) of 0.61. The UR-CST and Belle Vue Estate sites in Kigali are typical of urban background hilltop locations (Figure 1), unlike the urban background/roadside comparison where Kalisa et al. (2018) found significant differences. The much smaller dataset of 337 paired hourly average $PM_{2.5}$ measurements at the Gacuriro and CMU-Africa sites yields a slope of 0.967±0.03 (and an effectively zero intercept) with the ODR fit. For the sake of simplicity (as the succeeding analysis focuses on longer-term comparisons), the measurements at all four sites are averaged into a single time series providing hourly average $PM_{2.5}$ values from March 2017 to July 2018.

A comparison of the O_3 measured at the matched residential and university locations (Gacuriro and CMU-Africa; Belle Vue Estate and UR-CST) shows that most paired values are in reasonable agreement around the 1:1 line (Figure S4). An ODR fit of the 1-hour O_3 values measured at Gacuriro and CMU-Africa yields a slope of 0.97±0.01 with negligible intercept. A comparison of the UR-CST and Belle Vue estate O_3 measurements is complicated by the significantly higher O_3 apparently measured at the UR-CST site during the mid-July to mid-September period. The reason for these differences is unclear, as temperature and relative humidity (two variables likely to affect sensor performance) measured at the two urban background locations are identical. This issue of potentially high O_3 at UR-CST needs to be investigated further; here, we take a conservative approach and present the data set as a unified O_3 time-series (as we do for $PM_{2.5}$), except the UR-CST measurements are excluded from the current analysis. This restricts our analysis to the March-November 2017 period (as there is only a week of data for December 2017 from the Belle Vue estate site), which are the two 2017 wet seasons (March-April-May or MAM and ON) and the long dry season (June-July-August-September, JJAS).

Unlike O_3 and $PM_{2.5}$, differences in CO concentrations were sometimes observed at the residential locations and the campus sites in Kigali (Figure S5 of the SI). The highest concentrations were observed usually at the campus sites, which might reflect the greater traffic seen by these sites compared to residential neighborhoods. However, the Gacuriro neighborhood did experience two hours when CO exceeded 3 ppm. Overall, the study average CO concentrations were similar at UR-CST and Belle Vue Estate at 0.446±0.322 ppm and 0.447±0.283 ppm (average and standard deviation) respectively. CO was 0.504±0.499 ppm at CMU-Africa and 0.404±0.320 ppm at Gacuriro. Concentrations at rural Mugogo were even lower, at 0.225±0.097 ppm. However, the measured CO concentrations at all locations were low and far below US EPA standards (35 ppm for 1 hour). The WHO has not set guideline values for CO. The higher standard deviations at the urban sites compared to the rural site suggests that the urban areas are more likely to see large spikes of CO.

Average monthly patterns of $PM_{2.5}$ and changes from 2017 to 2018

The unified Kigali time series for ambient $PM_{2.5}$ is summarized as monthly average $PM_{2.5}$ values, covering the period between March 2017 and July 2018 (Figure 2). The least data were collected in June-July 2018 (100 and 64 hours) due to maintenance issues. For all other months, between 298-744 hours of data are available, averaging 634 hourly values per month (88% of a 30-day period). The monthly average $PM_{2.5}$ values range from 25 µg/m³ in May 2018 to 102 µg/m³ in July 2017, all higher than the WHO annual guideline of 10 µg/m³; most months also exceed the WHO's first interim target (WHO IT 1) of 35 µg/m³. The study average hourly $PM_{2.5}$ in Kigali is 52.4±33.7 µg/m³; the large standard deviation suggests significant temporal variability that will be explored further in later sections.

A comparison of the overlapping months shows a reduction of almost 40% from 2017 to 2018 for March, April, and May, and 51% for July, while the June monthly average is practically unchanged. However, June and July 2018 had the least data coverage (as noted in the preceding paragraph), and fewer than 300 hours of data are available for June 2017. The March-May comparisons are more robust with 1,683 and 1,942 hours of data available in 2017 and 2018 respectively. April 2017 had a larger fraction of non-working day data (43%) compared to the other five months (30-34%), but similar reductions are seen when the comparison is restricted to working days. An examination of spatially-resolved reanalysis data (Siebert et al., 2019) for rainfall (Rwanda Meteorology Agency, 2019) in Kigali (Figure S6) shows that the UR-CST site during MAM 2018 experienced 38 rainy days (≥5 mm/day), compared to 16 such rainy days in MAM 2017 at Gacuriro. Total rainfall was also substantially higher in 2018; 612 mm at UR-CST in MAM 2018 compared to just 180 mm at Gacuriro in MAM 2017. The more frequent and heavier rainfall in MAM 2018 could at least partly explain the significantly lower $PM_{2.5}$ in MAM 2018 compared to MAM 2017.

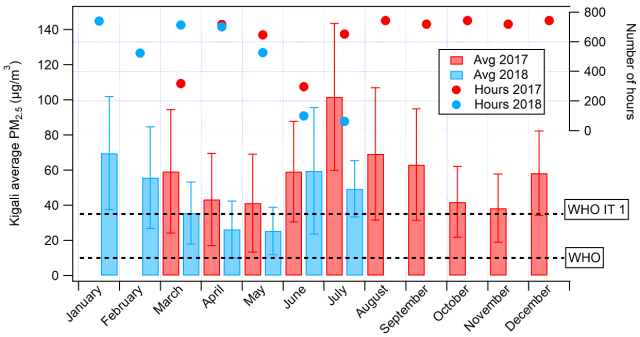

Figure 2: *Monthly average $PM_{2.5}$ measured in Kigali over the course of this study. Red bars indicate 2017 and blue bars indicate 2018 data. Error bars are one standard deviation of the hourly measurements in that month. Solid round markers show the number of hourly data points collected in that month. Horizontal dashed lines indicate WHO annual guideline and the WHO's first interim target (IT 1).*

Working day/non-Working day and seasonal differences in $PM_{2.5}$

Figure 3 shows the distribution of hourly $PM_{2.5}$ values measured in Kigali across each of the four seasons: long wet season (MAM 2017 and MAM 2018), long dry season (JJAS 2017, henceforth JJAS), short wet season (ON 2017, henceforth ON), and short dry season (December 2017 and January-February 2018, henceforth DJF.) Note that August and September are both considered "dry" here. As Figure 2 showed, the monthly average $PM_{2.5}$ value for September is closer to that of August and June, and noticeably higher than the average $PM_{2.5}$ for October and November. Additionally, September 2017 saw just 17 mm of rainfall, compared to 104 mm of rainfall in October-November 2017 (Figure S6). This suggests that our classification is appropriate for 2017.

Figure 3 shows that ambient $PM_{2.5}$ levels are higher in the dry seasons than in the wet seasons. The hourly data collected with the RAMPs on non-working days (nWD) in MAM 2018 and ON

shows a pear-shaped pattern, in contrast to the hourglass shape of MAM 2017 nWD data; this is also seen in the working day (WD) data. This suggests that there are fewer high-concentration periods in the later wet season months. WD $PM_{2.5}$ values are higher than the nWD concentrations for all seasons by 19-27%. However, as discussed earlier, WD $PM_{2.5}$ values are scaled upwards by 1.69 based on a comparison with the Kalisa et al. (2018) wet season filter measurements while nWD $PM_{2.5}$ values are similarly upscaled by 1.39 - a difference of 22%. Thus, this weekday-weekend effect could reflect Kalisa et al.'s results for April 2017 and needs to be investigated further for other months of the year.

Table 3 summarizes the seasonal averages for working days. The dry season average $PM_{2.5}$ concentrations are about two times the following wet season average; to be exact, JJAS/ON is 1.89 (95% confidence intervals 1.82-1.97) and DJF/MAM 2018 is 2.11 (95% CI 2.03-2.19). This was also observed for non-working days. Higher pollution in the dry season was also observed by Kalisa et al. (2018) for $PM_{2.5}$ in Kigali (though in campaigns of 2-4 weeks in each season) and by DeWitt et al. (2019) for BC at Mt Mugogo, who attribute the higher dry season concentrations to regional biomass burning. Kalisa et al. measured a median daily $PM_{2.5}$ of 126 µg/m³ (IQR 113-141 µg/m³) over June 15-30, 2017. The median for our JJAS hourly measurements (which does not include data for the June 15-28 period) was 66.2 µg/m³ (IQR 45.4-95.4 µg/m³). It seems that the June 15-28 period saw higher pollution based on Kalisa et al.'s results, but other parts of the same dry season were relatively cleaner based on our results, showing the importance of long-term monitoring.

BC and AAE in Kigali

A total of 6,850 hours of BC data were collected at UR-CST between July 6, 2017-April 24, 2018. The equivalent BC mass concentration (as measured at 880 nm) hourly averages ranged from 0.14 µg/m³ to 49.6 µg/m³, with a study average of 4.04±2.86 µg/m³. This value is significantly higher than the values observed in urban areas of developed countries; e.g. in the early 2000s, urban ambient BC was 2 and 1 µg/m³ respectively in the states of New Jersey and California in the USA after decades of reductions (Kirchstetter et al., 2017). The Kigali BC values are comparable to mid-sized urban areas in China and India. In Hefei (central China), annual average BC in 2012-2013 was 3.5 µg/m³ (Zhang et al., 2015). In Pune (western India), the average BC over 2015-2016 was 3.9 µg/m³ (Kolhe et al., 2018).

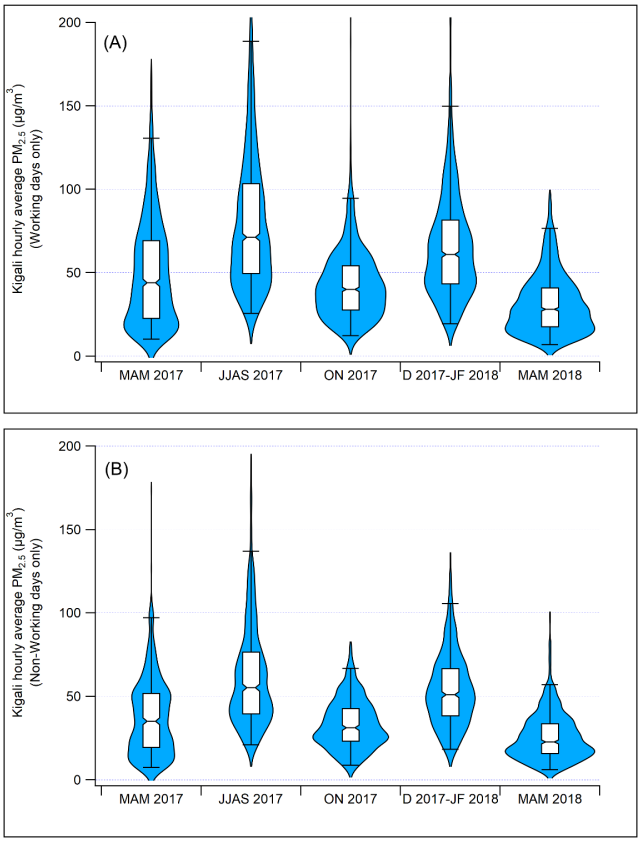

Figure 3: *Distribution of 1-hour average $PM_{2.5}$ in each of the seasons over the course of this study, grouped into (A) working day and (B) non-working day measurements. March-May (MAM) and October-November (ON) are the wet seasons; June-September (JJAS) and December-February (DJF) are the dry seasons. The boxes show the interquartile range (IQR), i.e. the 25th and 75th percentiles of data; the notch in each box shows the median value; and the whiskers mark the 2nd and 98th percentile of all data. The violin plot shows the relative distribution of all data. Y-axis truncated at 200 µg/m³ for visual clarity.*

BC working day seasonal averages are summarized in Table 3. Non-working day seasonal averages were 0.2-0.9 µg/m³ lower. The difference between the wet and dry seasons (dry season BC 40-60% higher) in Kigali is lower than the factor-of-four difference observed in similarly-tropical Pune (Kolhe et al., 2018) or even at the rural Mt Mugogo site in Rwanda, where DeWitt et al. (2019) found dry and wet season BC different by almost a factor-of-three.

The study average AAE was 1.53, with most values between 0.9-2.1 (Figure S7). As shown in Table 3, AAE was slightly lower in

Table 3: *Seasonal $PM_{2.5}$ and BC statistics for this study in Kigali, Rwanda*

Season	WD $PM_{2.5}$ average ± SD (µg/m³) (A)	Average diurnal minimum WD $PM_{2.5}$ (µg/m³) (B)	Regional WD $PM_{2.5}$ (%) C = B/A	WD BC average ± SD (µg/m³)	AAE
MAM 2017	49.9 ± 31.7	21.3	42.7	N/A	N/A
JJAS	81.2 ± 42.3	57.3	70.6	5.20±3.56	1.47±0.15
ON	42.9 ± 21.0	22.8	53.2	3.20±2.24	1.57±0.16
DJF	66.0 ± 31.6	50.7	76.8	4.52±2.96	1.52±0.16
MAM 2018	31.3 ± 17.7	22.8	73.0	3.30±2.40	1.59±0.19

the dry seasons than in the wet seasons. Monthly average AAE values ranged from 1.39-1.59, lower than the 1.5-1.9 range seen at rural Mt Mugogo (DeWitt et al., 2019); this could indicate a greater contribution from vehicular sources to ambient BC in urban Kigali. Higher AAE (>1.8) values were almost always only seen at BC concentrations below 5 μg/m³ (Figure S7), while higher BC concentrations showed AAE values around 1.4 or less, which suggests that fossil fuel combustion is associated with higher BC levels, but the background BC is dominated by biomass burning.

Diurnal profiles of $PM_{2.5}$ and BC

Figure 4 shows the average diurnal pattern of $PM_{2.5}$ and BC for the five seasons (four for BC) during the study period for which we have a large number of measurements. Only working day data are shown for visual clarity; a similar pattern is observed for the non-working day measurements. As noted earlier, $PM_{2.5}$ concentrations are substantially higher in the dry seasons than in the wet seasons. A morning peak (likely related to traffic) is observed between 8 AM-10 AM local time for all seasons except MAM 2017. Concentrations then fall during the day (as the boundary layer height increases) before rising back up in the evening, likely a combination of evening emissions and lower boundary layer heights. The MAM 2017 night-time highs (and to some extent the JJAS night-time highs) remain at that level until the morning traffic peak. For the other three seasons, the morning traffic peak is higher than the night-time high values. While the BC concentrations follow a similar diurnal profile, in all seasons the maxima occur during the morning rush hours, with concentrations lowest in the afternoon. In further contrast to the $PM_{2.5}$ dry/wet seasonal differences, the wet and dry season BC concentrations are much closer to each other, though dry season BC is still higher.

The highest $PM_{2.5}$ concentrations are observed at night-time during MAM 2017 and JJAS, with average $PM_{2.5}$ around 80 μg/m³ between 8 PM-12 AM in the wet season (MAM 2017) and around 100 μg/m³ between 7 PM-11 PM in the dry season (JJAS). In contrast, the highest average $PM_{2.5}$ levels in other seasons were observed during the morning rush hour - around 60 μg/m³ between 7 AM-10 AM in ON, around 90 μg/m³ between 8 AM-10 AM in DJF, and around 45 μg/m³ between 8 AM-10 AM in MAM 2018. The BC maxima all occur during the morning rush hour between 7 AM-8 AM except for DJF, when levels are marginally higher between 8 AM-9 AM than during the preceding hour.

The morning and evening peaks at similar times for $PM_{2.5}$ and BC indicate that these PM peaks are related to combustion emissions. Figure 5 shows the diurnal variation in AAE during the dry and wet seasons. In both cases, AAE is lower between 6 AM to 9 AM, when BC mass concentrations are higher; the lowest median AAE is 1.4 between 7 AM-8 AM during the dry season. However, the night-time peaks in $PM_{2.5}$ and BC are associated with higher AAE values, when hourly medians approach 1.6-1.7. These differences suggest that while the morning BC and $PM_{2.5}$peaks are mostly associated with fossil-fuel vehicular emissions, the night-time BC and $PM_{2.5}$ peaks are additionally influenced by domestic biofuel use. However, compared to previous studies showing AAE values of ~1.1±0.3 for fossil fuel BC-dominated pollution, the AAE values observed in Kigali are often higher, suggesting that there is always some biomass burning influence (associated with BrC).

Local and regional contributions to $PM_{2.5}$ and BC in Kigali

We use OBM (Diamantopoulou et al., 2016; Pikridas et al., 2013) to estimate the regional and local contributions to ambient $PM_{2.5}$in Kigali. During the dry season, a higher regional background is likely, as transported pollution may not be rained out. The background can include regional biomass burning, dust, and other upwind emissions as well as secondary $PM_{2.5}$. The OBM assumes that the minimum value of a seasonal diurnal profile based on hourly measurements is the seasonal average regional contribution. Then, the seasonal average local contribution is the difference between the seasonal average ambient $PM_{2.5}$ and the seasonal average regional contribution. These estimates of regional contribution assume the ambient concentrations at the minima are entirely regional, but there may be local vehicular and domestic biofuel emissions all day. Thus, the results are the upper bound of the regional contribution and the lower bound for the local contribution.

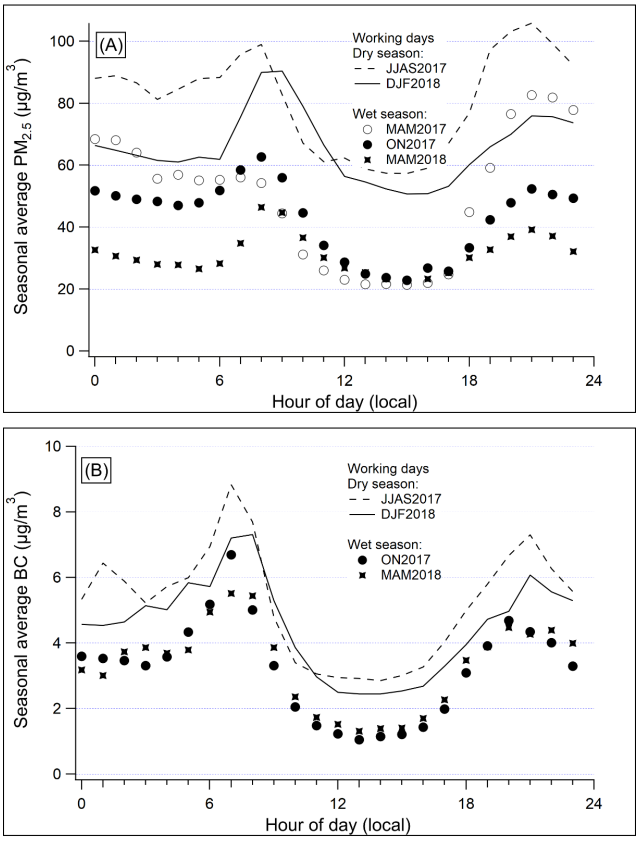

Figure 4: *Average diurnal patterns of (A) $PM_{2.5}$ and (B) BC for each season over the course of this study. Data restricted to working days, with similar patterns observed for non-working days.*

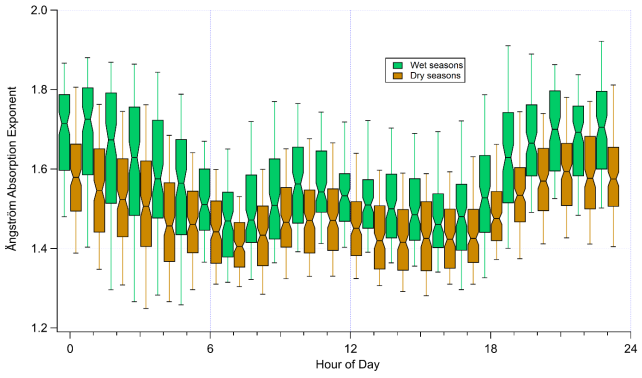

Figure 5: *Diurnal variations in Angstrom Absorption Exponent (AAE) during the dry and wet seasons suggest vehicular emissions and domestic biofuel use contribute to ambient air pollution at different times of the day. The boxes show the IQR (25th and 75th percentiles of data); the notch shows the median; whiskers indicate the 9th and 91st percentiles. The corresponding hourly boxes for the two types of seasons are offset for visual clarity.*

Table 3 summarizes the seasonal statistics required to calculate the average regional contribution to Kigali ambient $PM_{2.5}$ for each season. The minimum average $PM_{2.5}$ values were observed between 3-4 PM local time in all seasons. About half the ambient $PM_{2.5}$ in Kigali appears to be from regional sources in the wet season, and the regional share rises to over 70% in the dry seasons. Put another way, local sources contribute at least half the observed $PM_{2.5}$ during the 2017 wet seasons, and one-fourth of ambient $PM_{2.5}$ during the two dry seasons. The MAM 2018 wet season appears to be an anomaly compared to the two other wet seasons, but in terms of absolute $PM_{2.5}$ concentrations, the regional component is about the same in MAM 2018 as during the 2017 wet seasons.

Following a similar procedure for BC, the regional contribution on working days is 54-55% in the JJAS and DJF dry seasons, but only 33% and 40% in the wet MAM 2018 and ON seasons. The regional contribution to Kigali BC on non-working days is 7-8% higher during JJAS, ON, and MAM 2018, which likely reflects lower local emissions (including car-free Sundays). Overall, local sources are larger contributors to ambient BC than to $PM_{2.5}$ in Kigali, ranging from about half in the dry season to two-thirds in the wet seasons.

The impact of Sunday car-free hours on urban background air pollution

An estimate of the impact of traffic-related air pollution (TRAP) at non-roadside, urban background locations can be obtained by comparing the RAMP and BC measurements on "car-free" Sundays with the measurements on all other Sundays. In this study period, $PM_{2.5}$ (BC) measurements were made on 16 (11) "car-free" Sundays that can be compared to data from 42 (30) regular Sundays. However, in Rwanda, the car-free policy is only in effect on Sunday mornings. The time-resolved data from the RAMPs and BC monitor allows investigation of the benefits of this policy for the specific periods when such policies are in effect.

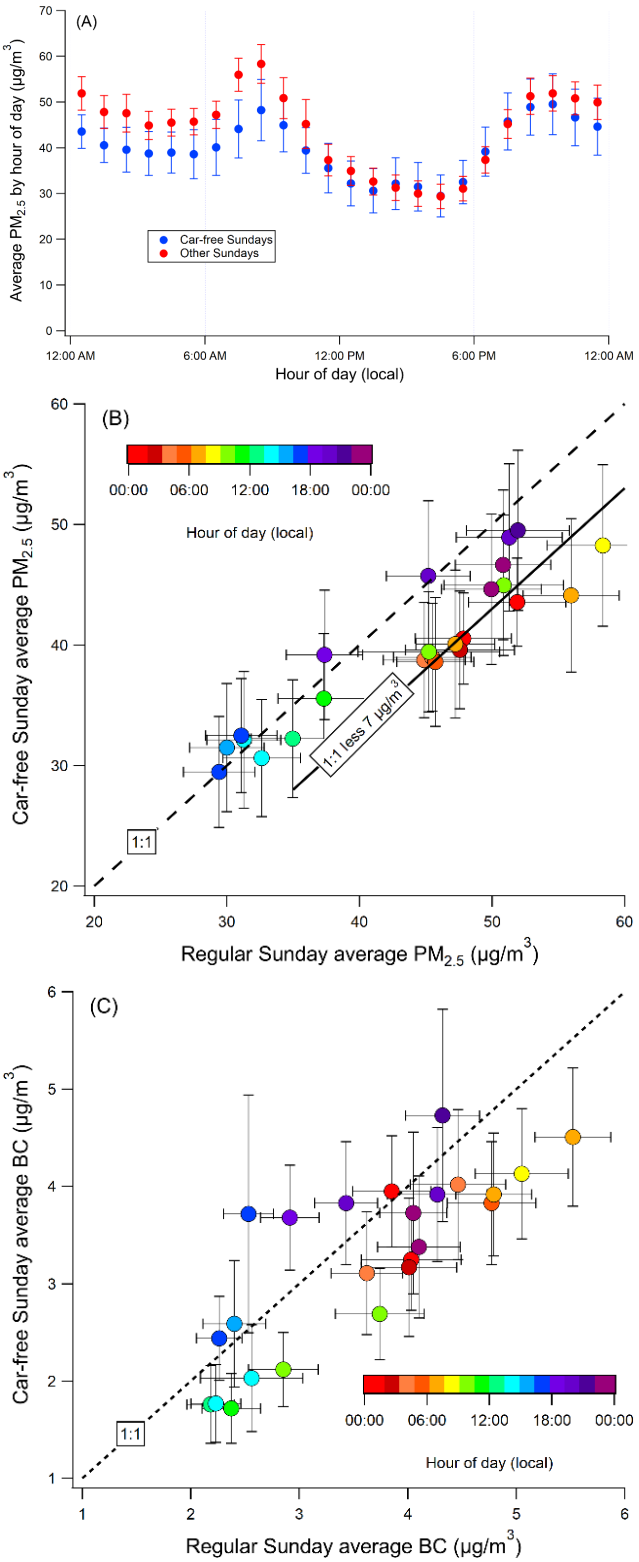

Figure 6: *Impact of limiting vehicular traffic on Sunday mornings in Kigali, Rwanda. Panel (A) shows the average hourly $PM_{2.5}$ measured over 16 car-free Sundays and 42 regular Sundays between April 2017-May 2018. Panel (B) shows the same $PM_{2.5}$ data as a scatter plot. Panel (C) shows a scatter plot of the corresponding BC concentrations. Error bars are the standard error.*

Figure 6 shows a comparison of the hourly average $PM_{2.5}$ between regular Sundays and car-free Sundays. The average hourly $PM_{2.5}$ concentration is lower in the morning period by an average of 7 µg/m³. In the afternoon and evening periods, the diurnal patterns converge, indicating no significant difference in pollution. The reduction in ambient $PM_{2.5}$ is highest between 7 AM-9 AM, when $PM_{2.5}$ on regular Sundays is highest; during these hours the average $PM_{2.5}$ is reduced by 10-12 ug/m³ on car-free Sundays or 20% of $PM_{2.5}$ at the same time on regular Sundays. BC concentrations are lower by about 1 µg/m³ between 7 AM-10 AM on car-free Sundays, which is 18%-28% of the corresponding concentrations on regular Sundays. However, there are also similar differences for pre-7 AM "business-as-usual" times on these Sundays, and so the observed reductions during the morning car-free periods may not be directly attributable to the car-free policy.

Ozone in Kigali and Mugogo

Only 942 hours of RAMP data are available from rural Mt Mugogo for JJAS (dry season) and 881 hours for ON (wet season), compared to the 2,400 and 1,464 hours of O_3 measurements in Kigali for the same seasons. The maximum 8-hour average O_3 value at rural Mugogo as measured by the RAMP using the gRAMP calibration models were 34.4 ppb in dry JJAS and 33.0 ppb in wet ON – similar, unlike the significantly higher O_3 observed in urban Kigali during the dry season (JJAS) compared to the wet season (MAM and ON). However, as discussed previously, the gRAMP model (hybrid-RF) is positively biased at Mt Mugogo during ON. The RCO reference monitor at this site shows a seasonal difference, with maximum 8-hour O_3 concentrations of 40.6 ppb in JJAS and 30.6 ppb in ON (restricting the comparison to common periods with the RAMP data.) This is consistent with previous findings at this site (DeWitt et al., 2019). The RAMP-specific Mugogo QR model, which is not seasonally biased, shows maximum 8-hour O_3 concentrations of 33.9 ppb in JJAS and 26.5 ppb in ON – below the reference values but capturing the seasonality. A closer examination of the data (for common periods) indicates that both the reference monitor and the gRAMP model show the maximum 8-hour O_3 on the same day of the dry season - September 2, 2017. The RAMP O_3 maximum (34.4 ppb) occurs between 8 AM-4 PM, during which time the reference monitor average was 40.3 ppb.

The average 1-hour O_3 concentrations in Kigali during March-November 2017 were 16 ppb in the wet season (MAM 2017 and ON) compared to 22 ppb in the dry season (JJAS). The maximum O_3 values, 66 ppb (1-hour average) and 57 ppb (8-hour average), were observed in the dry season. Previous measurements over Kigali during aircraft takeoff and landings in the MOZAIC campaign (Sauvage et al., 2005) over 1997-2003 also showed significantly higher O_3 concentrations in the dry season in the lower troposphere.

The current US EPA standard for 8-hour average O_3 is 65 ppb and the WHO guideline value for 8-hour average O_3 is 50 ppb. O_3 in Kigali is higher than the WHO guideline on 10 days over our sampling period, of which six are in July. However, the highest 8-hour average occurs on August 30, 2017 between 10 AM-6 PM. All 8-hour averages over 50 ppb occur during the daytime, for 8-hour periods beginning usually at 10 AM or 11 AM.

Conclusions and recommendations for future work

We have presented the results of a long-term ground-based monitoring campaign, the first of its kind in Kigali, Rwanda. Lower-cost and relatively low maintenance RAMP monitors were used for this study, with local verification and correction of sensor calibrations by collocation with reference monitoring during an overlapping campaign and at RCO.

O_3 pollution in Kigali was usually below WHO guidelines, but the 50-ppb threshold could be exceeded in the dry season. Periods of high ozone can be identified using low-cost sensors and a general calibration, though local calibrations improve sensor performance. The RAMP $PM_{2.5}$ (before filter-based correction) correlates strongly with filter-based $PM_{2.5}$ on non-working days when TRAP and other working-day contributions are lower, but the RAMP $PM_{2.5}$ values were still a significant underestimate. These differences suggest that the size distribution of $PM_{2.5}$ in Kigali is quite different from that in Pittsburgh, with substantial contributions from sub-300 nm particles (where low-cost optical sensors are less sensitive). Future studies with lower-cost monitors should include collocated filter-based measurements or short-term intensive studies with aerosol sizing instruments (e.g. a scanning mobility particle sizer, SMPS) to account for such differences.

The Health Effects Institute (2019) (HEI) using data from the Global Burden of Disease Study 2017 estimates that the population-weighted annual average $PM_{2.5}$ in Rwanda was 43 µg/m³ in 2017 (and the same for 2015-2016). Our study found that the study average $PM_{2.5}$ in Kigali is 52±34 µg/m³ at residential or university locations, which can be considered as "urban background" sites. Kalisa et al. (2018) show that pollution levels can be significantly higher at the roadside in Kigali than at the urban background locations where our measurements were made. Air pollution in the low-lying valleys of Kigali can also be higher (Henninger, 2013). RAMP measurements at Musanze in late 2017 showed $PM_{2.5}$ concentrations at this rural site were moderately correlated (r^2 = 0.54) with Kigali $PM_{2.5}$ and about 20% lower (not shown). These results suggest that the HEI population-weighted average for Rwanda and other similar satellite-based estimates could be underestimates that need to be updated with ground-based monitoring, like the campaign presented here.

Three recent studies – this paper, DeWitt et al. (2019), and Kalisa et al. (2018) – have found that air pollution is significant in urban and rural Rwanda, with considerable spatial variability due to local conditions and sources. Hence, future studies with low-cost monitors should include a variety of locations, such as low-lying areas and roadside locations. Our results should be

considered a lower estimate of the $PM_{2.5}$ pollution that Kigali residents are exposed to – which is concerning since the average monthly concentrations in our 16+ month study were above the WHO annual guideline and often over WHO's first interim target.

Long-term monitoring over five seasons shows that differences in $PM_{2.5}$ between working days and non-working days are smaller than the differences between the dry and wet seasons. The average dry season $PM_{2.5}$ levels are about two times the succeeding wet season $PM_{2.5}$ while the comparable seasonal differences in BC are about 40-60%, indicating that transported non-BC (e.g. dust) or low-BC (e.g. forest fires) pollution is important in the dry season. Particulate pollution in Kigali has a distinct diurnal profile, as the morning rush-hour usually results in the maximum BC and $PM_{2.5}$ due to traffic pollution, and minima occur in the afternoons. The higher AAE values associated with the overnight BC and $PM_{2.5}$ peaks suggest that domestic biofuel use could also be a significant contributor to overnight air pollution.

Overall, local sources could contribute half the $PM_{2.5}$ and two-thirds of the BC in the wet seasons in Kigali, which means that local pollution control policies can significantly improve Kigali's air quality. In MAM 2018, heavier local rainfall may have helped reduce ambient $PM_{2.5}$ levels, though the January-July 2018 average $PM_{2.5}$ was 43.7 $\mu g/m^3$, still above the WHO's first interim target for annual average $PM_{2.5}$. New pollution control policies are being implemented in Rwanda, including doubling the number of car-free Sundays and increased duties on older imported used cars. Continued monitoring is essential to evaluate the impact of these policies.

Air pollution in Kigali is also influenced by substantial regional contributions especially in the dry season; for example, the dry season daily minimum $PM_{2.5}$ was over 50 $\mu g/m^3$, or over 70% of average $PM_{2.5}$. About half the BC during the dry season also appears to be regional, which suggests a significant biomass burning influence in the regional/background contribution in line with DeWitt et al. (2019). Hence, controls on regional biomass burning are essential to reducing $PM_{2.5}$ concentrations in Kigali. Studies with more advanced instrumentation (such as an AMS) would help better quantify the time-resolved contribution of different local and regional sources to air pollution in Rwanda, which in turn would help identify additional air quality interventions.

This study was carried out in collaboration with the University of Rwanda and RCO (co-authors on this paper) and with the support of local residents who hosted the RAMPs. Towards the end of this study, the Rwanda Environmental Management Authority (REMA) acquired a reference monitoring station as well as eight RAMPs to establish their own air quality monitoring network. Low-cost monitors can significantly reduce the costs of air quality monitoring for developing countries, but local support and buy-in are keys to success.

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Author contributions

Conceptualisation: RS, ALR, PJ; methodology: RS, JG, HLD; data collection: ASK, VB, SG, CS, NJW, EK; data analysis and validation: RS, CM, ASK, SG, HL, EK; writing/initial draft: RS; writing/revisions: all authors; student supervision: JG, RS, PJ, PA; project leadership and management: RS; funding acquisition: ALR, PJ, RS

Data availability

The RAMP data (gRAMP-based gas concentrations and "BAM-equivalent" $PM_{2.5}$ mass concentrations, without filter-based correction) and BC data are available at https://doi.org/10.6084/m9.figshare.8074436.v1

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Appendix

Further investigation of the performance of Pittsburgh gRAMP calibration models and comparison with local "Mugogo" models

We further explore the performance of the Pittsburgh hybrid RF gRAMP model by comparing it with the Mugogo QR model, which had the best overall performance for O_3 among the Mugogo models. Figure S2 shows scatter plots of the RAMP-reported concentrations (as 8-hour running averages) for the two calibration approaches against the reference monitor, separated into the dry (June-July-August-September, JJAS) and wet (October-November, ON) seasons. However, the gRAMP model is positively biased in ON, with most of the data above the 1:1 line and clustered around the +30% line. In other words, the gRAMP model overpredicts O_3 during the wet season. The Mugogo model (panels B and D) performs better (expected since it was trained on just under half of the underlying 15-minute dataset), yielding O_3 concentrations clustered around the 1:1 line.

RAMP $PM_{2.5}$ data compared to filter-based 24-hour "reference" concentrations

For local calibration verification, we compare data from RAMP #140 (deployed in Gacuriro) with the filter-based 24-hour $PM_{2.5}$ concentrations reported by Kalisa et al. (2018) for the UR-CST site (about 5 km from Gacuriro) in April 2017 (wet season). Kalisa et al. (2018) reported that in Kigali, workday $PM_{2.5}$ was almost 50% higher than non-workday $PM_{2.5}$. The RAMP and filter-based $PM_{2.5}$ are strongly correlated (Figure S3) on eight out of ten non-workdays, with a correlation coefficient (r^2) of 0.77 and a slope of 1.39±0.06 (forced through the origin). On April 2 (Sunday), there was construction activity on the CST site close to the filter sampler, which could explain high PM levels that would not be seen in Gacuriro. It is not clear what was different about April 15 (Saturday), when the filter-based $PM_{2.5}$ was comparable to or higher than the highest workday PM levels. In this paper, all weekend RAMP $PM_{2.5}$ values are scaled upwards by 1.39. The comparison of working day data between filter-based $PM_{2.5}$ and the RAMP $PM_{2.5}$ is worse (r^2 of 0.08); this could reflect local variations on specific days as the sites are 5 km apart. We use the slope (1.69±0.18) of a fit forced through zero to scale up all weekday RAMP $PM_{2.5}$ data. Due to RAMP malfunctions, no RAMP data were collected for the dry season days (June 15-29) when Kalisa and co-workers collected filter-based measurements, so the wet season scaling factors are used for all RAMP data.

Reference

Kalisa, E., Nagato, E.G., Bizuru, E., Lee, K.C., Tang, N., Pointing, S.B., Hayakawa, K., Archer, S.D.J., Lacap-Bugler, D.C., 2018. Characterization and Risk Assessment of Atmospheric $PM_{2.5}$ and PM_{10} Particulate-Bound PAHs and NPAHs in Rwanda, Central-East Africa. Environ. Sci. Technol. 52, 12179–12187. https://doi.org/10.1021/acs.est.8b03219

Figure S1: *The RAMPs and the external Met-One NPMs in the lab (left) and a unit deployed in Kigali, Rwanda (right).*

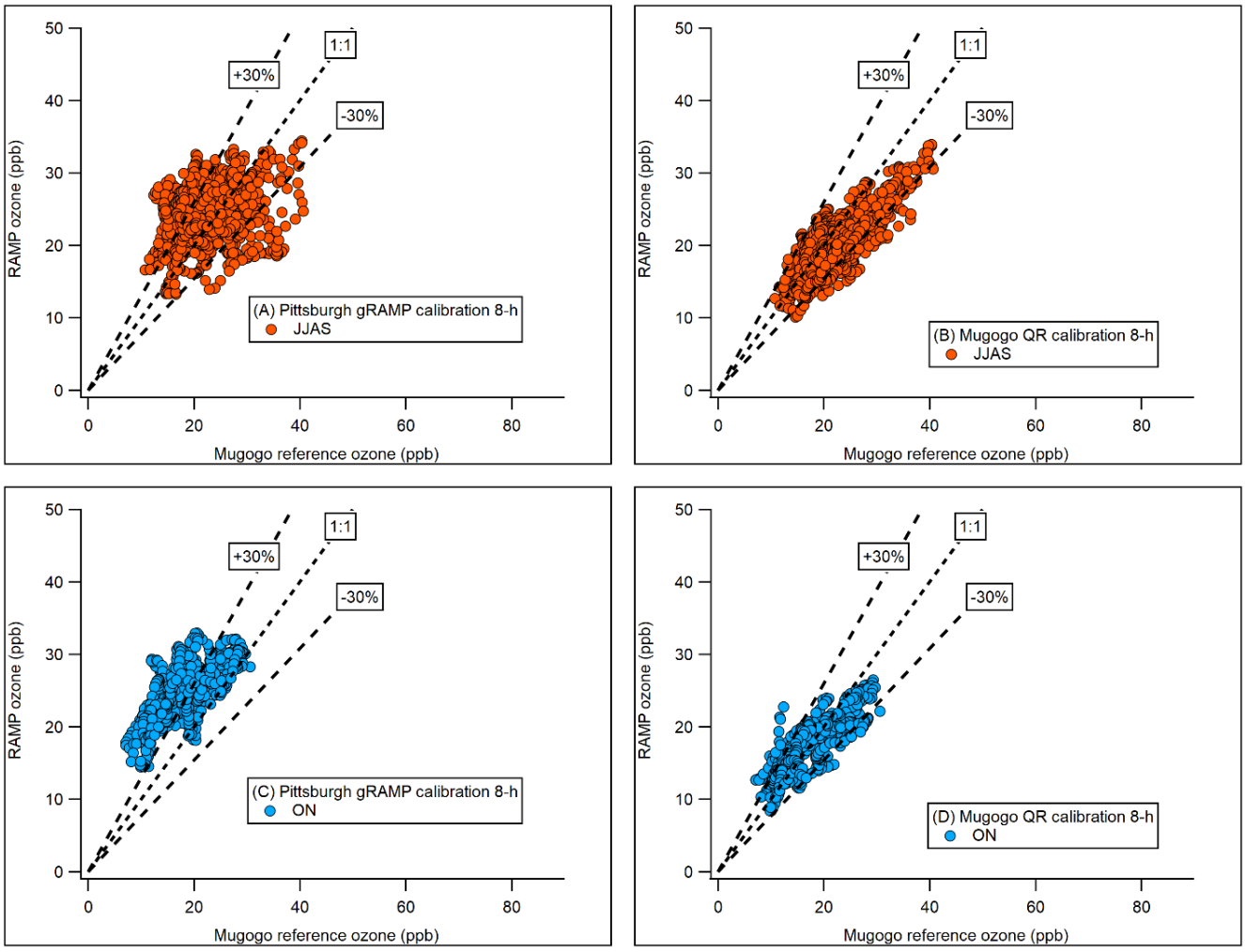

Figure S2: *Comparison of RAMP calibration models for ozone with reference monitor data at the Mt Mugogo Climate Observatory. The "Pittsburgh gRAMP" model (A,C) is developed on collocations in Pittsburgh, PA, USA. The "Mugogo QR" model (B,D) is based on a collocation with the reference monitor at Mugogo; the data shown here includes both training (28 days) and testing (32 days) data. JJAS (A,B) is the dry season and ON (C,D) is the wet season.*

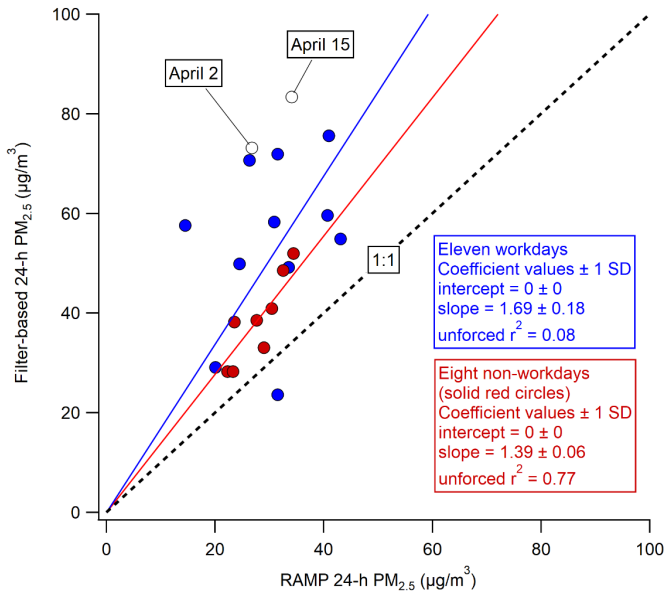

Figure S3: *Developing scaling factors for optical RAMP $PM_{2.5}$ measurements by comparison with filter-based 24-hour $PM_{2.5}$ measurements (reported by Kalisa et al. 2018). Solid blue circles indicate workdays. Solid red circles show eight non-workdays that are used to develop the non-workday scaling factor, which excludes two non-workdays that experienced significantly higher filter-based $PM_{2.5}$ than even workdays (shown by the open circles.) The filter measurements were conducted at the University of Rwanda campus, while these RAMP measurements were conducted about 5 km away in the Gacuriro neighborhood.*

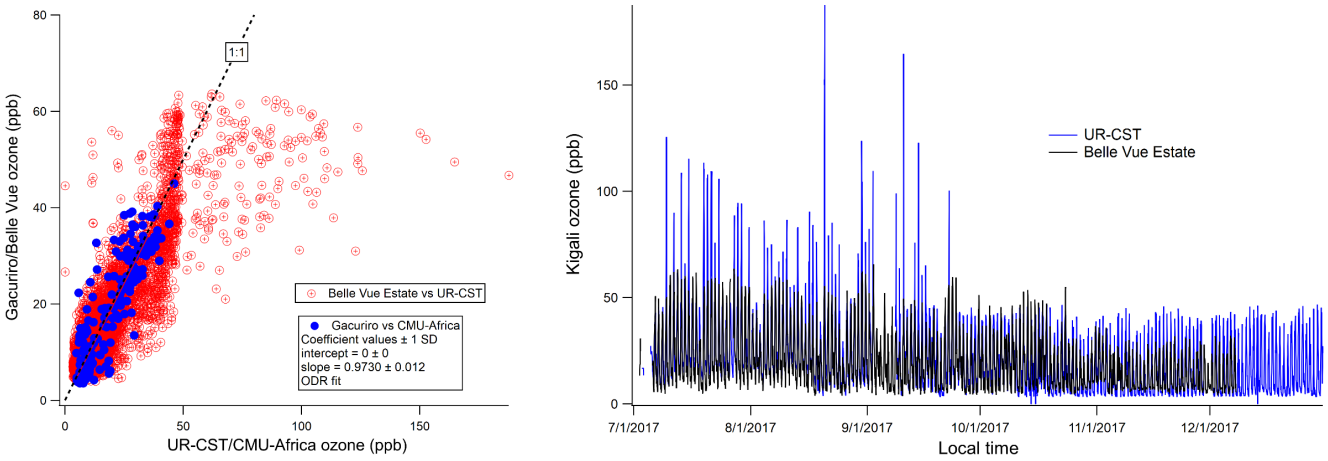

Figure S4: *Ozone measurements in Kigali over the course of this study. (Left) Scatter plot shows most of the paired measurements across the city agree with each other. (Right) However, there are periods when the UR-CST O_3 is much higher than at Belle Vue estate.*

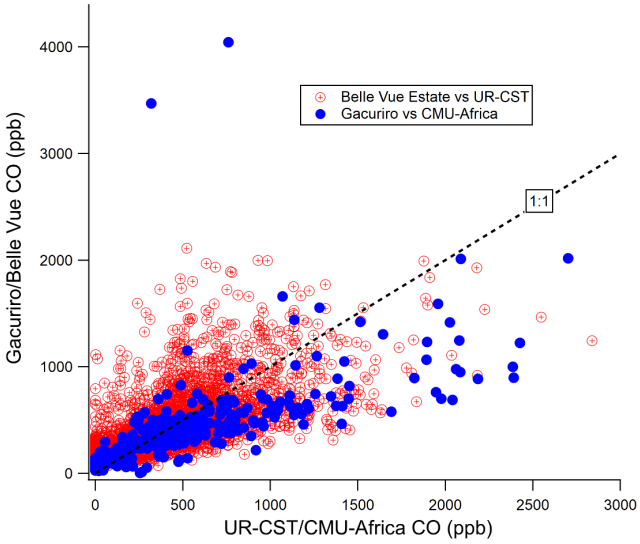

Figure S5: *Intraurban variability of 1-hour CO concentrations across Kigali.*

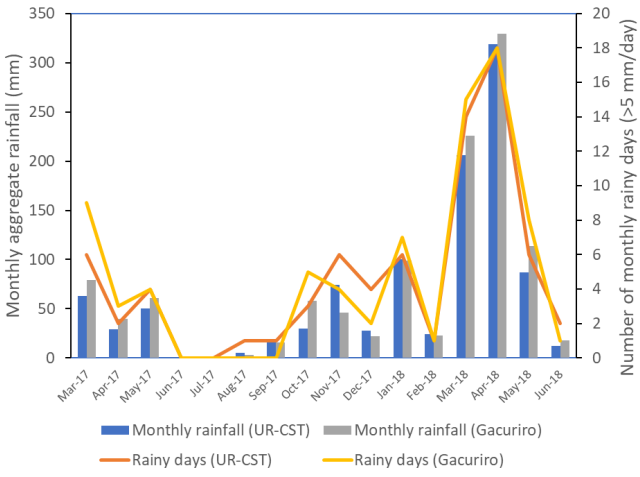

Figure S6: *Spatially-resolved rainfall data using the ENACTS-Rwanda methodology, from the Rwanda Meteorological Agency. MAM 2018 saw significantly more rainfall and more rainy days than other periods.*

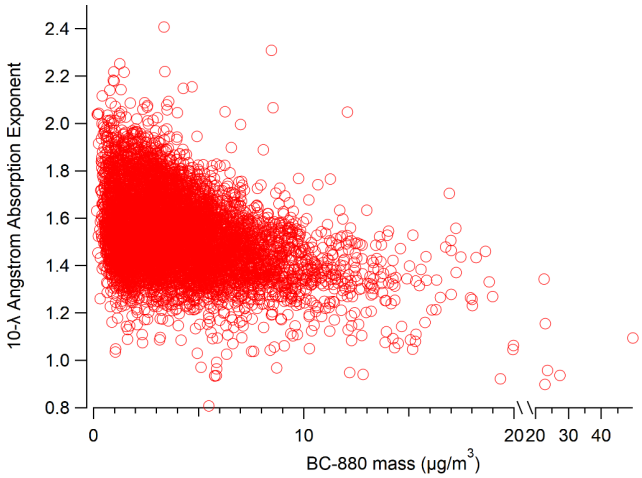

Figure S7: *Higher BC concentrations are associated with lower AAE values.*