Again, the authors thank the reviewers for their time and assistance with this manuscript. We have followed the majority of their suggestions and believe that their suggestions have certainly improved the manuscript. Where we have not followed suggestions, we hope that we offer our reasoning and clarify the issues that the reviewers may have had (both in our answers and by making changes within the text to clarify our meanings).
We offer our responses on a point-by-point basis below. We have italicized and bolded our responses. Please also see our marked-up changes in the paper following.

Response 1:

ACPD 2018
Seasonal and diurnal variability in air pollutants and short-lived climate forcers measured at the Rwanda Climate Observatory
By DeWitt et al.
Upon second review of the manuscript, the manuscript has been improved, yet still requires major revision for publication in ACP.
Comments in green are unaddressed comments from the prevision reviewer comment report and would be important to address before publication.
Reviewer general comments:
It is difficult for a mountain site to inform on the air quality in Kigali as well as on mitigation efforts within Rwanda, and even more so on the comparison between RCO and country averages. Yet this data set is important and merits publication. The story surrounding this data still needs work.
The discussion of PM2.5 in general should be removed as the study focuses on BC, O3 and to some extent CO. These pollutants should also be added the title.
As stated in the last author response, the authors still disagree: while BC is not the same as PM2.5, acknowledging a potential dual-season impact of biomass burning (both north and south biomass burning seasons) for equatorial East Africa PM (and BC IS PM) is part of the larger story. This is also seen with the tracing of BC and FRP later on in the paper.
When and how often is RCO within the boundary layer compared to in the free troposphere? Line 134 does not adequately answer this issue. See the following references:
We did not have any measurements of into/out of free troposphere (balloon soundings, etc) and there are no high-resolution meteorological models in Rwanda, specifically, that would allow us to answer this specifically with any more certainty. We have done what we can with the available models and data without developing our own, complex model. Places such as central Europe have more met stations per area and higher resolution met models; these are still under development in Rwanda (though, there was a funded project for the UK met office to help Rwanda to do just this!!, so hopefully any future papers from the RCO team will better answer this question).
Nyeki, S., Li, F., Weingartner, E., Streit, N., Colbeck, I., Gäggeler, H. W., and Baltensperger, U.: The background aerosol size distribution in the free troposphere:

an analysis of the annual cycle at a high-alpine site, J. Geophys. Res., 103, 31749–31761, https://doi.org/10.1029/1998JD200029, 1998.

Zanis, P., Ganser, A., Zellweger, C., Henne, S., Steinbacher, M., and Staehelin, J.: Seasonal variability of measured ozone produc- tion efficiencies in the lower free troposphere of Central Europe, Atmos. Chem. Phys., 7, 223–236, https://doi.org/10.5194/acp-7-223-2007, 2007.

Zellweger, C., Forrer, J., Hofer, P., Nyeki, S., Schwarzenbach, B., Weingartner, E., Ammann, M., and Baltensperger, U.: Par- titioning of reactive nitrogen (NOy) and dependence on me- teorological conditions in the lower free troposphere, Atmos. Chem. Phys., 3, 779–796, https://doi.org/10.5194/acp-3-779- 2003, 2003.

Ozone chemistry, references still required:

We have added, as well as a couple on wildfires and ozone. However, Houston is a special ozone case (highly reactive VOCs plus NOx from the Houston ship channel driving a lot of the ozone formation, or at least in the years that that particular article was published) and Toronto is also an urban environment. Both have very different meteorology and emissions compared to Rwanda. Ozone and biomass burning have been linked in other (cited in the article) works in sub-Saharan Africa, which would be more relevant for this paper.

Baier, B. C., Brune, W. H., Lefer, B. L., Miller, D. O. and Martins, D. K.: Direct ozone production rate measurements and their use in assessing ozone source and receptor regions for Houston in 2013, Atmos. Environ., 114(Journal Article), 83–91, doi:10.1016/j.atmosenv.2015.05.033, 2015.

Geddes, J. A., Murphy, J. G. and Wang, D. K.: Long term changes in nitrogen oxides and volatile organic compounds in Toronto and the challenges facing local ozone control, Atmos. Environ., 43(21), 3407–3415, doi:10.1016/j.atmosenv.2009.03.053, 2009.

Monks, P. S., Archibald, A. T., Colette, A., Cooper, O., Coyle, M., Derwent, R., Fowler, D., Granier, C., Law, K. S., Mills, G. E., Stevenson, D. S., Tarasova, O., Thouret, V., von Schneidemesser, E., Sommariva, R., Wild, O. and Williams, M. L.: Tropospheric ozone and its precursors from the urban to the global scale from air quality to short-lived climate forcer, Atmos Chem Phys, 15(15), 8889–8973, doi:10.5194/acp-15-8889-2015, 2015.

Reviewer specific comments:
Title:
– I would again encourage the authors to specify which "air pollutants and short-lived climate forcers" they studied. I really think a specific title would benefit the visibility of the article. For example, "**Seasonal and diurnal variability in O3, BC and CO measured at the Rwanda Climate Observatory"**
Changed as suggested.

Abstract:
– Line 15: replace "air pollution is still largely unstudied in sub-Saharan Africa" to "Air pollution is understudied in sub-Saharan Africa".

Changed.

– Avoid parentheses. The information within them is important and should be part of the sentences.

changed

– Season numbers are confusing. The seasons are much clearer now in the text, but it would be useful to use the month letters here in the abstract for clarity. The idea here is to make the abstract accessible to an international readership.

Added month letters

– Line 23: rewrite for clarity. For example, "Rwanda is […] East Africa with less than 20% urbanization but is currently undergoing rapid development."

– Line 26: give standard deviation on the 5 microg/m3 value.

– Line 36: are there only one or two rainy season(s)?

Two rainy seasons. There are two, but they are both simply described as rainy, so are often referred to collectively as 'rainy season'. However, the authors have changed to plural for clarity.

– Line 41: "that deserves immediate attention" – I would encourage the authors to either specify how or to omit this comment.
Omitted

Introduction:
The introduction was streamlined and reads much better. However Figure 1 needs 2-3 more sentences of description to clarify the data points. I don't understand the x-axis. Are there really 1600 papers reporting an annual PM2.5 exposure (and why not concentration?) of 15 microgram/m3? I recommend using a few references to make the same point. For example compare O3 and/or BC Paris, London, Beijing, LA, Mumbai, Johannesburg and add the new Rwanda data. Don't use PM2.5 if there is no PM2.5 data in the paper.

The figure, as described in the figure caption, is the World Bank annual PM2.5 exposure data/model/etc (which is the only really 'global' dataset of 'air pollution', which is why it was used) in a country versus the number of papers when searching the web of science for 'air pollution' + 'that country name'.

For increased clarity, we have changed this sentence to say: "Despite this, little scientific research has been published on air quality in Africa, which can be approximated by the number of paper results from the search terms 'air pollution + country name.' World Bank collected data and model approximations estimate higher PM2.5 exposure in African versus European Countries (Figure 1). Again, we are setting up a problem of potential air quality issues + Africa, and PM2.5 is the most widely available data, so we are using it—in a way we feel is fair—as a proxy for air quality.

- Line 53: update reference to a more up-to-date reference.

Updated

- Line 55: astral should be austral

Changed

Methods:

– Table 1: needs synthesized information. For example, additional columns could include minimum and maximum concentrations observed by each instrument, annual or seasonal average. Table 1 should be a quick reference for the community to see how this station compares to other mountain sites.

Added

– I recommend that the authors add a data processing section in their methods. How did they quality control the data?

This is already in the text, please see the instrumentation section. We have added a few more details as well. Data processing sections are not often included in manuscripts of this nature and data processing was not different than any standard field campaign or long-running measurement.

Results and Discussion:

– Line 175-176: were the generator spikes often? The generator's influence could be substantially impacting the data, and it would be important to convince the reader of the trustworthiness of the data set.

No, the site was also part of the Rwanda broadcasting authority so power connectivity was generally good throughout the year, the generator was over 500 m away from the site as stated in the text and only one generator, and again, we removed any spikes. One generator, very occasionally active, 500 m from the station would not affect the conclusions from this data set.

– Black carbon concentrations are high enough to be reported in microg/m3.

We will remain with ng/m3, as both are acceptable units and we do not feel regenerating the graphs are necessary if the units used are clearly labelled.

– Figure 2: why is temperature constant at the beginning of the measurement period? Data quality control is incomplete still for BC (straight grey line around 9/1/16). Are the lighter colour traces averages, running averages, 8-h running averages? extrapolations? Specify.

We specified in the figure caption that they are daily averages (as in, one calendar day). We have added that they are 24 hours averages. I also do not see where the temperature is constant (have checked)? Rwanda has fairly consistent temperatures so it might be hard to see small differences on the scale. Again, there is simple a connection issue with graphing (when no data, a line connects the two existing data points), NOT data quality control issues, as the authors stated in the previous iteration of the author report.

– Figure 3: Absolute values are more meaningful to highlight an air quality problem. The authors can plot all traces on the same graph and have 3 different y-axis. The authors must be consistent in their graphing – each graph has different types of error representations and leads to confusion. Choose one and use throughout each panel for clarity.

We respectfully wish to remain with normalized graphs, as we feel they highlight the changes by season nicely and again, there are absolute values in other graphs. 3 different y-axes would be quite busy for this three paneled figure. Additionally, this way you can directly see how each pollutant changes in relation to the seasons and to each other in terms of overall change (magnitude of change). All error in this panel is 95% confidence interval and shaded, there is no difference in this, just in the magnitude of this interval per averaging unit.

– Figure 4: include a graph for RCO to effectively compare the three sites. Elaborate on the significant different in BC concentrations in DJF between Kampala, Addis and RCO.

Nonetheless, comparing two urban sites with RCO is not so meaningful since they are affected by local sources to highly different extents. The authors could also remove this graph entirely.

As we answered in the last iteration of this author response, we have decided to keep RCO removed, as again, as stated also by the reviewer several times, BC and PM is not directly comparable but one may give information about the other. This graph is to show that there may be similarities between seasonal BC and/or PM2.5 in equatorial Africa, due to experiencing transport from the two biomass burning seasons during the two dry seasons in equatorial Africa (note that Kampala is in the equatorial East Africa region, while Addis Ababa is not). We have also moved this graph, and we discuss further down below. We do not understand where the reviewer would like us to elaborate: we do elaborate on the differences in this section. Kamapla is close to Rwanda, and as the FRP, BC, and hysplit backtrajectories show, plus the o3 data in Brazzaville (also equatorial Africa, though west) show a duel transport from both the northern and southern biomass burning seasons.

– Figure 5: MODIS data should not be presented in the rainbow color scale. I recommend using **a two colour bar so that it is clearer whether the FRP is low or high (like the blue-red color bar).** *The authors answer that they have changed the color, but the submission version likely has a mistake as it still contains the rainbow color scale.* ***Ack, wrong picture added, this was changed****.* The excellent match between FRP and BC concentrations is highly significant and should be further discussed in the paper. This comparison is striking! ***This goes along with the PM discussion later on (please see further down for a larger explanation and changes), but we are trying to drive home the fact that BC increases in equatorial east Africa in both biomass burning seasons, despite the changes in location. We have added this sentence to drive home the point: "This suggests that transport from regional biomass burning has a twice-yearly effect on BC concentrations in Rwanda, despite the different locations of the biomass burning in sub-Saharan Africa."***

This also feeds into the PM discussion above, we hope the changes make the reason for including PM more clear and satisfy the reviewer's concerns.

– Figure 11: I have issues with the meaning of this figure. The comparison is problematic. Rwanda's bar on the figure is from ONE SINGLE SITE, a regional site **whereas the comparison to other countries is an average of a MANY**, and does not add value to the context. BC data could be compared to other background and mountain sites – but cannot between countries. Furthermore, if the authors want to highlight a pollution problem, **then a better approach could include highlighting maximum daily pollutant levels (and/or exceedances) instead of averages from different sites.**

The two reviewers disagreed about this figure, but the authors have decided to remove. Highlighting daily pollutant levels from multiple sites is beyond the scope of this paper.

– Figure 10: it is difficult to compare between panels. Place all graphs on one x-axis labelled Day 1-Day4 and then colour code with the different dates. Could the authors use microg/m3 instead of ng/m3?

Changed onto the same axes. Left as ng/m3 (see reasoning above).

– Section 3.3.3 – the discussion about PM2.5 in Addis Ababa and Kampala are outside the scope of the study. This analysis does not bring more understanding to Rwanda, particularly since no PM2.5 are being reported in this study.

The discussion is to highlight that equatorial Africa may have pollution from both biomass burning seasons (an issue because it is heavily populated) while other areas of Africa might not. This goes along with the ozone data from the MOZAIC and SAFARI campaigns that are discussed in the paper. This also follows the FRP-BC agreement the reviewer wishes we highlighted more in their review. For these reasons, the authors believe that this data IS relevant to discuss. However, as the reviewer has pointed out, this may not be well set up in the paper as-is. We have now changed the title of 'global and East African etc section' to '**Potential Twice-Yearly Influence Biomass Burning in equatorial Africa*****'. By highlighting that Addis Ababa does NOT have a double peak in PM2.5, but Kampala does (and at the same time as Rwanda) it does suggest that a larger regional trend may be present. We have revised this section 3.3.3 to hopefully make this point more clearly to the reviewer and the other readers, including trying to tie O3 in to this story.***

– Line 467 – PM3.5 ;)

Changed.

– The MOZAIC campaign discussion could be better places under an ozone section.
The MOZAIC campaign is showing seasonal trends in ozone (that take place twice per year), which is relevant for the seasonal trend section. We have tried to tie it into the overall picture of the paper more clearly here, however.

Conclusion:
Point 1: The use of PM2.5 in this study is inconsistent. There is no PM2.5 data presented, yet Figure 1 and Figure 10 focus on PM2.5. The PM2.5 discussion is therefore misleading. Why not stick to BC discussion only? It would streamline the discussion.
Removed PM2.5 from this conclusion (but see above response for why we believe it should remain in the main text).
Point 2: Higher emissions do not equate to higher ozone.
We removed this point as it went into the first point and was redundant.
Point 3: excellent
Point 5: Care should be taken in using this first-ever data set for Rwanda to speculate on improving emission inventories and mitigation issues.
Very fair point, changed to: "The measurements we have provided in this study will be useful in advancing atmospheric science in Rwanda, which has limited long-term and in-situ atmospheric data."
Reviewer technical corrections:
There are important changes that the authors can make to improve the quality of the writing and thus the efficiency of their communication. I would like to point out the following grammar and syntax recurring issues in the manuscript:
1. The word "this" should be followed by a noun. "Despite this," and "This is" is incorrect/inaccurate syntax in scientific writing (see line 47, line 218, line 538***).***
Changed these three lines, did a search and these were the only instances.
2. Sentences longer than 2-3 lines of text need to be revised for syntax and conciseness. (see lines 94-99)
Shortened sentences where possible.

Reviewer 2:

Suggestions for revision or reasons for rejection (will be published if the paper is accepted for final publication)

The authors have done a very good job of improving the flow of the manuscript. As I stated in my original review, this is an important dataset in an area with few measurements and thus adds important new information. I do believe the authors have improved the manuscript and would recommend for acceptance after a few minor comments are addressed to improve clarity.

Line 24, I would recommend adding either regional or local to this sentence to improve clarity, "…which coincide with the two (regional or local?) biomass burning

seasons."
Added regional

Line 55, The southern Africa burning season is more than just JJA, but extends until October in southern regions (austral spring). This can be seen in Figure 5 and previous literature.
Changed

Line 83, these data are not data about climate change really, but are of SLCP. This is similar for line 581. These measurements are not of impact of climate change really. I would recommend removing that or explaining the linkages better.
This is really setting up the Rwanda Climate Observatory itself, as this is the first paper on it.

Line 96, this sentence is incomplete. I believe it should be "Rwanda has…"
The authors could not determine what sentence the reviewer was referring to.

Line 198 what is spatial resolution of NCEP reanalysis used? ***Standard 2.5 x 2.5 degrees. Added to the text.***

Line 290, I believe this should be figure 8.
We think now it should be figure 7 with the removal of another figure.

Lines 315-353, this analysis is really interesting. It is inconclusive at the end, but as described in conclusions line 531, points to some research needs. I would recommend that the authors add a sentence or two summarizing this assessment and clarifying that there is still uncertainty of where this high fossil fuel comes from or if it is not being calculated correctly as info is not for local conditions before beginning section 3.3. Reading through I felt it changed abruptly. This inconclusiveness is an interesting and relevant finding and I believe should be highlighted.
The authors have added these sentences:

"However, this inconclusiveness highlights the need for further study, as ascribing a source to local pollution is important. Further work on emissions profiles relevant to sub-Saharan Africa could clarify these issues. Cookstoves, cookfires, agricultural and trash burning, and older diesel combustion emissions are all likely sources of BC and could be targeted by government policy. Understanding the most important source of local pollution is important for developing efficient government policies for air quality."

Line 379, which estimates are you referring to here? Are these estimates for this area in particular? Or similar areas?

Estimates for sub-Saharan Africa, added this clarifying statement in the text.

Line 381, I do not believe that your results show that targeting local emissions could bring measureable decreases in ambient PM or resultant impacts. Your local BC is estimated to be ~1ug/m3. Obviously there are associated PM and PM precursor emissions related to the BC emissions that account for the 1ug/m3, but how much? That last part isn't known as there aren't PM measurements – and thus I think jumping from BC to PM to health impacts is too strong of a statement. I would recommend removing or altering this statement.

Deleted this statement.

Line 400, in order to see the peak in evening emissions in local cooking/generators, the station would have to be within the boundary layer generally. However, Line 389 suggests that ozone peak in evening is from transport above the boundary layer. This is confusing to me. It does appear that ozone is high starting ~20:00 and BC ~19:00. It could be possible that this hour is enough for the station to change from being within the boundary layer to out of it. If this is the case, then I would recommend in this section adding the times that the pollutants peak to clarify "evening".

Unfortunately, we have no real way of knowing exactly when the station is in or out of the boundary layer in a precise hourly manner (low resolution met models, not many met stations at the time of this paper, though that was in the works for Rwanda!, no soundings). Complex topography makes it difficult. We have tried to add times and clarify our findings in this section. The BC peak at 6 pm, we believe, is cooking/generator driven while the higher overall nighttime BC is, we believe, boundary layer driven.

We have added this statement: BC had mid-morning and early evening (~ 6 pm) peaks that coincided with both cooking times and kerosene/generator use times (sunset at 6 pm each night), indicating local influence on BC before the later lowering of the boundary layer.

Line 414, I would recommend adding the exact dates of the case studies when they are first mentioned here.

We have added

Line 424, it seems the peaks in BC in May are all in the morning during cooking times, yet on average (from fig 8) the peak is seen in the evening. I found that interesting. I would recommend adding more time ticks to the legend of this figure. Trying to look for cooking times (which I assume are 6-8am and 6-8pm) it is hard to see without more ticks. Also, I would recommend that the labels of all graphs with time on x-axis are labelled and indicate the convention used (month/day/year) in order to avoid confusion.

Figure 8 has ticks every hour and they are all the same hours labelled, and are all hours of the day? So we are unsure what the reviewer wished for here, as these are all hourly averaged data. We have added a grid

Line 439, I would recommend that the authors add a sentence or two to transition from BC to PM here. Also, I would recommend subscripting 2.5 In PM2.5.

We have added and have subscripted PM2.5

Line 462, as mentioned in previous reviews, the peak seen in Hersey et al (2015) is not due biomass burning. The peak is due to local emissions during winter, which in many sites is from domestic burning. South Africa is impacted by biomass burning more in Aug-Oct (see e.g. SAFARI aerosol papers, Queface et al, 2011; Tesfaye et al, JGR, 2011; Horowitz et al. ACP, 2017, etc.).

Changed (and added extra references)

Line 467, PM3.5 should be PM2.5.***Changed*** Line 482, there was a SAFARI2000 campaign as well, which was larger. I don't think it would change the conclusions here, just that there have been more recent measurements from SAFARI than in 1993 (line 486).

There were not more recent measurements in central/equatorial Africa however (the earlier campaign had measurements in the congo area (brazzaville), while the latter seemed to only go up to Zambia/Zimbabwe). However, we have added a reference to the more-recent SAFARI campaign as it should be included:

(Line 507-520): "More recent measurements were made in a 2000 SAFARI campaign, but not as far north as the previous SAFARI campaign (Otter et al., 2002) and the positioning of the measurements could have also had an effect on O3 seasonality, as southern Africa is more influenced by biomass burning from August-October."

Figure 3, I would recommend adding more labels to the ticks as it is difficult to eye-ball the 3-month dry and wet seasons with other two labels. This makes it more difficult for the reader to easily pick out the differences.

We have added a grid

Figure 12, I would not include the daily WHO recommendations on monthly averages of data as these are two different averaging times. I think this point needs more context in explaining the differences and should be in the text only.

Removed, and also removed the WHO recommendation discussion as it did not flow well with the text and did not add to the discussion at hand.

Seasonal and diurnal variability of O_3, BC, and CO measured at the Rwanda Climate Observatory

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Abstract

Air pollution is understudied in sub-Saharan Africa, resulting in a gap in scientific understanding of emissions, atmospheric processes, and impacts of air pollutants in this region. The Rwanda Climate Observatory, a joint partnership between MIT and the government of Rwanda, has been measuring ambient concentrations of key long-lived greenhouse gases and short-lived climate-forcing pollutants CO_2, CO, CH_4, BC, and O_3 with state-of-the-art instruments on the summit of Mt. Mugogo (1.586° S, 29.566° E, 2590 m above sea level) since May 2015. Rwanda is a small, mountainous, and densely populated country in equatorial East Africa, currently undergoing rapid development but still at less than 20% urbanization. Black carbon concentrations during Rwanda's two dry seasons (DJF and JJA), which coincide with the two regional biomass burning seasons, are higher at Mt. Mugogo than in major European cities with daily (24 hour) during the dry season of around 5 $\mu g\ m^{-3}$ (daily average range from less than 0.1 – over 17 $\mu g\ m^{-3}$ for the entire measurement period). BC baseline concentrations during biomass burning seasons are loosely correlated with fire radiative power data for the region acquired with MODIS satellite instrument. The position and meteorology of Rwanda is such that the emissions transported from both the northern and southern African biomass burning seasons affect BC, CO, and O_3 concentrations in Rwanda. Spectral aerosol absorption measured with a dual-spot Aethalometer varies seasonally due to changes in types of fuel burned and direction of pollution transport to the site. Ozone concentrations peaked during Rwanda's dry seasons (daily measured maximum of 70 ppbv). Understanding and quantification of the percent contributions of regional and local (beyond large-scale biomass) emissions is essential to guide policy in the region. During the rainy seasons, local emitting activities (e.g., cooking, transportation, trash burning) remain steady, regional biomass burning is low, and transport distances are shorter as rainout of pollution occurs regularly. Thus local pollution at Mugogo can be estimated during this time period, and was found to account for up to 35% of annual average BC measured. Our measurements indicate that air pollution is a current and growing problem in equatorial East Africa.

1. Introduction

According to recent data collected and published by the World Bank, particulate air pollution in most African countries is above the annual average guideline values recommended by the World Health Organization (WHO). Despite this, little scientific research has been published on air quality in Africa, which can be approximated by the number of paper results from the search terms 'air pollution + country name.' World Bank collected data and model approximations estimate higher

346 $PM_{2.5}$ exposure in African versus European Countries (Figure 1). The WHO reported in
347 2013 that one in eight premature deaths globally can be linked currently to poor air
348 quality (WHO, 2013), while another, more recent report, showed that these deaths are
349 concentrated in developing countries (World Health Organization, 2016). Black
350 carbon (BC) is one of the major air pollutants emitted from Africa, mainly from
351 biomass burning as it is widespread on the continent during certain seasons. In
352 addition to affecting health, BC contributes to atmospheric heating and thus to climate
353 change (Ramanathan and Carmichael, 2008). Widespread crop fires in northern and
354 southern Africa, prevalent in boreal winter (December-January-February, DJF) and
355 austral winter and part of austral spring (June-July-August, JJA and September-
356 October), respectively, are known to increase aerosol and ozone concentrations in
357 this region and transported molecular and aerosol fire tracers associated with
358 elevated ozone have been measured as far as the Pacific and Indian Oceans (Field et
359 al., 2016; Real et al., 2010).

360 Rwanda is located in the middle of the two major seasonal biomass burning
361 regions of sub-Saharan Africa. Wide-scale biomass burning occurs to the north of
362 Rwanda during December-January-February (DJF) and to the south during June-July-
363 August (JJA). Rwanda's climate may exacerbate fire haze pollution effects, as Rwanda
364 experiences two dry seasons that occur at the same time as these two continental
365 burning seasons, making long range transport with low rainout efficiency likely.
366 Rwanda's prevalent wind direction also changes from northerly (DJF) to southerly
367 (JJA) at the same time as the large-scale biomass burning area shifts from north-
368 central Africa to southern Africa. Increase in incidence and amount of biomass

369 burning is thought to be one consequence of climate change in this region (Niang et
370 al., 2014). Southern Africa's biomass burning is also influenced significantly by human
371 activity, not just the climate (Archibald et al., 2010). Rwanda is positioned to
372 experience both large-scale (transported) haze due to fires and human activities and
373 local, diffuse emissions.

374 In addition to air quality issues, climate change (related to air pollution) may
375 also adversely affect Rwanda. The major pollutants from or ultimately increased by
376 biomass burning (particles, carbon monoxide, ozone) are also known short-lived
377 climate forcers. The main products exported (coffee and tea), the livelihood of the
378 majority of Rwandans (agriculture), and power (currently almost half of Rwanda's
379 power is hydroelectric) are all potentially affected by climate change. These issues
380 are similar across the region. Central Africa is expected to receive increased severe
381 rainstorms, which may lead to erosion and an uptick in vector-borne diseases (Niang
382 et al., 2014). Rwanda's mountainous topography and ubiquitous hillside agriculture
383 makes Rwanda venerable to floods and landslides. However, there is limited on-
384 ground data on air quality and climate change in Africa.

385 In order to advance our scientific understanding of air pollution, climate
386 change, and their impacts in Africa through generation of on-the-ground data, MIT
387 and the government of Rwanda have established the Rwanda Climate Observatory
388 (RCO). The RCO has a goal to measure long-lived greenhouse gases and short-lived
389 climate forcers/pollutants in East Africa. Since May 2015, CH_4, CO, CO_2, O_3, and BC
390 concentrations have been continuously measured, and N_2O measurements were
391 added in February 2017. The RCO is a part of the Advanced Global Atmospheric Gases

Experiment (AGAGE) network, a global network of high-frequency trace greenhouse gas measurements (Prinn et al., 2000), and is the first station of its kind in Africa. Rwanda was chosen as a location due to several factors. These factors include government interest from Rwanda and willingness to take on station maintenance, Rwanda's interest in growing its technical sector, readily available infrastructure in Rwanda to support the project, and a gap in climate data in this area of the world.

Here we present first results on diurnal and seasonal variations in short-lived climate forcers/pollutants related to air quality, focusing on O_3, CO, and BC observed at the RCO. This dataset is unique and unprecedented to the region. Information on the concentrations, sources, and time-dependent concentration variations of these air pollutants is essential in this rapidly changing area of the world. Data will not only advance our understanding of air pollution and climate change in the region but also potentially inform future policies on air pollution with sound science.

2. Experimental Methods: Rwanda Climate Observatory

2.1 Rwanda Climate Observatory Environment

The RCO is located in the Northern Province of Rwanda, near Byangabo on the summit of Mt. Mugogo (1.586° S, 29.566° E, and 2590 m above sea level). Mt. Mugogo is about 70 km (aerial distance) to the north-west from Kigali, the capitol of Rwanda (population of approximately 1 million), 20 km (south-west) from the next major city, Musanze (population of around 100,000), and 60 km north-east from the Lake Kivu region (Gisenyi, Rwanda and Goma, DRC, combined population of approximately 1 million). A dirt road reaches the base of the mountain, about 500 m below the summit

where the RCO is located, and a diesel generator is installed on the road at the base. Inlets were installed on both the roof of the Observatory (10 m above ground level) for O_3 and BC) and on a Rwanda Broadcasting Authority Tower (35 m above ground level) for CO, CO_2 and CH_4. There is a small Rwandan army camp adjacent to the measurement site and a eucalyptus forest and a mix of agricultural fields and scattered rural houses surround the immediate vicinity of the RCO (Figure 2).

The high altitude and remote positioning of Mt. Mugogo allows sampling of regional air masses from throughout East Africa depending on prevailing meteorological conditions, as well as local pollution (as the dense population but low urbanization of Rwanda means that direct human influence is ubiquitous except within the national parks). Kigali and the Lake Kivu region are approximately 1000 m in altitude below the station height and their altitude (~1500 m) can be used as the base of local pollution. The majority of air masses transported to Mugogo originate below 5 km above ground level. Approximately 20% of yearly air masses measured at Mugogo's summit originate from 0-1 km above ground level (certainly within the polluted boundary layer), and approximately 36% below 2 km (potentially within the polluted boundary layer) (from HYSPLIT analysis). During mid-day, Mugogo's summit is likely within the regional polluted boundary layer, but during the later evening it is likely above. Complicating this issue is the network of farms and houses along the mountainside near Mt. Mugogo.

2.2 Instrumentation and Calibration

437 Details on the instruments sampling at the RCO are compiled in Table 1. $PM_{2.5}$
438 BC (particulate matter $_{2.5}$ micrometers in diameter or less) was measured using a
439 Magee Scientific 7-wavelength Aethalometer with dual-spot technology that is able to
440 correct for filter loading artifacts (Drinovec et al., 2015). A cyclone $PM_{2.5}$ impactor was
441 installed on the inlet to remove larger particles and covered with an insect net. Air
442 was passed through a filter once per day to collect blank data and examined to ensure
443 the instrument baseline was correct. If high, the filter was changed and the blank
444 rerun. Flow was calibrated once per year and after major instrument movement and
445 changes, while the optical performance was calibrated with a neutral density filter kit
446 once per year. Data was recorded every minute at a 5 liter per minute (LPM) flow
447 rate and particles were captured on a quartz fiber filter tape. The air stream was not
448 dried and the relatively humidity (RH) was not controlled, which could lead to
449 increased uncertainty during periods of high relative humidity. RH recorded at the
450 station varied by approximately 5% over the day and from 60-85% monthly,
451 depending on the season. The 880 nm channel was used to calculate the concentration
452 of BC but all channels were examined to determine reasonable data (comparing to
453 literature values). Five minute data (not pictured) was used to detect very local
454 pollution and remove influence of short-lived local fires and BC from the generator
455 500 m below the station. Spikes in BC concentrations that lasted for less than 15
456 minute with values higher than 25,000 ng m^{-3} were removed, along with
457 corresponding CO.
458 CO mixing ratios were measured in real-time using a cavity ring-down
459 spectrometer (G2401, Picarro, USA). Sampled, laboratory, and calibration air were

460 dried with a Nafion drier inside an Earth Networks calibration box to increase the
461 accuracy of the Picarro water vapor correction (Welp et al., 2013). Three NOAA-
462 standard calibration tanks were used for calibration spanning normal ambient
463 concentrations and calibrations were performed once per day initially to check for
464 linearity of instrument's response (Gasore and Physics, 2018). An O_3 monitor (T400,
465 Teledyne Advanced Pollution Instrument, USA) was used to measure O_3. Regular
466 checks were performed using internal span and zero O_3 calibrations, and non-passing
467 data was removed. Flow was calibrated two to three times per year.
468 Meteorological data (ambient temperature, relative humidity, pressure, wind
469 speed, wind direction and rainfall) were collected with an automatic weather station
470 (WXT520, Vaisala, Finland). The weather station was attached to a fixed, hinged arm
471 35 m above ground level and connected to the communications tower, level with the
472 $CO/CO_2/CH_4$ inlet, with a 2 m clearance from the tower. The weather station was
473 calibrated when delivered and recalibrated during repairs (once during the two year
474 measurement period).
475 In addition to the described instrument checks and data quality control
476 procedures, station technicians visited the station once daily (except Sundays) and
477 performed visual checks of all instruments except the meteorological station, which
478 was examined once per quarter manually by climbing the tower. They also notified
479 the station chief scientist immediately of any issues (instrument warnings, generator
480 issues, data coverage outages) and worked to address these issues.
481 ***3. Results and Discussion***
482 **3.1 Seasonal Variation in BC, CO, and O_3**

Figure 3 shows a summary of the data, including daily and 15 minute averaged BC, O_3, and CO data and meteorological data. Daily averages were examined to probe overall increases in regional pollutants, while 15 minute averages were used to detect local pollution.

Rwanda has two rainy seasons roughly occurring in March-April-May (MAM) and September-October-November (SON), and two dry seasons during December-January-February (DJF) and June-July-August (JJA). This generalized definition and durations of the seasons are used the purpose of comparing data for multiple years and is used throughout this paper. High variations in BC concentrations can be seen in the BC time series (Figure 3) ranging from below 100 to above 20,000 ng m^{-3}, with an average value of 1,700 ng m^{-3} (standard deviation: 1,600 ng m^{-3}). Peak concentrations corresponded to dry seasons. CO and O_3 mixing ratios also increased during the dry seasons compared to the rainy seasons, though not as pronounced as the BC increases. This decrease is partially due to the efficient rainout of black carbon particles during the rainy season. The diurnal, weekly, and monthly variations in concentrations of each species, normalized to their average, are shown in Figure 4.

It has been known for some time that wide-scale biomass burning in sub-Saharan Africa has a large seasonal effect on the atmosphere (e.g., Archibald et al., 2010; Crutzen and Andreae, 1990). Understanding and separating these seasonal effects from anthropogenic emissions can be difficult without continuous data sets both during and outside of this period, especially as both biomass burning and anthropogenic emissions in this region of the world emit BC, CO, and PM. Biomass

505 burning emissions have also been shown to affect O_3 formation under the right
506 meteorological conditions.
507 To explore the sources of BC and CO, at the RCO, seven-day HYSPLIT back
508 trajectories were run every 6 hours using NCEP/NCAR reanalysis meteorological data
509 (2.5 x 2.5 degree resolution) (Kalnay et al., 1996). This analysis provided insights on
510 the approximate origin and trajectories of air masses before arriving at RCO
511 measured at the RCO. These HYSPLIT back trajectories were separated into DJF,
512 MAM, JJA, and SON and are shown with MODIS satellite fire count data colored by fire
513 radiative power (FRP, W m^{-2}) (Figure 5). The MODIS fire count data and radiative
514 power are used strictly for qualitative, not quantitative, purposes in this work. Here
515 we observe that, as major biomass burning sites moved to the north and west in DJF,
516 transport direction was also primarily northerly, and as biomass burning move to
517 Southern Africa in JJA, the prevailing wind directions were also southerly. Although
518 Rwanda itself had few large-scale fires, its geographical position and meteorology
519 meant that it experienced transported fire haze from both major burn seasons. Black
520 carbon measured at the station tracked fairly well with summed daily FRP for sub-
521 Saharan Africa (Figure 5). This suggests that transport from regional biomass
522 burning has a twice-yearly effect on BC concentrations in Rwanda, despite the
523 different locations of the biomass burning in sub-Saharan Africa.
524 To further examine pollution transport to the RCO, the HYSPLIT back
525 trajectory geographical areas were gridded (using the R Openair package, (Carslaw
526 and Ropkins, 2012)) and merged, using date and time, with measured BC
527 concentrations and mixing ratios of CO. This was done to generate concentration-

528 weighted back trajectories (cwt) for each season (more details on cwt available in
529 (Hsu et al., 2003; Seibert et al., 1994))(Figure 6). Trajectory time in each grid and
530 arrival time of each air mass were taken into account in this model to predict the
531 likely source regions and emission concentrations of pollutants measured at the RCO.
532 This was done to determine likely source regions of air pollution at the RCO by
533 comparing arrival times of air masses to the RCO and the time series of pollutants.
534 This method has proven fairly effective at identifying emission sources when
535 comparing predicted emission regions to emissions inventories (Lupu and Maenhaut,
536 2002) and is good as a rough estimate of emission regions with no apriori
537 information (Kabashnikov et al., 2011).
538 BC and CO appeared to originate from similar areas, as expected due to their
539 overlapping sources of inefficient combustion and biomass burning. During JJA,
540 significant BC and CO appeared to originate from southern Africa and Madagascar, as
541 well as from local sources near the RCO. During DJF, the source of these pollutants
542 appeared to be much closer to the RCO, as major fires in the DRC and Uganda were
543 also closer to the station. Throughout the measurement period, but particularly DJF,
544 the Lake Kivu region also appeared to be a source of BC and CO. The Lake Kivu region
545 is densely populated and use of both cook stoves and diesel generators is common.
546 In addition to direct emissions of BC and CO, other emissions such as volatile
547 organic compounds and oxides of nitrogen from biomass burning are known to affect
548 tropospheric O_3 concentrations (Jaffe and Wigder, 2012; Sauvage et al., 2005). It
549 appears that such emissions could have played a role in the observed seasonal
550 increase in O_3 mixing ratios of approximately 20 ppb in DJF and 25 ppb in JJA above

551 rainy season levels at the RCO. This increase of about 5 ppb O_3 during JJA versus DJF
552 was potentially due to the mixing of biomass burning emissions with anthropogenic
553 emissions from east African cities such as Nairobi, Dar Es Salam, and Kampala during
554 the JJA dry season. It also could have been the result of generally higher solar
555 radiation during the JJA season in Rwanda (Safari and Gasore, 2009). A mix of biomass
556 burning and anthropogenic emissions from southern Africa could have been
557 transported to Rwanda after photochemical aging and processing. Direct source
558 apportionment of O_3 is difficult as O_3 is formed from the right combination of VOCs,
559 NOx, and favorable meteorological conditions (Baier et al., 2015; Geddes et al., 2009;
560 Gong et al., 2017; Monks et al., 2015). During the DJF dry season, fires are closer to
561 Rwanda and away from major urban areas. During June and July, a loose correlation
562 (R=0.47 and 0.45, respectively) between O_3 mixing ratios and BC concentrations was
563 observed, while no correlations (R=-0.04, -0.15, and 0.07) were observed in
564 December, January, and February.

565 **3.2 Absorption Angstrom Exponent and BC Source Apportionment**

566 It is important to understand the pollution emission sources in East Africa,
567 beyond large-scale biomass burning, in order to enact policies and actions to reduce
568 these emissions. One way scientists have estimated fuel combustion versus biomass
569 burning BC particulate is by measuring the color of the particles (wood smoke
570 particles have enhanced absorption in the UV, while fossil fuel combustion particles
571 have flat absorption over all wavelengths)(Kirchstetter and Thatcher, 2012;
572 Sandradewi et al., 2008). The Aethalometer's seven wavelengths allow measurement
573 of the wavelength-dependent aerosol absorption and the calculation of absorption

574 coefficients that can be used to infer the potential sources of BC aerosol (Drinovec et
575 al., 2015; Sandradewi et al., 2008) measured. Theoretically, from the wavelength
576 dependence of aerosol absorption, BC from fossil fuel and wood smoke can be
577 differentiated (Sandradewi et al., 2008). Though this two-component model can
578 provide a valuable knowledge on knowledge on source attribution of BC this model
579 has some limitations. This model is more accurate if calibrated to local conditions as
580 burning and aging during transport affects aerosol 's wavelength-dependent
581 absorption (Dumka et al., 2013; Harrison et al., 2012), as different fuels and wood
582 biomass burning creates aerosol with different radiative properties and the standard
583 model, based on European studies, has been shown to be less applicable in developing
584 countries (Garg et al., 2016).
585 From the Aethalometer data, wavelength dependence of absorption
586 coefficients and the absorption Ångstrom exponent (AAE) were calculated and
587 compared to literature values of biomass burning and fossil fuel combustion (Figure
588 7). The AAE is a dimensionless property commonly used to characterize the
589 wavelength-dependent absorption of BC and gives clues on the source and/or aging of
590 BC when compared to laboratory and other ambient studies (Chung et al., 2012; Lack
591 and Langridge, 2013; Russell et al., 2010; Yuan et al., 2016). The AAE values assigned
592 for the standard Aethalometer model separating the BC from biomass burning and
593 fossil fuel combustion are two and one, respectively (where two represents an
594 average AAE for woodsmoke of different types and ages) (Kirchstetter et al, 2004;
595 Sandradewi et al, 2012; Drinovec et al. 2015). In this work, standard mass absorption
596 cross-sections (MACs) for each wavelength provided by the manufacturer of the

597 Aethalometer were used to calculate the absorption coefficient (b_{abs}) at each
598 wavelength. For pure BC from fossil fuel, $b_{abs} \sim 1/\lambda$ and the AAE between two
599 wavelengths (470 nm and 950 nm) is 1 using the equation $\ln(b_{abs}\lambda_1/b_{abs}\lambda_2)/\ln(\lambda_2/\lambda_1)$.
600 The average AAE (averaged for entire measurement period between July 2015
601 and January 2017) was calculated to be 1.65 (+/- 0.14) at the RCO using the 470 and
602 950 wavelength absorption and MACs (Figure 10)(Sandradewi et al., 2008; Drinovec
603 et al. 2015). These wavelengths were chosen as the AAE calculated from 470 and 950
604 is generally comparable with other literature values(Saarikoski et al., 2012). The
605 calculated AAE values were on par with AAE calculated from measurements taken in
606 areas heavily influenced by biomass burning (Chung et al., 2012; Lack and Langridge,
607 2013; Russell et al., 2010; Saleh et al., 2013; Sandradewi et al., 2008; Yuan et al.,
608 2016). Past studies have reported an AAE of 1.2-$_{2.5}$ for biomass burning aerosol
609 (Andreae and Gelencsér, 2006; Chung et al., 2012; Russell et al., 2010; Saleh et al.,
610 2013, 2014). While daily only small variations (+/- 0.05) for AAE were observed (,
611 significant seasonal differences in this value were found, with monthly averaged
612 values ranging from 1.5 (dry season) to 1.9 (at the end of the long rainy season). This
613 seasonal difference is shown with the 30 day running mean of the AAE (Figure 7).
614 Studies in southern Africa measuring savanna and crop burning found an AAE of
615 around 1.45 for ambient black carbon aerosol, and in the dry season savanna and crop
616 burning are the prevalent type of large-scale biomass burning in sub-Saharan Africa
617 (Russell et al., 2010). The AAE calculated from the Aethalometer data at the RCO was
618 higher during the rainy season when local emissions dominated our measurements
619 (Figure 7). Eucalyptus burning, the most prevalent burning near the station (for

charcoal making, cooking fires, brick kiln fuel) was measured in laboratory experiments to have a higher AAE than savanna burning (AAE of 1.71 +/- 0.50 calculated between 405 and 781 nm wavelengths)(Chung et al., 2012). Eucalyptus trees and savanna burning were certainly not the only two types of solid biofuel influencing measurements at the station, but the difference in AAE of aerosols produced from different fuels means that the AAE will have large variations based on fuel wood or other biomass used and this was reflected in our data.

Using the Aethalometer model with standard inputs not accounting for the different types of fuel used in East Africa versus Europe, a high influence of fossil fuel black carbon emissions was calculated: in the dry season, over 50% of black carbon was assigned to be fossil fuel in origin (Figure 7). Fossil fuel emissions certainly influenced the pollution at the RCO, as air masses from Kigali, Kampala, Nairobi, and Dar es Salaam were transported to the station. These cities have high black carbon emissions from generators, fossil fuel power stations, and older diesel vehicles but would also have significant biomass cook stove emissions (Gatari and Boman, 2003; Koch et al., 2009; Mkoma et al., 2009; van Vliet and Kinney, 2007). However, at <10% fuel demand of fossil fuel (all types, see Table 2) versus >90% wood and charcoal fuel demand, even if the g BC per kg fuel from diesel was 4x higher, and all fossil fuel use was unregulated diesel (unlikely), well under half of the measured BC should be from fossil fuel combustion emissions. Aging with transport would increase the AAE of the aerosol, not decrease, so aging should not cause this seasonal difference as transport distances of BC are longer during the dry seasons.

642 In order to gain more insights into the sources of BC we also examined the
643 BC:CO. CO is also released by inefficient combustion and the ΔBC: ΔCO ratio can be
644 different for different emission sources. In order to calculate this ratio we first
645 converted the CO mixing ratios to concentrations (in $\mu g\ m^{-3}$), and then subtracted the
646 95th percentile values for CO and BC from their respective concentrations. For the
647 entire data set, the ΔBC: ΔCO (both in $\mu g\ m^{-3}$) ratio was 0.014 (R^2 0.79, n = 40523).
648 The ΔBC: ΔCO ratio varied seasonally, with monthly average peaks reaching 0.016 in
649 December, February, and July and lows below 0.01 in April. The average ratio of 0.014
650 for the measurement period was almost twice as high as in biomass burning plumes
651 sampled over West Africa in an aircraft campaign (0.0072) (Moosmüller and
652 Chakrabarty, 2011) but on par with or lower than measurements taken during the
653 INDOEX campaign in the Indian Ocean (Dickerson et al., 2002). A study in Germany
654 and Mexico found a correlation between diesel vehicle use and higher BC:CO
655 (Baumgardner et al., 2002), while other studies have also found an increased
656 ΔBC: ΔCO during periods more influenced by biomass burning (Pan et al., 2011). A
657 study in India found no correlation in biomass-burning and fossil fuel-influenced
658 ΔBC:ΔCO air masses (Sahu et al., 2012), as there are a wide range of ratios measured
659 from the same source (Dickerson et al., 2002; Sahu et al., 2012). The high ΔBC:ΔCO
660 ratio at the RCO could be due to the prevalence of older diesel engines in the country,
661 which emit more BC to CO than newer engines (Cai et al., 2013), but, as the highest
662 value occurs during the Rwanda dry seasons and the continental biomass burning
663 seasons , likely the ratio is governed in part by rainout as BC is more easily removed
664 by wet deposition than CO. In this study, we were not able to use this ratio to further

separate biomass burning BC from fossil fuel combustion BC. However, this inconclusiveness highlights the need for further study, as ascribing a source to local pollution is important. Further work on emissions profiles relevant to sub-Saharan Africa could clarify these issues. Cookstoves, cookfires, agricultural and trash burning, and older diesel combustion emissions are all likely sources of BC and could be targeted by government policy. Understanding the most important source of local pollution is important for developing efficient government policies for air quality.

3.3 Examination of Local and Regional Pollution

The continuous collection of BC, CO and O_3 data during the dry and rainy seasons allowed examination of both transported and local pollution. Here we define local pollution as pollution originating within twelve hours transport time under typical wind speed conditions (<150 km, including both Rwanda and the border areas with DRC and Uganda). During Rwanda's rainy seasons, the continental fire count is also at a minimum, reducing large-scale biomass burning influence. The region's emissions are from small-scale agricultural burning, charcoal making, cooking fires, brick production (located in the valley below the station and throughout the region), vehicles, diesel and heavy fuel-oil power plants, and diesel generators. These activities continued throughout the rainy season and dry season at similar rates.

The baseline daily average BC concentration in the rainy season remained at 0.5-1 $\mu g\ m^{-3}$ after 12 hour periods without rain, which could be considered as contributions of small but numerous diffuse emission sources to daily BC concentration in this region. These values, while significantly below those during the biomass burning affected seasons, are not negligible. If all BC during the rainy seasons

688 is assumed to be local in origin (within one day of transport, as typically rain occurs
689 each day during the rainy season), and this level remained the same throughout the
690 year, yearly average contribution of local emissions to BC would vary between 18-
691 100% of the total measured BC concentration at the RCO. The shoulder months of
692 September and February have been removed from this calculation as they have both
693 rain and biomass burning influence, but on a yearly scale, around 35% of BC
694 concentration measured at the station could originate from local emissions. This
695 estimate is a high estimate as transport of BC is still possible above the boundary
696 layer, but it is on par with previous estimates of the contribution of savanna and
697 forest burning BC emissions versus other emission sources in sub-Saharan Africa
698 (Bond et al., 2013).

699 **3.3.1 Diurnal Variations in BC, CO and O_3**

700 Diurnal variations in concentration of pollutants can provide important
701 insights into information on local as well as regional pollution emission sources.
702 Boundary layer height and whether or not the station is measuring the free
703 troposphere or the polluted boundary layer is also important for understanding
704 diurnal changes in pollutant concentrations (Nyeki et al., 1998). Diurnal variations in
705 BC concentrations, CO mixing ratios and O_3 mixing ratios observed at RCO in different
706 seasons are shown in Figure 8. At the RCO, the O_3 mixing ratio exhibited a diurnal
707 cycle with a peak in concentration in the evenings (after ~ 8 pm), with steady levels
708 through the night and a minimum during mid-day. The increase of O_3 in the later
709 evening is likely mainly regional O_3 transported above the boundary layer measured
710 at night (as the boundary layer height lowered), but some regionally formed O_3 could

711 also be transported to the station by the evening. Similar diurnal O_3 profiles were
712 found at other mountain locations remote from urban centers (Zhang et al., 2015).
713 This diurnal pattern persists in all seasons (Figure 8) and occurred on daily time
714 scales. The differences in diurnal minima and maxima were highest in the June-
715 August period, and lowest in the December-February period. This difference may be
716 due to the differences in biomass burning proximity (far in JJA, closer in DJF), primary
717 wind direction (southerly versus northerly), and also solar intensity (highest in JJA,
718 (Safari and Gasore, 2009)).

719 BC had mid-morning and early evening (~ 6 pm) peaks that coincided with
720 both cooking times and kerosene/generator use times (sunset at 6 pm each night),
721 indicating local influence on BC, before the station was outside of the boundary layer
722 in the evening. These peaks occurred approximately two hours before the O_3 peak
723 each evening, further indicating some regional or local influence. Regional transport
724 of BC higher in the atmosphere should be greater in JJA/DJF (more BC) and solely
725 boundary-layer driven BC concentration changes would be greater during these
726 times, but the normalized diurnal changes from daily baseline to daily peak remain
727 similar throughout the seasons. Additionally, no persistently higher nighttime (after 8
728 pm) BC baseline levels were observed in these data. CO mixing ratios had a similar but
729 less pronounced diurnal variation.

730 **3.3.2 Case Study: High and Low Periods of Black Carbon**

731 Seasonal variations are too long to fully capture local pollution events. To
732 further examine local pollution in 2016, high BC time periods during DJF (2/12-2/16)
733 and JJA period (8/3-8/6), and one period of low black carbon in the MAM period

(5/18-5/22) were examined for their BC:CO ratio and correlation, relationship of O_3 to CO, and AAE (Figure 9). From this figure, no clear trends are observed. The BC:CO is 10 with an R^2 of 0.48 for the polluted DJF period, 8 with an R^2 of 0.47 for non-polluted period in May, and 16.6 with an R^2 of 0.72 for the polluted JJA period. The average AAE for the May period was 1.79, for February 1.53, and for August 1.53 as well. Unfortunately, no O_3 data was available for the August period. O_3 in February was loosely correlated with CO (R^2 0.17) and averaged 39 ppbv, with a peak value of 43. O_3 in May had averaged 26 ppbv with a peak of 34 ppbv, and no correlation with CO.

During the May period, spikes in very local pollution can be seen (Figure 10). These hour plus increases in BC happen at regular cooking times in the valley and, due to their shorter (hourly) time scales of rise and fall, cannot be explained by changes in boundary layer conditions. The diurnal patterns of increased BC during cooking times persist during the polluted period, but on a baseline of regional pollution. Some of the diurnal variability in black carbon background can be attributed to boundary layer conditions, seen with the slow and steady changes over the course of the day not confined to the timescales of activity in the valley.

3.3.3 Potential Twice-Yearly Influence Biomass Burning in equatorial Africa

The BC in Rwanda has peaks in both dry seasons, and these peaks correlate well in time with the FRP in sub-Saharan Africa, as shown in Figure 5. However, the site in Rwanda is one site, and drawing a conclusion on regional seasonal pollution trends is difficult without other data. BC is only one component of $PM_{2.5}$. Other components of $PM_{2.5}$ include dust, organic carbon, nitrates, sulfates, and ammonium.

BC is indicative of combustion, and when BC rises due to combustion processes, often $PM_{2.5}$ will rise (though combustion aerosol contains a significant organic fraction).

Although no continuous measurements of BC are widely reported in sub-Saharan Africa, recently the US Embassies in Addis Ababa, Ethiopia, and Kampala, Uganda have begun continuously measuring $PM_{2.5}$ concentrations. The raw data is collected and reported online on the OpenAQ platform (OpenAQ.org). This dataset on $PM_{2.5}$ concentrations in major cities over different seasons in this region has been valuable in gaining basic insights into the seasonal characteristics of $PM_{2.5}$ concentrations in the region (Figure 11). While $PM_{2.5}$ is not the same as BC, biomass burning is thought to be a major contributor to $PM_{2.5}$ in sub-Saharan Africa. By examining the $PM_{2.5}$ concentration in a city in the same region as Rwanda (equatorial east Africa) and a different region (further north), increased understanding on the impact of the dual biomass burning seasons for different regions in sub-Saharan Africa's air quality can be understood.

The $PM_{2.5}$ concentrations in both Addis Ababa and Kampala showed clear seasonal patterns, though the seasonal patterns differed at the two sites. Addis Ababa (Ethiopia) is much further north than Rwanda and Ethiopia is in general higher in elevation than Rwanda (though at 2355 m, not higher than the RCO) and closer to the Indian Ocean. In Addis Ababa, the dry season is also in DJF, but measured $PM_{2.5}$ concentrations were low during this season. HYSPLIT back trajectory calculations confirmed that air masses during this time of the year originated over the ocean, not from the continent. Kampala, Uganda is close to Rwanda, near the equator, and has similar seasonality. Rainy and dry season extrema are shown in the available

779 Kampala $PM_{2.5}$ data, with an enhancement during February and JJA of around 15 to
780 25-30 μg m^{-3}, respectively, above $PM_{2.5}$ concentrations during other months.
781 While not pictured here, South Africa has the most air quality monitoring stations of
782 any sub-Saharan African country. Results from these stations show a $PM_{2.5}$ peak in
783 the southern burning season (June-October), though June-July was mostly due to local
784 heating (Hersey et al., 2015) and August-October was related to biomass burning
785 (Horowitz et al., 2017; Tesfaye et al., 2011).
786 From these data, it appears that African countries near the equator may be
787 positioned to experience six months per year of transported regional fire haze, from
788 both the northern and southern biomass burning seasons. This is potentially unique
789 to the region and this effect may be seen in other pollutants and short lived climate
790 forcers. In fact, beyond BC and $PM_{2.5}$, the MOZAIC campaign in the late 1990s and
791 early 2000s measured ambient O_3 mixing ratios at the Nairobi, Kampala, and Kigali
792 airports. This campaign found Kigali, despite its smaller size and lower vehicle count,
793 to have the highest O_3 mixing ratios among them (Sauvage et al., 2005). They
794 measured a similar in magnitude increase in surface O_3 mixing ratios during the JJA
795 season in Rwanda as our measurements at the RCO, although DJF was not measured
796 in their work.
797 O_3 measurements were made in Brazzaville, Republic of the Congo during
798 January and February O_3. While much further west than Rwanda, in Brazzaville O_3
799 mixing ratios also increased during January and February, parallel to Rwanda, with
800 monthly averages during January and February 25 ppb greater than the minimum of
801 <30 ppb in April (Sauvage et al., 2005). This suggests influence from northern

hemisphere biomass burning to O_3 mixing ratios at Brazzaville. O_3 in JJA at Brazzaville was almost 30 ppb higher than in January and February, however, so transport of air mass from the south and southern Africa biomass burning had a greater influence on O_3 in the region than transport from the north and biomass burning in central Africa. The 1992 SAFARI campaign also measured O_3 in sub-Saharan Africa throughout all seasons, and measured a seasonal ozone concentration peak during the JJA period for central and southern Africa (Thompson et al., 1996). A separate, large peak for DJF was not as observable in the SAFARI data (Thompson et al., 1996). SAFARI measurements took place prior to 1993, meaning that significant development in sub-Saharan Africa could have taken place between the SAFARI campaign and the MOZAIC campaign (1997-2003) that could drive the increasing O_3 in DJF as well as JJA over a period of almost a decade. More recent measurements were made in a 2000 SAFARI campaign, but not as far north as the previous SAFARI campaign (Otter et al., 2002) and the positioning of the measurements could have also had an effect on O_3 seasonality, as southern Africa is more influenced by biomass burning from August-October. The SAFARI campaign measured the total column O_3, not the ground-level O_3 mixing ratios, so data are not directly comparable.

4. Conclusions

In this work, we present the first long-term and continuous measurements of short-lived climate forcers for a nearly two-year period from July 2015 to January 2017 at the Rwanda Climate Observatory located at Mt. Mugogo in Rwanda. From these observations, we find that:

1. During Rwanda's two dry seasons, transported pollution led to high black carbon and carbon monoxide levels at the RCO, surpassing concentrations measured in many major cities elsewhere. Emissions from large-scale crop and savanna fires could have a wide-reaching effect on this region and likely drive the increased BC and O_3 measured during DJF and JJA by our study and O_3 by past studies in equatorial Africa. The dense population of equatorial East Africa and the double impact of the two fires seasons could lead to significant public health problems for the population in Rwanda and equatorial East Africa as exposure to elevated levels of $PM_{2.5}$ and BC concentrations occurs six months out of the year.
2. Local emissions beyond large-scale biomass burning influence were constant and estimated to contribute up to 35% of the annual average measured black carbon concentration, if black carbon during the rainy season was assumed to be completely local (Rwanda and neighboring countries) in origin (ranging from 0.5-1 $\mu g\ m^{-3}$ daily average measured BC). These local emissions, from different combustion sources (e.g., cooking fires, inefficient diesel generators and engines with sub-standard fuel use, solid biomass fuel burning, small agricultural fires), are likely concentrated in the densely populated Rwanda and Lake Kivu economic area. Rwanda's population is growing quickly. As these local emissions are related to

population density, air pollution will likely increase unless there is government intervention.

3. Different combustion fuel and burning practices in Europe and East Africa calls into question the accuracy and applicability of a two-component model for estimating BC from fossil fuel combustion and biomass burning using AAE approximations for biomass burning and fossil fuel combustion aerosol measured in Europe for use in East Africa. There may also be different mass absorption cross-sections for aerosols measured at the RCO than in Europe or North America. This shows the need for multiple on-ground measurements to fully understand pollution sources in different regions of the world, notably in Africa. However, seasonal variations in the wavelength dependence of ambient BC particles did point to different sources of BC particles and this should be further explored in future studies.

4. The measurements we have provided in this study will be useful in advancing atmospheric science in Rwanda, which has limited long-term and in-situ atmospheric data.

These data and analyses, while acknowledging the high influence of regional biomass burning, also show that measurable decreases in air pollution could be achieved within eastern and central Africa with targeted local policies, emphasizing cleaner diesel vehicles and generators, reduced wood-fuel reliance for cook stoves, and improved cook stoves to burn biomass fuel more efficiently. Currently, over 2

million households in Rwanda rely on wood burning (including charcoal) for cooking. While reducing this number will have significant economic costs, putting in place infrastructure for alternative cooking fuels (pellet stoves, LPG stoves, electrical stoves) could help the country avoid even higher local air pollution emissions and associated adverse impacts as the population grows. Diesel-fueled minibuses, common transport between towns in Rwanda and within Kigali, and older diesel vehicles are also high emitters of black carbon but newer vehicles with emissions control technology may be economically beyond the reach of local bus companies and citizens. Continuing to grow electrical capacity and connection will reduce the use of kerosene lanterns and diesel generators, and will reduce air pollution if additional energy capacity is achieved through renewable sources (solar, hydropower). The huge influence of regional biomass burning, exacerbated by equatorial East Africa's meteorology, and the potential influence of anthropogenic emissions from major cities on O_3 formation in these regions must also be examined as this area develops. Halting slash-and-burn agriculture, reducing trash incineration, and developing ways to warn the population during periods of high pollution from naturally occurring savanna and forest fires should be an important agenda for regional discussions on environmental, public health, and other development issues.

6. Future Work

The government of Rwanda is working to establish an air quality and climate change monitoring network throughout the country to measure ambient criteria air pollutants and other key climate change related components of atmospheric pollution. Building knowledge of air quality and climate change related emissions in this data-

poor area of the world is essential to fill the large data and knowledge gap in this region. Adding ground-based measurements, comparing measurements to satellite data, using data to evaluate and improve existing emission inventories, improving accuracy of global/regional air quality and climate change models, and using data for quantification of impacts of air pollution and climate change will help local governments design appropriate mitigation strategies rooted in data and local context.

7. Data Availability

This data will be made available at the AGAGE website, https://agage.mit.edu/data/agage-data. All data used in this article will be made available as of publication and data from this project on a rolling basis after quality control.

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919
920

921 Table 1: Instruments used in this study and measurement period used for analysis

INSTRUMENT	SPECIES MEASURED	MEASUREMENT PERIOD	AVERAGE VALUE	MIN VALUE	MAX VALUE
PICARRO G2401 CAVITY RING DOWN SPECTROMETER	CO_2, CO, CH_4, H_2O	MAY 2015-JANUARY 2017	215 (CO ppbv)	63(CO ppbv)	663(CO ppbv)
MAGEE SCIENTIFIC AE33 7-WAVELENGTH AETHALOMETER	BLACK CARBON ($PM_{2.5}$, CYCLONE IMPACTOR ON INLET)	MAY 2015-JANUARY 2017	1692 (ng m^{-3})	8 (ng m^{-3})	17445 (ng m^{-3})
TELEDYNE T400 API	O_3	MAY 2015-JANUARY 2017	40 (ppbv)	10 (ppbv)	84 (ppbv)
VAISALA WXT	MET PARAMETERS (RH, WS, WD, T, P)	JULY 2015-JANUARY 2017			

Table 2:

Fuel Demand in Rwanda (2016, Rwanda Ministry of Infrastructure)

Fuel Type	Demand
Petrol	120442 kL
Diesel	178529 kL
Kerosene	22288 kL
Heavy Fuel Oils	59292 kL
Jet-A	18235 kL
Wood (charcoal + natural)	4,200,000 metric tons

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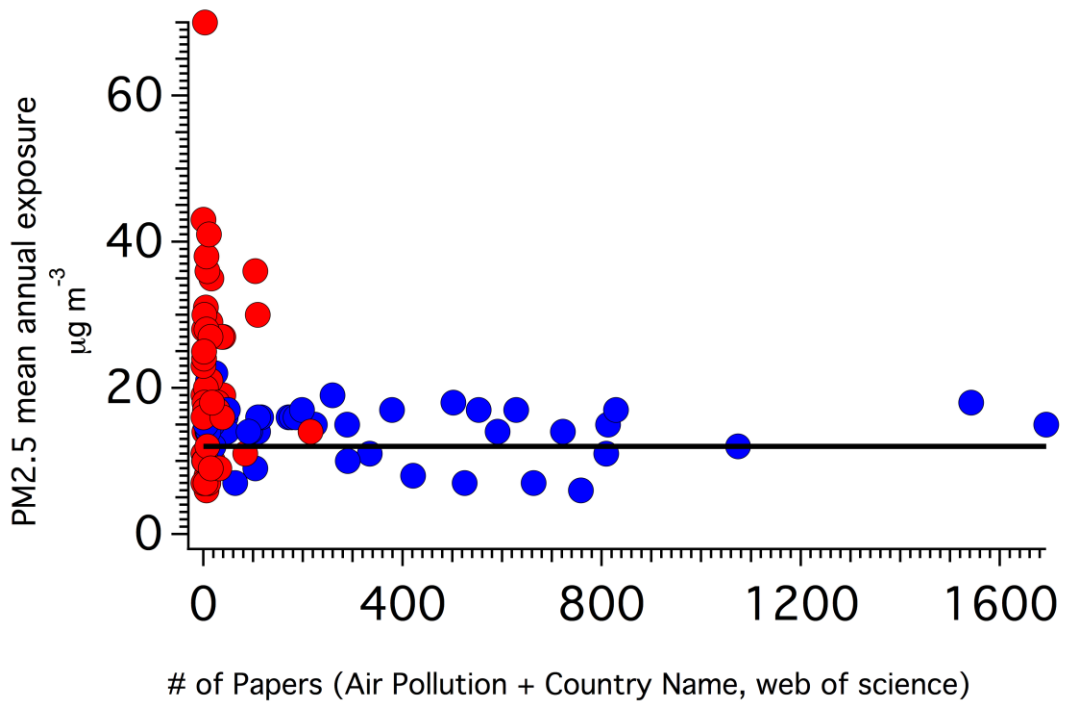
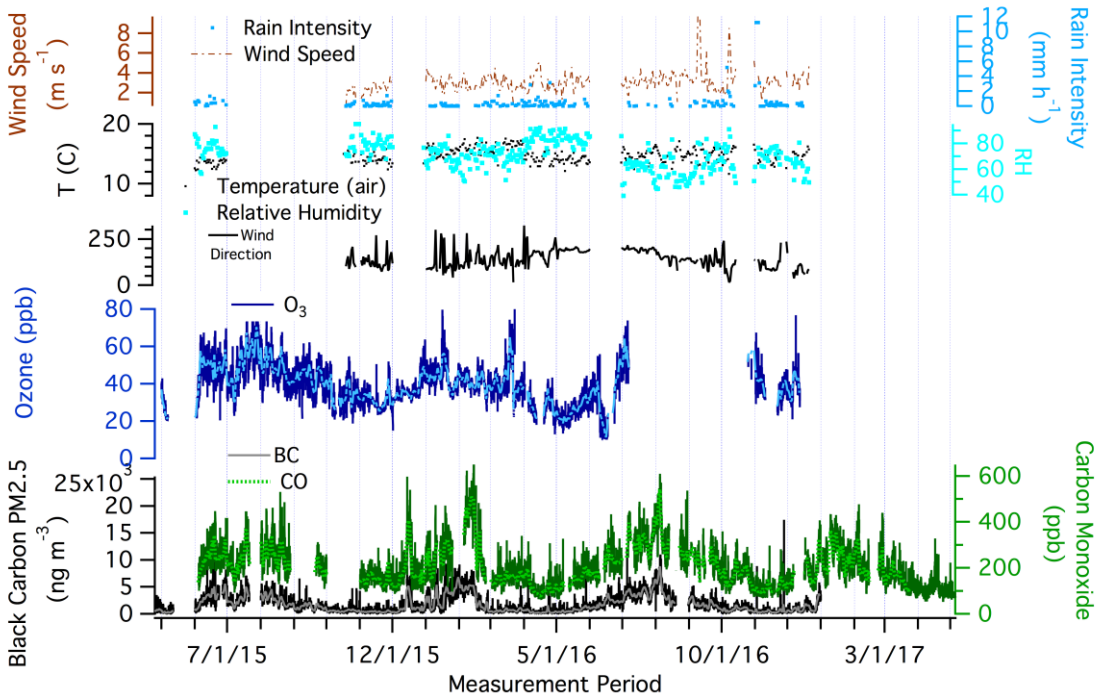


Figure 1: Africa (red) and Europe (blue), $PM_{2.5}$ mean annual exposure (https://data.worldbank.org/indicator/en.atm.pm25.mc.m3) and paper count of country + air pollution (from Web of Science).

Figure 2. From top left moving counter-clockwise: an aerial view of RCO at Mt. Mugogo Main Peak, the station with towers in the background, and the location of Mt. Mugogo in Rwanda (blue pin) in relation to Kigali (yellow pin).



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1165 **Figure 3**. From the top down up: (a) wind speed (red dotted) and rain intensity (blue

1166 dash) daily average values; (b) temperature (black) and relative humidity (light blue)

1167 values; (c) ozone (dark blue, light blue) (15 minute, daily); (d) black carbon (black,

1168 grey) and carbon monoxide (dark green, light green) (15 minute, daily) average

1169 concentrations.

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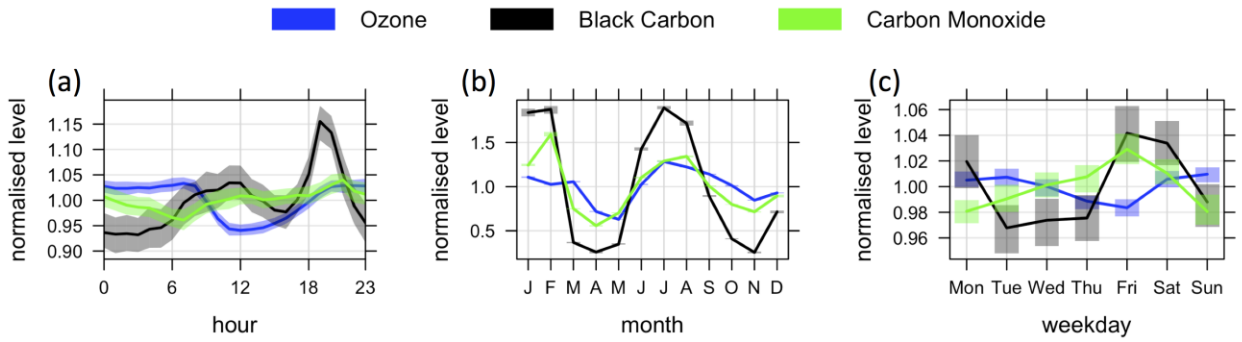

Figure 4. Normalized temporal variations of O_3 mixing ratios, CO mixing ratios, and BC concentrations by: (a) hour (diurnal) (b) month, and (c) day of the week. Shaded areas are 95% confidence intervals.

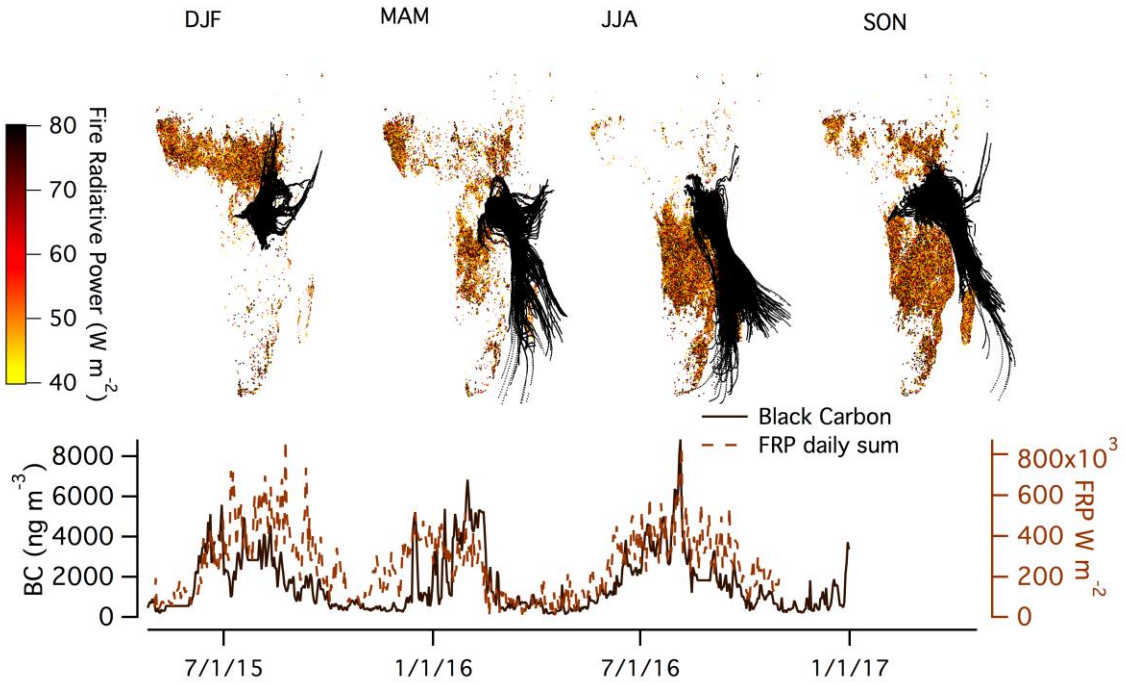

Figure 5. (a) Seasonal fire radiative power data acquired with the MODIS instrument and back trajectories of air masses (generated with the HYSPLIT model) reaching the Rwanda Climate Observatory for the period May 2015 to January 2017. Seasons in Rwanda are split into: short dry season, December-January-February (DJF), long rainy season, March-April-May (MAM), long dry season, June-July-August (JJA,) and short rainy season, September-October-November (SON). (b) The time series of daily average BC concentration and the daily sum of Fire Radiative Power (W m^{-2}) from the pictured data bound by the furthest HYSPLIT backtrajectory reaches each season (box defined by the most north, south, east, and west point the HYSPLIT backtrajectories reach).

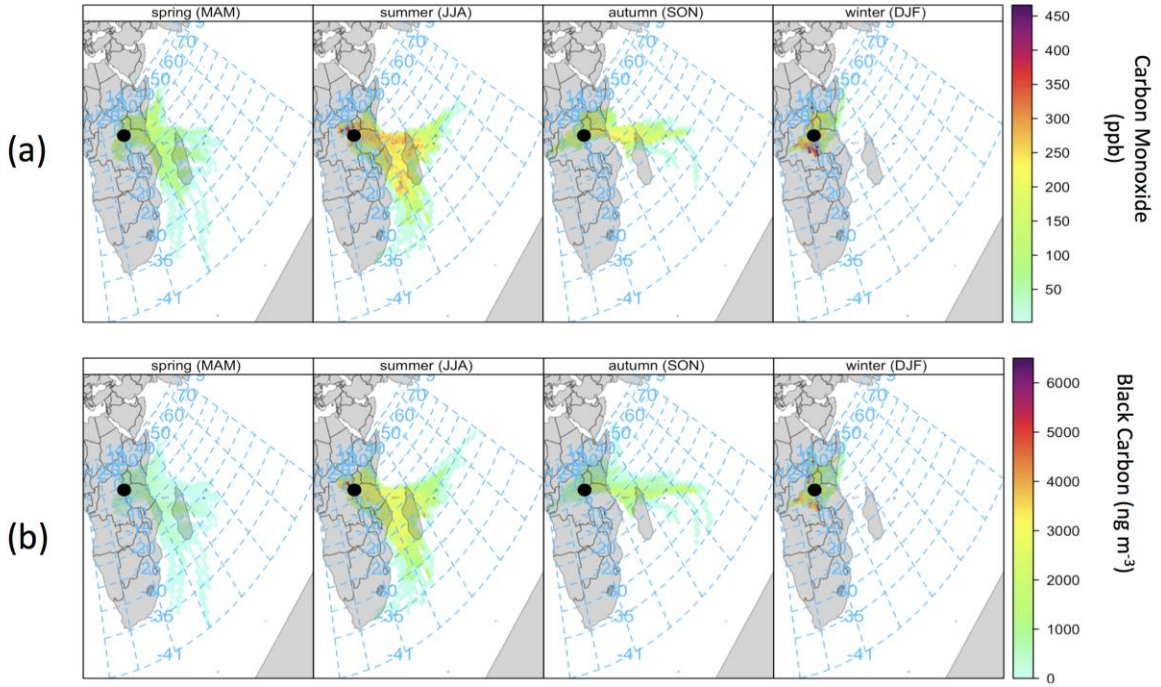

Figure 6. Concentration-weighted back trajectories of (a) CO and (b) BC, separated by season, for measurements at the Rwanda Climate Observatory (black dot) for the period of July 2015-January 2017.

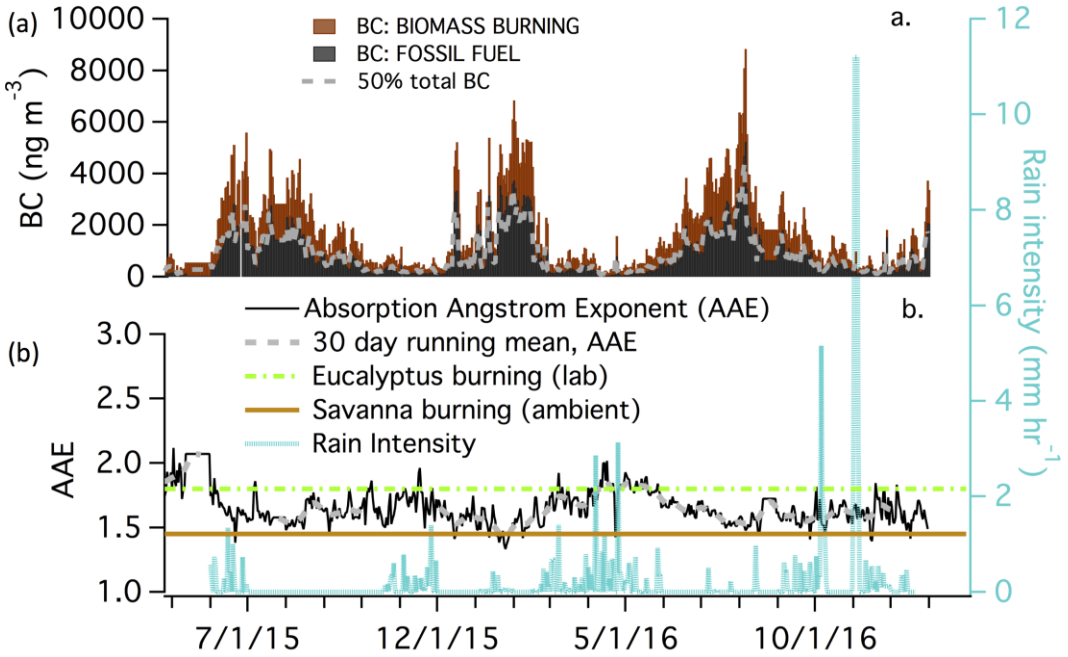

Figure 7. (a) Time series of contributions of fossil fuel combustion and biomass burning to BC concentrations observed at RCO. (b) Daily average absorption Angstrom exponent (AAE) measured at RCO (black line), rain intensity, and published AAE for Eucalyptus burning ((Yuan et al., 2016), laboratory studies, green line) and savanna burning ((Russell et al., 2010), ambient, brown line) also shown as reference.

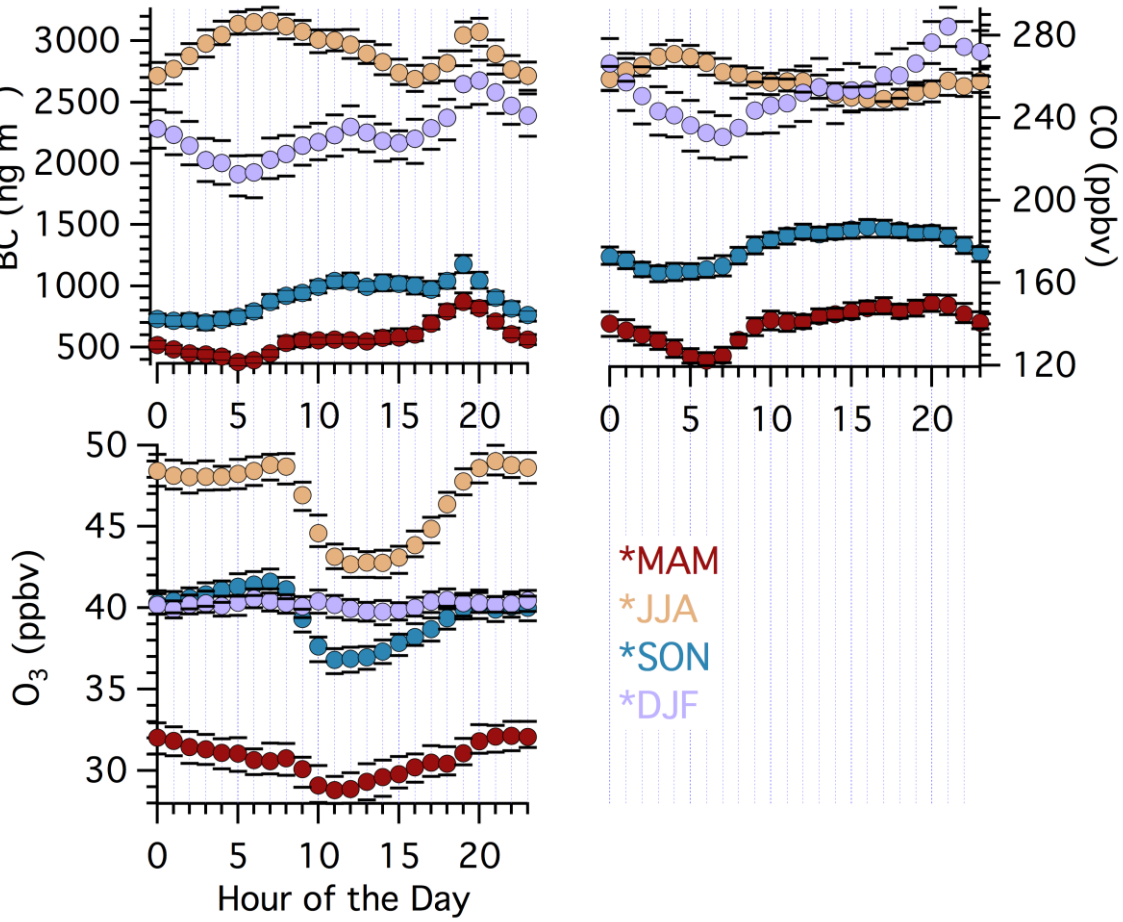

Figure 8. Seasonally separated diurnal profiles of (a) BC concentrations, (b) CO mixing ratios, and (c) O_3 mixing ratios, colored for each season. The circles represent mean concentrations and the lines represent 95% confidence intervals.

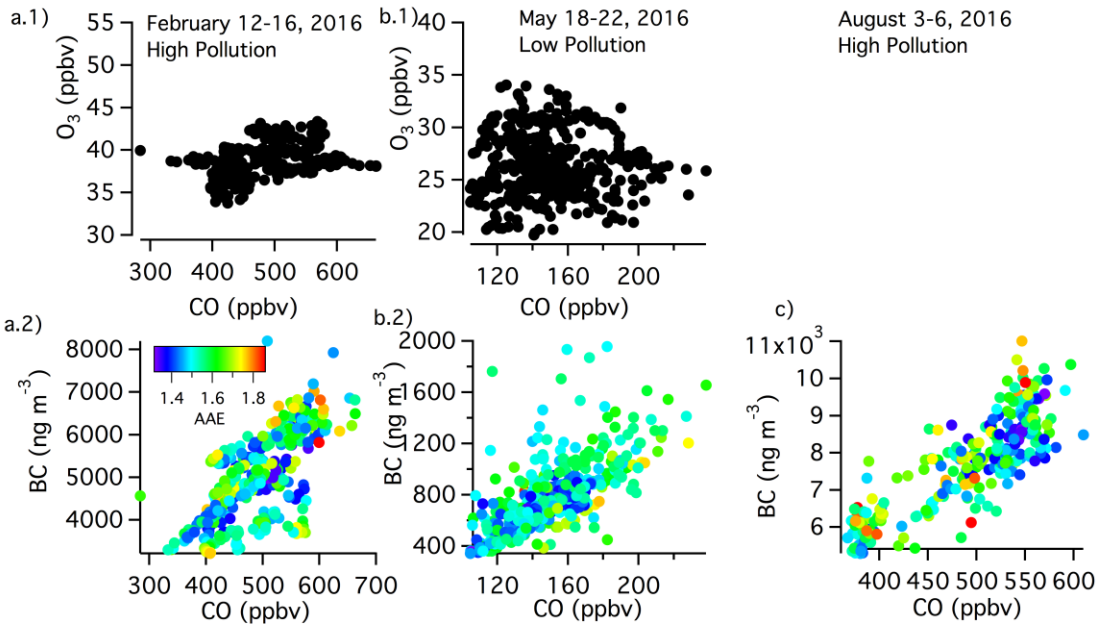

Figure 9: Polluted period in DJF (a), non-polluted period in MAM (b), and polluted period in JJA (c). Comparison of O3 and CO in a.1 and b.1, and comparison of BC and CO, color-coded by AAE, in a.2, b.2, and c for each respective period.

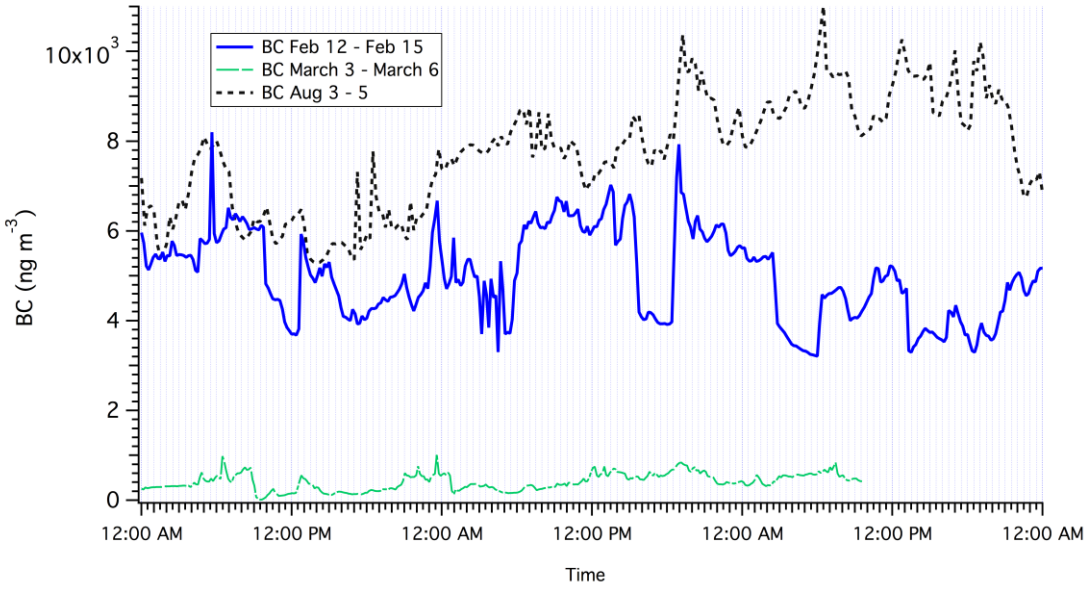

Figure 10: Case study of BC in a polluted period in February (blue line), a non-polluted period in March (green line), and a polluted period in August (dotted black line).

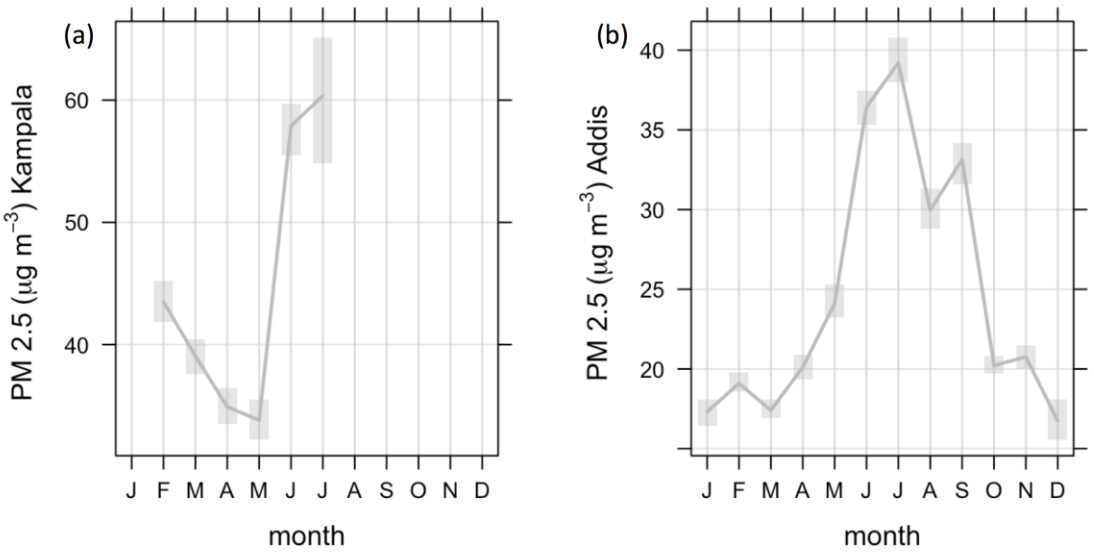

Figure 11: Monthly means of $PM_{2.5}$ concentrations measured at the US Embassies in (**a**) Kampala, Uganda (as available) and (**b**) Addis Ababa, Ethiopia (right) from January-December 2016/2017 (as available). Shaded areas are 95% confidence intervals. Lines indicate daily average WHO recommendation for healthy $PM_{2.5}$ limits.