The authors would like to thank the two reviewers for their time and effort. We have considered each point carefully and address both reviewers below. Their contributions have hopefully strengthened this paper, and we have made major revisions as advised. Author comments are in blue.

Anonymous Referee #1 Received and published: 2 March 2018 This manuscript reports on the new Rwanda Climate Observatory, an atmospheric trace gas station as part of the established AGAGE network. The station was funded by a collaboration between MIT and the government of Rwanda. The data presented in this report represent the first year and a half of the operation of the atmospheric station and focuses on BC, CO and O3. Season and diurnal plots along with air mass back trajectories are presented. This data set is valuable for the air quality community, however the data in this study is simply reported and lacks a synthesized approach to validate publication in ACP. In addition, there are erroneous claims on ozone production. At the moment, this manuscript does not represent a significant advancement in the science of air quality. I would have to recommend rejection, although I would be C1 ACPD Interactive comment Printer-friendly version Discussion paper happy to review a new submission. I have suggestions to help improve the usefulness of this publication and I outline below my major comments and identify minor issues. Reviewer general comments: The goal of the publication is unclear. What are the others hoping to achieve with this study? The title suggests the discussion of pollutants AND short-lived climate forcers, but climate implication of BC, CO or O3 are not addressed in the paper. BC, CO, and O3 are short-lived climate forcers (see IPCC report). We have added text to discuss this in the intro and the conclusion. The introduction suggests the resolution of the air quality problem in Africa, but the study focuses solely on Rwanda. The introduction has been significantly shortened; we did not mean to suggest a resolution for the entire continent. The BC time series suggest local episodic experiences, but no case studies of highly polluted days are presented. A case study has been added. The ozone diurnal at a remote site is drastically different than at urban sites, but the mechanisms for these differences are not explained. This was explained in the text and we discuss in more detail now (see specific answers to this point below). In addition, it is difficult for a mountain site to inform on the air quality in Kigali and even more so on mitigation efforts within Rwanda. The authors point out that, while a mountain site, Rwanda is highly populated with low urbanization and the mountain site was within an hour walk from a major town and near settlements. These settlements are where the majority of Rwandans live (12 million people, 1 million in Kigali) and thus the air pollution in these areas is relevant and likely somewhat representative. The authors conclude that local reduction in emissions would improve air quality in Rwanda, yet their measurements suggests that the majority of high BC and CO concentrations observed at RCO are regionally impacted. Therefore, the mitigation strategies proposed by the authors wouldn't be so effective in my opinion. Throughout the manuscript, it was acknowledged that biomass burning and regional impacts were greater than local emissions. However, Rwanda can currently only control its own emissions, and enough emissions are Rwanda's that this was suggested with many caveats. We have now added text to discuss regional fires and possible recommendations for mitigation while softening the local regulations language. Can recommendations for air quality improvement on a regional scale be made based on the presented data? Can the authors show a high pollution period with high frequency data to further support the importance and relevance of high-frequency measurements? High frequency data was used to exclude very local sources, look at diurnal profiles, showcase local spikes in pollution. A case study with the presented data would be valuable. From what I can see from Figure 9 - an interestingly high BC episode in Aug-Sept 2016 would be worth investigating. This period unfortunately does not have ozone measurements. We have chosen a different period where all instruments were working for a case study. Why don't the authors show CH4 and N2O data? Will it be part of a follow-up publication? Yes, this data will be presented in a future publication as it was a graduate student's thesis to model this data. The authors felt it was a natural split to discuss long-lived GHG species (CH4, N2O, CO2) in one paper and air pollutants in a second. Correlation plots are missing to investigate co-transported pollutants at RCO. R2 values were presented in the paper, which were derived from correlation plots. The authors chose not to show correlation plots in an effort to reduce the number of plots. When and how often is RCO within the boundary layer compared to in the free troposphere? This is now added in the methods section, Mugogo description. . An interesting study could involve measuring pollutants in Kigali and correlating them to air mass age once they reach RCO. We did not measure in Kigali and have no measurements there during the presented time period (none existed). This would be a completely separate project beyond the scope of this paper. Similar case study work has been done by (Gao et al., 2017; Zhang et al., 2015) of which, Zhang et al 2015 the authors already cite (line 433). Higher ozone precursors do not necessarily lead to higher ozone. (erroneous conclusion lines 615-624). Ozone production is not linear. Please familiarize yourselves with ozone chemistry. (examples of review references: Baier et al., 2015; Geddes et al., 2009; Monks et al., 2015)

It is not linear, no, but more precursors do have an effect on ozone production. The authors did not suggest this was a linear relationship in the text but have added a caveat 'with right meteorological conditions.' This area has high VOCs (rainforest) and likely high NOx (diesel vehicles), with no measurements of either unfortunately. So we cannot speak to NOx or VOC limited. We have also added that potentially higher solar intensity could contribute.

Finally, to further improve the manuscript, I recommend that the authors thoroughly revise their manuscript to present the information more precisely and concisely. In particular, the authors should focus on revising the syntax of their sentences. A rule of thumb I can recommend: if the sentence does not add new information, delete it.

The authors have revised the manuscript to be shorter as suggested.

I address these issues further in my specific comments. Reviewer specific comments: Title: Much of the manuscript focuses on back trajectories and I think it might be valuable to include that aspect in the title. I would also encourage the authors to specify which "air pollutants and short-lived climate forcers" they studied. Why not simply write O3 and BC? Also, there is no discussion on climatic impacts in the study.

We have discussed climate impacts further in the text.

Abstract: Line 15: The statement "air pollution is largely understudied in sub-Saharan Africa" should be supported. Why is it understudied? Because there is a lack of knowledge and expertise? A lack of funds? A lack of interest? The authors feel as if this is beyond the scope of the paper to speculate on this, as this is not scientific: it is a fact that it is largely understudied and that is now illustrated in Figure 1. Be specific. Line 23: 20% of what? Of the population? Yes, of the population. Lines 26-27: unclear that Rwanda has 4 seasons in one year (or that the two seasons represent the time since the beginning of the measurements). Rwanda does not have 4 seasons: it has two rainy and two dry seasons, as stated. Line 37: name examples of major East African capital cities We have removed this line. It is unclear within the abstract what are the major findings of the study. The authors should include quantitative data in their abstract. We have added more quantitative information as suggested. Introduction: In general, the introduction is ineffective. It is too long and too broad. The introduction could be more effective by focusing on Rwanda's air quality rather than on Africa's air quality. The introduction begins on page 3, and the first time Rwanda is mentioned is on page 6. I recommend that the authors revise those three pages on African air quality into one short paragraph of 5-6 sentences. Furthermore, I recommend introducing the AGAGE network much sooner in the introduction and mention the network in the abstract since it is the first network station in Africa!

The introduction is now much shorter and we have added more details on site selection at the end of the introduction and in the methods section.

Line 50: I would disagree that little scientific research has been performed on air quality in Africa, unless it is in comparison with the Europe and North America (which would need to be specified). I would argue that important work on air quality in Africa has been done since the 80s. (See (Stevens, 1987) as an example) The authors would argue that much has changed in Africa since the 80s. Perhaps it would be more effective for the authors to identify gaps in knowledge, rather than downplaying the existing research. The authors did not mean to offend or downplay pas research, and have tried to include as many past studies as possible. However, the authors maintain that long-term on-ground data is not prevalent on the continent, and particularly in East and equatorial Africa, except in certain countries (like South Africa). Lines 55-56: more recent references can also be included here. This has been removed Line 72: the authors say "past studies" but only reference one single study. Lines 83-85: add SAFARI campaigns (Otter et al., 2002; Swap et al., 2002b, 2002a) and Cape Point GAW station (Brunke and Scheel, 1998) This sentence has been removed as per the reviewer's request to shorten the introduction. Lines 98-101: the authors argue that long-term high-frequency measurements are important and needed, but this study focuses on monthly averages. Did the authors consider showing a case study with high frequency episodes? The authors did do an initial examination of the high frequency spikes in pollution; however, no emerging trends were found. A case study has been added. Discussion paper Lines 112-114: do the authors mean in comparison to Nairobi? Lines 120-121: add reference Lines 127-131: add reference Lines 134-137: unclear. What is meant here? Lines 144-146: missing reference Line 161: is the goal of the study really to understand air pollution in all of Africa? I recommend revising for a Rwandan context. Methods: Section 2.1: As a reader, I would be interested in knowing at the beginning of this section why RCO was chosen as the location for the AGAGE network. Was the intention to capture regional air pollution (as mentioned in the last sentence)? To sample free tropospheric air masses? This has been added In the methods section. Line 190: what checks are in place at the station to ensure the diesel generator exhaust fumes are not sampled? Very high short-lived spikes in BC were removed, and the generator was 500 m below the station. Table 1: additional columns could include minimum and maximum concentrations observed by each instrument, calibration frequencies, LODs, etc. I recommend that the authors add a data processing section in their methods. Data processing was standard, and we have added a calibration frequency column, and the minimum and maximum can be observed in the graphs. How did they quality control the data? Results and Discussion: Figure 2: why is temperature constant at the beginning of the measurement period? Appears that data quality control is incomplete for temperature and CO trace (dotted lines between gaps in data).Thank you for pointing this out, as it would be confusing to readers, we now realize. This was due to a graphing issue (the graphing program takes no data and draws a line to the next). We have split the data between gaps and graphed it separately, it was not related to the data quality control. Are the lighter colour traces averages, running averages, extrapolations? Specify. This was specified in the figure caption, they are daily averages. Figure 3: Why did the authors choose to use normalized values. Wouldn't absolute values be more meaningful to highlight and air quality problem? The authors have shown absolute values in the previous figure (figure 2). Normalized values are shown to show each pollutant on the same graph for comparisons. The authors must be consistent in their graphing - each graph has different types of error representations. Choose one and use throughout each panel. All averaged graphs have the same error, 95% confidence intervals. The authors think that different types of graphs can have errors represented different ways for clarity of graph. Figure 4: include a graph for RCO to effectively compare the three sites. Explain why data is incomplete for Uganda. 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. RCO was not included as it was shown in Figure 3 and is BC, not PM. No PM data exists publically at this time in Rwanda over time. Uganda data is not complete, as the instrument had only been running for the number of months shown (it has more data now, but this is less relevant for the data set shown). We have elaborated on the differences in Kampala and Addis in the text. (Line 268: the authors could add the WHO's lines to their plots as a graphic reference point. Lines 278-282: I believe the authors are suggesting that local air pollution is more problematic than regional air pollution? The authors agree that regional pollution/biomass burning is problematic and were not trying to suggest that it was more important than regional air pollution. However, there is some local pollution which should be explored. We can see we were not as clear as we would have liked about this issue, so we have reworked this section. Rwanda's local emissions are more controllable at this point so they were more discussed (as regulations would require one government), and these local emissions are what will grow with population increases and development, NOT biomass burning (except for slash and burn agriculture, potentially). However, their data for RCO suggests the opposite, that in fact regional air pollution elevates the background level to such high concentrations that addition cooking fires do not make a significant contribution to concentrations measured at RCO. This result might be difficult and problematic for mitigating air pollution in Rwanda. 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 bluered color bar). We have changed the color. The excellent match between FRP and BC concentrations is highly significant and should be further discussed in the paper. This comparison is striking! In the caption - do not use short/long to describe the different seasons. They are all of the same length - 3 months. We have removed. Figure 6: I have issues with the meaning of this figure. The comparison is problematic. Rwanda's bar is from RCO, a regional site whereas the comparison to other countries is an average of a number of sites throughout countries. This figure is unfortunately meaningless. BC data could be compared to other background and mountain sites - not 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. The authors believe that yearly averages show long-term pollution concentrations and RCO's context globally is interesting and relevant, but more text has been added to this section to show that these numbers are not directly comparable. The authors do not believe it is meaningless to put the RCO numbers in an international context. Figure 7: be consistent with panel readings (top to bottom) when Figure 2 is bottom up. Figure 8: Diurnal profiles are clearly not influenced by local emissions. Traffic peaks are not observed in the morning, nor in the evening. A discussion on boundary layer breakup is missing from the discussion. Also, I have never heard of a nocturnal boundary layer collapsing in the evening (lines 430-432); this explanation is wrong. This was worded incorrectly and has been removed. We have also added a discussion about boundary layer height. However, as there is a peak in the evening and morning that is distinct (and not a 'u' shape like the ozone) we do believe there is some influence from local emissions, as there is significant cooking in the valley around the station and there is high diesel generator use in the surrounding area. Rwanda is highly densely populated, so even a 'remote' mountain site is near a significant number of households. This shape persists throughout all seasons, so is not just black carbon aloft (as this would decrease in the seasons where there is less biomass burning). Figure 9: Why are running averages shown? What additional information to they provide? Discuss. The running mean is shown to show that AAE seems more seasonally/regionally, and less locally, influenced, as it is very similar to the daily averages. This text is added. Lines 476-482: show graphically, like in a pie chart. However, how important is this Rwandan information if pollution at RCO is regional? Lines 489-491: same issue as above - RCO measures regional air and so source apportionment would need to include surrounding countries' contributions. The authors feel it is important to include this information for future modelers or studies in the region. Rwanda also has good statistics on fuel use, as opposed to other nearby countries. The authors also believe that the numbers are more important than visualization in this case. We have also removed some of this discussion as per your earlier comment. Lines 569-578: show graphically Line 588: rainout is hypothesized as having an impact on the BC:CO ratio. The authors show precipitation data in Figure

9. They therefore have the information to investigate this effect accurately. Yes, we have investigated this and it is a reason. However, there is also non-local precipitation that may be affecting black carbon concentration in other areas, that may then lower the concentration of black carbon transported to the station.

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 syntax (ex. Line 50, line 111, line 278 and more). 2. When enumerating a list, all listed items must be the same type of word. Either all nouns, all verbs, etc. a. ex line 59: "are known to increase aerosol and O3 conc and to transport aerosol. . ." b. lines 105-106: "to increase. . .. And to improve. c. Lines 127-129 rewrite the listed items so they can be correctly enumerated d. Lines 175-179: revise syntax 3. Sentences longer than 2-3 lines of text need to be revised for syntax and conciseness. Specifically: Line 54: replace "certain" with "dry" Lines 56-61: syntax error - split into two sentences. (see point #2) Line 59: rephrase because aerosol fire tracers are molecular Lines 68-72: syntax error - rephrase Lines 158-159: unnecessary sentence; this message is continually repeated. Line 223: specify "regular" Lines 232-236: Move whole paragraph to the caption of the figures. Line 238: delete "it has been known for some time that"

The authors thank the reviewer for these suggestions and have addressed.

Additional references: 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. Brunke, E.-G. and Scheel, H. E.: Surface Ozone Measurements at Cape Point, in AtC8 ACPD Interactive comment Printer-friendly version Discussion paper mospheric Ozone: Proceedings of the XVIII Quadrennial Ozone Symposium L'Aquila, Italy, 12-21 September 1996, edited by R. D. Bojkov and G. Visconti, p. 7, Parco Scientifico e Tecnologici d'Abruzzo., 1998. Gao, J., Zhu, B., Xiao, H., Kang, H., Hou, X., Yin, Y., Zhang, L. and Miao, Q.: Diurnal variations and source apportionment of ozone at the summit of Mount Huang, a rural site in Eastern China, Environ. Pollut., 222, 513-522, doi:10.1016/j.envpol.2016.11.031, 2017. 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. Otter, L. B., Scholes, R. J., Dowty, P., Privette, J., Caylor, K., Ringrose, S., Mukelabai, M., Frost, P., Hanan, N., Totolo, O. and Veenendaal, E. M.: The Southern African Regional Science Initiative (SAFARI 2000)âA˘ ´r: wet season campaigns, South Afr. J. Sci., 98(3-4), 131-137, 2002. Stevens, C. S.: Ozone formation in the greater Johannesburg region, Atmospheric Environ. 1967, 21(3), 523-530, doi:10.1016/0004-6981(87)90035-7, 1987. Swap, R. J., Annegarn, H. J. and Otter, L.: Southern African

Regional Science Initiative (SAFARI 2000)âA˘ ´r: summary of science plan, South Afr. J. Sci., 98(3-4), 119-124, 2002a. Swap, R. J., Annegarn, H. J., Suttles, J. T., Haywood, J., Helmlinger, M. C., Hely, C., C9 ACPD Interactive comment Printer-friendly version Discussion paper Hobbs, P. V., Holben, B. N., Ji, J., King, M. D., Landmann, T., Maenhaut, W., Otter, L., Pak, B., Piketh, S. J., Platnick, S., Privette, J., Roy, D., Thompson, A. M., Ward, D. and Yokelson, R.: The Southern African Regional Science Initiative (SAFARI 2000)âA˘ ´r: overview of the dry season field campaign, South Afr. J. Sci., 98(3-4), 125-130, 2002b. Zhang, L., Jin, L., Zhao, T., Yin, Y., Zhu, B., Shan, Y., Guo, X., Tan, C., Gao, J. and Wang, H.: Diurnal variation of surface ozone in mountainous areas: Case study of Mt. Huang, East China, Sci. Total Environ., 538, 583-590, doi:10.1016/j.scitotenv.2015.08.096, 2015.

This manuscript presents over a year worth of measurements of air pollutants from the Rwanda Climate Observatory. This is an important dataset in an area where there are few long-term measurements. Thus, this manuscript does add important new information to our understanding of atmospheric composition in Africa and is appropriate for publishing in ACP. I would recommend major revisions before it is accepted. In general, I believe that the manuscript focuses too much on biomass burning impacts on the site and does so too early. The site and its data are important contributions to the scientific literature, however, this gets lost in the current structure of the manuscript. I think because the data and site weren't first fully explained and characterized, the analysis of the data that follows is confusing to me in parts. I believe to improve this, the manuscript does need to be reorganized and edited, and that is why I am recommending major revisions.

The authors have reorganized the paper as suggested and significantly shortened the introduction. Additionally, we have tried to balance the biomass burning versus local emission discussion to take into account both reviewer's points.

I would recommend that the paper is re-focused to firstly be on presenting the site and its measurements. As written now, the data are presented, but then quickly the focus moves to other sites and back trajectories. I would recommend that the authors first present these measurements fully and fully characterize the site. To assist with the former, I would recommend adding a table with the values of pollutants measured (e.g. annual average, seasonal averages, etc.). A case study of polluted or non-polluted events (or both) could also be helpful to understand the drivers of pollution at the site as well.

We have added a case study as suggested. We have also added a table (table 2) as suggested.

In order to help characterize the site, I would recommend showing all the data including the met data (including local wind direction and speed) in addition to the air mass history through hysplit and the other GHG measurements for completeness.

We are unsure what the reviewer means here. We do not wish to show GHG measurements, as that is in a future paper and we worry it will confuse the discussions shown here (a graduate student has written his thesis on the long-term GHG measurements and we felt it was a natural split between air pollutants and GHGs). We do show met data in Figure 2, and we have added wind direction. HYSPLIT is shown in two figures.

I would recommend adding graphs of temperature, rain and solar radiation to Figure 3. Solar radiation could be important to explaining ozone, and so would be helpful to present.

We do not have solar radiation data quality controlled at this time (there were lightning issues) and do not discuss solar radiation data in the methods section (so not sure how the reviewer knew we had solar radiation data). But we have added a reference from Safari and Gasore about solar radiation in Rwanda, as it does appear to increase in JJA vs DJF.

One thing that is not clear to me is if this site is generally within the boundary layer or not. This is key to the explanation of the diurnal cycle (e.g. ozone analysis in section 3.2.1), however it is not clear to me what/where the site is sampling. This is also important in understanding if the site is impacted by biomass and the potential impact on the measurements of this site, is not clear to me in the current biomass burning discussion in the manuscript. For example, on line 272, this peak in PM2.5 is reported in Hersey et al. (2015) is during winter and impacted by ground-level sources, and is not during biomass burning period. Figure 3 in Hersey et al. (2015) shows significant biomass burning in the region, however? (JJA, southern hemisphere winter, when there is also a peak in PM concentration). The Rwanda site is certainly impacted by biomass burning. Discussions about the site and the boundary layer have been added to the methods section.

Line-by-line recommendations I would recommend adding all the methods applied to the methods section. This includes details on Hysplit, MODIS, calculation of AAE, etc. These are currently in results as the ideas are introduced, however, I would recommend they should be in the methods section instead. We have moved HYSPLIT and MODIS discussions into the methods. Introducing AAE in the methods would be confusing in the author's opinion, as what it is feeds directly into the discussion of what it means. Starting line 251. Data from other sites are discussed in more detail before the data from the main site. I found this to be very confusing. I can see that there are few measurements in the area, so these could be helpful for comparison. However, I would recommend that they are then moved after the full presentation and analysis of the RCO data and are used to provide context. Information on the sites should be added to the methods as well. In addition, the back trajectories are discussed in 257 and not shown. It is suggested that transport occurs from southern Africa and Madagascar to Ethiopia – have others seen this? We have moved this discussion to the end of the paper as suggested, and removed part of the discussion.

Line 303 and Figure 5, this analysis is very interesting. For Figure 5, is the picture any different if you plot FRP only of the direction of the back trajectories? The point that main air flow changes during the seasons and, unfortunately for AQ, follows the biomass burning source region is very interesting. This can be seen in maps, but are the fires outside of the back trajectory direction (e.g. the Western African fires in MAM) artificially impacting the FRP Figure 5b? Also, I would recommend adding O3 and CO to Figure 5b to see their trends as well. MAM is not a period of time where BC is high, so the authors were not as concerned with this issue, additionally the FRP overall is low during this time. FRP and HYSPLIT are both inexact things, and the authors wished to convey with this figure that, when BC is high in Rwanda and the weather is dry, transport is from the major burn areas and biomass burning is high. This was not meant to be an exact comparison, as modeling or more detailed satellite measurements, beyond the scope of this paper, would be necessary for more quantitative comparisons (line 303). BC traces the best with FRP, and the authors wanted to reduce the complexity and repetitiveness of the presented graphs.

Line 303 states that MODIS is used qualitatively and not quantitatively, however FRP is quantified through the MODIS fire count data, so this seems to be contradicting the statement in 303. Answered above. Also fire count does not equal emissions or intensity so should not be directly comparable to BC. Line 316, what resolution were the geographical areas re-gridded to? I assume to match the input met resolution, but would state it. Gridded to 1 degree by 1 degree, now stated. Line 335, on the size of the maps in Figure 7, it is hard to see "local sources". The maps of cwt are not at good enough resolution to feel confident in local sources. In the discussion of ozone, would meteorology play a role in the seasonal differences of ozone? For example, does solar radiation change dramatically? Does the boundary layer change? Line 440, which profiles are flatter in the figure? As they are on the same scale, it is harder to see which has a relatively flatter shape. Ozone profiles. Line 552, for the aethalometer model does it take aging into account with the apportionment? If these are biomass burning aerosols that have been transported very far, they would be aged and would look different than local BC from burning. We have added a sentence about aging. Aging typically increases AAE, but is not explicitly taken into account in the source apportionment model. Figure 2, I would recommend adding shaded bars to the figure to denote rainy and dry season. Figure 2, The light green line is easily seen online, but not in the printed version. Perhaps white or yellow may stand out better? Also, what are the dots in the CO measurements? Gaps or zeros? These were gaps, we have fixed. Figure 6, I do like the comparison to other sites, however I would find additional information on the sites helpful. Are these just one site each or an average of sites. If it is the former, t hen I would recommend adding the name. If it is the latter, then I would recommend adding how many sites per country were used. Are they are all the sites from those countries in the EPA report? Also, for the top graph, does this have the same x-axis? Where then were the Rwandan urban measurements taken? More details have been added to the text. RCO is the only data point, we have made sure to highlight this in the text. Figure 7, looking at the maps, many of the concentration-weighted trajectories appear to me to be in the ocean (e.g. JJA (ozone esp. shows this), SON), though that is not the conclusion in the text. It is not clear to me why the highest concentrations would be over the ocean in this analysis. Also, in southern Africa, burning moves south as the season goes on (as shown in Figure 5). So the trajectories for JJA (in Figure 7) if they are coming from quite far south, then moving over the ocean to come back to land and that is where they have the highest concentrations, don't seem like they would cross the main burning areas to me. Ozone has been removed from this figure: ozone is formed in the atmosphere, thus source apportionment not exact. Madagascar has a number of fires, and that is likely the reason for the BC over the ocean. The authors believe that regional fire is impacting Rwanda and equatorial Africa in a significant way, and that the data shows this. Pollution is mixing in that region, especially during major burn periods, so exact identification of sources during wide-spread biomass burning episodes is difficult.

typos Line 82, I believe "later" should be "larger" Line 427, should read "...at RCO is different. . ."

Changed or removed

Seasonal and diurnal variability in air pollutants and short-lived climate forcers measured at the Rwanda Climate Observatory

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Abstract

Air pollution is still largely unstudied 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, 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, which coincide with the two biomass burning seasons, are higher at Mt. Mugogo than in major European cities with daily averages of 5 μg m^{-3}. 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 season, 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 that deserves immediate attention.

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 (Figure 1). The WHO reports that one in eight premature deaths globally can be linked currently to poor air quality, and these deaths are concentrated in developing countries (WHO, 2013). Black carbon (BC) is one of the major air pollutants emitted from Africa,

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mainly from biomass burning as it is widespread on the continent during certain seasons. In addition to affecting health, BC contributes to atmospheric heating and thus to climate change(Ramanathan and Carmichael, 2008). Widespread crop fires in northern and southern Africa, prevalent in boreal winter (December-January-February, DJF) and astral winter (June-July-August, JJA), respectively, are known to increase aerosol and ozone concentrations in this region and transported molecular and aerosol fire tracers associated with elevated ozone have been measured as far as the Pacific and Indian Oceans (Field et al., 2016; Real et al., 2010).

Rwanda is located in the middle of the two major seasonal biomass burning regions of sub-Saharan Africa. Wide-scale biomass burning occurs to the north of Rwanda during December-January-February (DJF) and to the south during June-July-August (JJA). Rwanda's climate may exacerbate fire haze pollution effects, as Rwanda experiences two dry seasons that occur at the same time as these two continental burning seasons, making long range transport with low rainout efficiency likely. Rwanda's prevalent wind direction also changes from northerly (DJF) to southerly (JJA) at the same time as the large-scale biomass burning area shifts from north-central Africa to southern Africa. Increase in incidence and amount of biomass burning is thought to be one consequence of climate change in this region (Niang et al., 2014). Southern Africa's biomass burning is also influenced significantly by human activity, not just the climate (Archibald et al., 2010). Rwanda is positioned to experience both large-scale (transported) haze due to fires and human activities and local, diffuse emissions.

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

In order to advance our scientific understanding of air pollution, climate change, and their impacts in Africa through generation of on-the-ground data, MIT and the government of Rwanda have established the Rwanda Climate Observatory (RCO) to measure long-lived greenhouse gases and short-lived climate forcers/pollutants in East Africa. Since May 2015, CH_4, CO, CO_2, O_3, and BC concentrations have been continuously measured, and N_2O measurements were 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, including government interest from Rwanda and willingness to take on station maintenance, Rwanda's interest in growing its technical sector, specifically focused on green growth,

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Rwanda's high and mountainous terrain, which means that stations located in Rwanda could measure pollution transported from the East and Central Africa region, 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, and discuss variations and air pollution sources. This dataset is unique and unprecedented to the region and this information on overall concentrations, sources, and time-dependent concentration variations of these air pollutants is essential in this rapidly changing area of the world to not only advance our understanding of air pollution and climate change in the region but also 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, 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.

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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, and approximately 36% below 2 km (from HYSPLIT analysis). During mid-day, Mugogo's summit is likely within the regional polluted boundary layer, but at other times of the day it is above. Complicating this issue is the network of farms and houses along the mountainside near Mt. Mugogo.

2.2 Instrumentation

Details on the instruments sampling at the RCO are compiled in Table 1. PM2.5 BC (particulate matter 2.5 micrometers in diameter or less) was measured using a Magee Scientific 7-wavelength Aethalometer with dual-spot technology that is able to

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correct for filter loading artifacts (Drinovec et al., 2015). A cyclone PM2.5 impactor was installed on the inlet to remove larger particles. Air was passed through a filter once per day to collect blank data. Flow was calibrated once per year and after major instrument movement and changes, while the optical performance was calibrated with a neutral density filter kit once per year. Data was recorded every minute at a 5 liter per minute (LPM) flow rate and particles were captured on a quartz fiber filter tape. The air stream was not dried and the relatively humidity (RH) was not controlled, which could lead to increased uncertainty during periods of high relative humidity. RH recorded at the station varied by approximately 5% over the day and from 60-85% monthly, depending on the season. The 880 nm channel was used to calculate the concentration of BC.

CO mixing ratios were measured in real-time using a cavity ring-down spectrometer (G2401, Picarro, USA). Sampled, laboratory, and calibration air were dried with a Nafion drier inside an Earth Networks calibration box to increase the accuracy of the Picarro water vapor correction (Welp et al., 2013). Three NOAA-standard calibration tanks were used for calibration spanning normal ambient concentrations and calibrations were performed once per day initially to check for linearity of instrument's response (Gasore, 2018). An O_3 monitor (T400, Teledyne Advanced Pollution Instrument, USA) was used to measure O_3. Regular checks were performed using internal span and zero O_3 calibrations. Flow was calibrated two to three times per year.

Meteorological data (ambient temperature, relative humidity, pressure, wind speed, wind direction and rainfall) were collected with an automatic weather station

(WXT520, Vaisala, Finland). The weather station was attached to a fixed, hinged arm 35 m above ground level and connected to the communications tower, level with the $CO/CO_2/CH_4$ inlet, with a 2 m clearance from the tower.

3. Results and Discussion

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. Five minute data (not pictured) was used to detect very local pollution and remove influence of short-lived local fires and BC from the generator 500 m below the station. Spikes in BC concentrations that lasted for less than 15 minute with values higher than 25,000 ng m^{-3} were removed, along with corresponding CO.

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 is partially due to the efficient rainout of black carbon particles during

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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 (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, and anthropogenic emissions contain O_3 precursors that can increase O_3 formation under the right meteorological conditions.

To explore the sources of BC and CO, at the RCO, seven-day HYSPLIT back trajectories were run every 6 hours using NCEP/NCAR reanalysis meteorological data (Kalnay et al., 1996). This analysis provided insights on the approximate origin and trajectories of air masses before arriving at RCO measured at the RCO. These HYSPLIT back trajectories were separated into DJF, MAM, JJA, and SON and are shown with MODIS satellite fire count data colored by fire radiative power (FRP, $W\ m^{-2}$) (Figure 5). The MODIS fire count data and radiative power are used strictly for qualitative, not quantitative, purposes in this work. Here we observe that, as major biomass burning sites moved to the north and west in DJF, transport direction was also primarily northerly, and as biomass burning move to Southern Africa in JJA, the prevailing wind directions were also southerly. Although Rwanda itself had few large-scale fires, its geographical position and meteorology meant that it experienced

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transported fire haze from both major burn seasons. Black carbon measured at the station tracked fairly well with summed daily FRP for sub Saharan Africa (Figure 5).

To further examine pollution transport to the RCO, the HYSPLIT back trajectory geographical areas were gridded (using the R Openair package, (Carslaw and Ropkins, 2012)) and merged, using date and time, with measured BC concentrations and mixing ratios of CO to generate concentration-weighted back trajectories (cwt) for each season (more details on cwt available in (Hsu et al., 2003; Seibert et al., 1994))(Figure 6). Trajectory time in each grid and arrival time of each air mass were taken into account in this model to predict the likely source regions and emission concentrations of pollutants measured at the RCO. This was done to determine likely source regions of air pollution at the RCO by comparing arrival times of air masses to the RCO and the time series of pollutants. This method has proven fairly effective at identifying emission sources when comparing predicted emission regions to emissions inventories (Lupu and Maenhaut, 2002) and is good as a rough estimate of emission regions with no apriori information (Kabashnikov et al., 2011). This method has low computational cost and is simple to set up, both of which are important for areas with limited bandwidth or computational capacity and this method can be repeated easily by in-country scientists.

BC and CO appeared to originate from similar areas, as expected due to their overlapping sources of inefficient combustion and biomass burning. During JJA, significant BC and CO appeared to originate from southern Africa and Madagascar, as well as from local sources near the RCO. During DJF, the source of these pollutants appeared to be much closer to the RCO, as major fires in the DRC and Uganda were

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also closer to the station. Throughout the measurement period, but particularly DJF, the Lake Kivu region also appeared to be a source of BC and CO. The Lake Kivu region is densely populated and use of both cook stoves and diesel generators is common.

In addition to direct emissions of BC and CO, other emissions such as volatile organic compounds and oxides of nitrogen from biomass burning are known to affect tropospheric O_3 concentrations, as they are precursors to O_3 formation (Jaffe and Wigder, 2012; Sauvage et al., 2005). It appears that such emissions could have played a role in the observed seasonal increase in O_3 mixing ratios of approximately 20 ppb in DJF and 25 ppb in JJA above rainy season levels at the RCO. This increase of about 5 ppb O_3 during JJA versus DJF was potentially due to the mixing of biomass burning emissions with anthropogenic emissions from east African cities such as Nairobi, Dar Es Salam, and Kampala during the JJA dry season. It also could have been the result of generally higher solar radiation during the JJA season in Rwanda (Safari and Gasore, 2009). Direct source apportionment of O_3 is difficult as it formed downwind of emissions, but a mix of biomass burning and anthropogenic emissions from southern Africa could have been transported to Rwanda after photochemical aging and processing. During the DJF dry season, fires are closer to Rwanda and away from major urban areas. During June and July, a loose correlation (R=0.47 and 0.45, respectively) between O_3 mixing ratios and BC concentrations was observed, while no correlations (R=-0.04, -0.15, and 0.07) were observed in December, January, and February.

3.2 Absorption Angstrom Exponent and BC Source Apportionment

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It is important to understand the pollution emission sources in East Africa, beyond large-scale biomass burning, in order to enact policies and actions to reduce these emissions. One way scientists have estimated fuel combustion versus biomass burning BC particulate is by measuring the color of the particles (wood smoke particles have enhanced absorption in the UV, while fossil fuel combustion particles have flat absorption over all wavelengths)(Kirchstetter and Thatcher, 2012; Sandradewi et al., 2008). The Aethalometer's seven wavelengths allow measurement of the wavelength-dependent aerosol absorption and the calculation of absorption coefficients that can be used to infer the potential sources of BC aerosol (Drinovec et al., 2015; Sandradewi et al., 2008) measured. Theoretically, from the wavelength dependence of aerosol absorption, BC from fossil fuel and wood smoke can be differentiated(Sandradewi et al., 2008). Though this two-component model can provide a valuable knowledge on knowledge on source attribution of BC this model has some limitations. This model is more accurate if calibrated to local conditions as burning and aging during transport affects aerosol 's wavelength-dependent absorption(Dumka et al., 2013; Harrison et al., 2012), as different fuels and wood biomass burning creates aerosol with different radiative properties and the standard model, based on European studies, has been shown to be less applicable in developing countries (Garg et al., 2016).

From the Aethalometer data, wavelength dependence of absorption coefficients and the absorption Ångstrom exponent (AAE) were calculated and compared to literature values of biomass burning and fossil fuel combustion (Figure 7). The AAE is a dimensionless property commonly used to characterize the

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wavelength-dependent absorption of BC and gives clues on the source and/or aging of BC when compared to laboratory and other ambient studies(Chung et al., 2012; Lack and Langridge, 2013; Russell et al., 2010; Yuan et al., 2016). The AAE values assigned for the standard Aethalometer model separating the BC from biomass burning and fossil fuel combustion are two and one, respectively (where two represents an average AAE for woodsmoke of different types and ages) (Kirchstetter et al, 2004; Sandradewi et al, 2012; Drinovec et al. 2015). In this work, standard mass absorption cross-sections (MACs) for each wavelength provided by the manufacturer of the Aethalometer were used to calculate the absorption coefficient (b_{abs}) at each wavelength. For pure BC from fossil fuel, $b_{abs} \sim 1/\lambda$ and the AAE between two wavelengths (470 nm and 950 nm) is 1 using the equation $\ln(_{babs}\lambda_1/b_{abs}\lambda_2)/\ln(\lambda_2/\lambda_1)$.

The average AAE (averaged for entire measurement period between July 2015 and January 2017) was calculated to be 1.65 (+/- 0.14) at the RCO using the 470 and 950 wavelength absorption and MACs (Figure 10)(Sandradewi et al., 2008; Drinovec et al. 2015). These wavelengths were chosen as the AAE calculated from 470 and 950 is generally comparable with other literature values(Saarikoski et al., 2012). The calculated AAE values were on par with AAE calculated from measurements taken in areas heavily influenced by biomass burning (Chung et al., 2012; Lack and Langridge, 2013; Russell et al., 2010; Saleh et al., 2013; Sandradewi et al., 2008; Yuan et al., 2016). Past studies have reported an AAE of 1.2-2.5 for biomass burning aerosol(Andreae and Gelencsér, 2006; Chung et al., 2012; Russell et al., 2010; Saleh et al., 2013, 2014). While daily only small variations (+/- 0.05) for AAE were observed (, significant seasonal differences in this value were found, with monthly averaged

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values ranging from 1.5 (dry season) to 1.9 (at the end of the long rainy season). This is shown with the 30 day running mean of the AAE (Figure 7). Studies in southern Africa measuring savanna and crop burning found an AAE of around 1.45 for ambient black carbon aerosol, and in the dry season savanna and crop burning are the prevalent type of large-scale biomass burning in sub-Saharan Africa (Russell et al., 2010). The AAE calculated from the Aethalometer data at the RCO was higher during the rainy season when local emissions dominated our measurements (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;

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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.

In order to gain more insights into the sources of BC we also examined the BC:CO. CO is also released by inefficient combustion and the ΔBC: ΔCO ratio can be different for different emission sources. In order to calculate this ratio we first converted the CO mixing ratios to concentrations (in μg m^{-3}), and then subtracted the 95^{th} percentile values for CO and BC from their respective concentrations. For the entire data set, the ΔBC: ΔCO (both in μg m^{-3}) ratio was 0.014 (R^2 0.79, n = 40523). The ΔBC: ΔCO ratio varied seasonally, with monthly average peaks reaching 0.016 in December, February, and July and lows below 0.01 in April. The average ratio of 0.014 for the measurement period was almost twice as high as in biomass burning plumes sampled over West Africa in an aircraft campaign (0.0072)(Moosmüller and Chakrabarty, 2011) but on par with or lower than measurements taken during the INDOEX campaign in the Indian Ocean (Dickerson et al., 2002). A study in Germany and Mexico found a correlation between diesel vehicle use and higher BC:CO (Baumgardner et al., 2002), while other studies have also found an increased ΔBC: ΔCO during periods more influenced by biomass burning (Pan et al., 2011). A study in India found no correlation in biomass-burning and fossil fuel-influenced

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ΔBC:ΔCO air masses (Sahu et al., 2012), as there are a wide range of ratios measured from the same source (Dickerson et al., 2002; Sahu et al., 2012). The high ΔBC:ΔCO ratio at the RCO could be due to the prevalence of older diesel engines in the country, which emit more BC to CO than newer engines (Cai et al., 2013), but, as the highest value occurs during the Rwanda dry seasons and the continental biomass burning seasons , likely the ratio is governed in part by rainout as BC is more easily removed 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.

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 μg 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

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biomass burning affected seasons, are not negligible. If all BC during the rainy seasons is assumed to be local in origin (within one day of transport, as typically rain occurs each day during the rainy season), and this level remained the same throughout the year, yearly average contribution of local emissions to BC would vary between 18-100% of the total measured BC concentration at the RCO. The shoulder months of September and February have been removed from this calculation as they have both rain and biomass burning influence, but on a yearly scale, around 35% of BC concentration measured at the station could originate from local emissions. This is a high estimate as transport of BC is still possible above the boundary layer, but it is on par with previous estimates of the contribution of savanna and forest burning BC emissions versus other emission sources (Bond et al., 2013). While transported savanna, woodland, and forest fire emissions appear to have a huge effect on Rwanda's air quality, targeting local emissions could bring a measurable decrease in PM exposure of the population.

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3.3.1 Diurnal Variations in BC, CO and O_3

Diurnal variations in concentration of pollutants can provide important insights into information on local as well as regional pollution emission sources. Diurnal variations in BC concentrations, CO mixing ratios and O_3 mixing ratios observed at RCO in different seasons are shown in Figure 8. At the RCO, the O_3 mixing ratio exhibited a diurnal cycle with a peak in concentration in the evenings, steady levels through the night and a minimum during mid-day. The increase of O_3 in the evening is likely mainly regional O_3 transported above the boundary layer measured at night (as the boundary layer height lowered), but some more locally formed O_3

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

BC had mid-morning and early evening peaks that coincided with both cooking times and kerosene/generator use times and with lower boundary layer height in the mornings and evenings. Like with O_3, changing boundary layer conditions also played a role in variations in BC concentrations over the day, as local boundary layer height increased during the day and decreased during the evening and morning hours, and the RCO altitude was above the boundary layer height often during the evening. These peaks persisted throughout the rainy and dry seasons, indicating some influence of local sources for these diurnal peaks as regional transport of BC higher in the atmosphere should be greater in JJA/DJF (more BC) and solely boundary-layer driven BC concentration changes would be greater during these times. CO mixing ratios had a similar but less pronounced diurnal variation.

3.3.2 Case Study: High and Low Periods of Black Carbon

Seasonal variations are too long to fully capture local pollution events. To further examine local pollution, high BC time periods during DJF and JJA period, and one period of low black carbon in the MAM period, were examined for their BC:CO

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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 ppbb 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 Comparison to Global and Eastern Africa Measurements

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Daily averages of BC at the station often exceeded 5 μg m^{-3}, and the yearly average BC measured at the station was greater than many rural measurement locations around the globe and on-par with urban measurements in North America and Europe, though much lower than measurements made in cities in China (Figure 11). While data from other countries is from multiple years and stations, this does give context to the Rwanda measurements globally. However, more relevant comparisons would be with other areas in Eastern Africa.

Recently the US Embassies in Addis Ababa, Ethiopia, and Kampala, Uganda have begun continuously measuring PM2.5 concentrations. The raw data is collected and reported online on the OpenAQ platform (OpenAQ.org). This dataset on PM2.5 concentrations in major cities over different seasons in this region has been valuable in gaining basic insights into the seasonal characteristics of PM2.5 concentrations in the region (Figure 12). The PM2.5 concentrations in both these cities 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 PM2.5 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 a long dry season during JJA and a short dry season during DJF. Rainy and dry season extrema are shown in the available Kampala PM2.5 data, with an enhancement during February and JJA of around 15 to 25-30 $\mu g\ m^{-3}$, respectively, above PM2.5 concentrations during other months. However, in Kampala during all months measured, including the rainy season where little regional biomass burning influence is likely, monthly averages remained above the WHO recommendations for air pollution levels at daily averages of 25 $\mu g\ m^{-3}$ or less and, despite having a lower population than Addis, were consistently higher in PM2.5 concentrations. South Africa

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has the most air quality monitoring stations of any sub-Saharan African country and results from these stations show a PM2.5 peak only in the southern burning season (JJA), not surprisingly missing transported pollution from the northern (DJF) burning season (Hersey et al., 2015). From these data, though there are only two data points, it appears that African countries near the equator may be positioned to experience six months per year of transported regional fire haze, from both the northern and southern biomass burning seasons.

Beyond BC and PM3.5, the MOZAIC campaign in the late 1990s and early 2000s measured ambient O_3 mixing ratios at the Nairobi, Kampala, and Kigali airports. This campaign found Kigali, despite its smaller size and lower vehicle count, to have the highest O_3 mixing ratios among them (Sauvage et al., 2005). They measured a similar in magnitude increase in surface O_3 mixing ratios during the JJA season in Rwanda as our measurements at the RCO, although DJF was not measured in their work. O3 measurements were made in Brazzaville, Republic of the Congo during January and February O_3. While much further west than Rwanda, in Brazzaville O_3 mixing ratios also increased during January and February, parallel to Rwanda, with monthly averages during January and February 25 ppb greater than the minimum of <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

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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. 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 appeared to have a wide-reaching effect on this region, reflected in increased PM2.5 in Kampala, a major East African city, for both biomass burning seasons and likely driving the increased O_3 measured during DJF and JJA by our study and 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

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population in Rwanda and equatorial East Africa as exposure to elevated levels of PM2.5 and BC concentrations occurs six months out of the year.

2. Ground level O_3 was enhanced during both dry seasons, likely due to the prevalent wide-scale biomass burning. Increased enhancement was observed during the JJA dry season when solar intensity was higher and the air masses originated from the southeast and likely included a mix of biomass burning and anthropogenic emissions (cooking fires, vehicles, industries). As this area develops and population grows, local as well as regional air pollution could become a major environmental and societal issue that could be a threat to national development goals.

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3. 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 μg 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 and, as these local emissions are

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

4. 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.
5. The measurements we have provided in this study will be useful in advancing atmospheric science in Africa, improve emission inventories and air pollution/atmospheric models in the region, and designing mitigation measures in the region, 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

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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 this 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

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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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Observatory and the University of Rwanda for providing laboratory space and infrastructure for instrument testing. We thank Dr. Arnico Panday who provided guidance during the initial stages of this project. We also wish to acknowledge the essential contributions of the Mugogo station technical experts Theobard Habineza, Modeste Mugabo, Olivier Shyaka, and Gaston Munyampundu, and RBA technician Yves Fidele, without which running this station would be impossible.

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

INSTRUMENT	SPECIES MEASURED	MEASUREMENT PERIOD	TIME RESOLUTION
PICARRO G2401 CAVITY RING DOWN SPECTROMETER	CO_2, CO, CH_4, H_2O	MAY 2015-JANUARY 2017	1 MIN
MAGEE SCIENTIFIC AE33 7-WAVELENGTH AETHALOMETER	BLACK CARBON (PM2.5, CYCLONE IMPACTOR ON INLET)	MAY 2015-JANUARY 2017	1 MIN
TELEDYNE T400 API	O_3	MAY 2015-JANUARY 2017	1 MIN
VAISALA WXT	MET PARAMETERS (RH, WS, WD, T, P)	JULY 2015-JANUARY 2017	1S

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

References

Andreae, M. O. and Gelencsér, A.: Black carbon or brown carbon? The nature of light-absorbing carbonaceous aerosols, Atmos. Chem. Phys., 6(3), 3419–3463, doi:10.5194/acpd-6-3419-2006, 2006.

Archibald, S., Nickless, A., Govender, N., RJ., S. and Lehsten, V.: Climate and the inter-annual variability of fire in southern Africa: a meta-analysis using long-term field data and satellite-derived burnt area data, Glob. Ecol. Biogeogr., 19(6), 794–809, 2010.

Baumgardner, D., Raga, G., Peralta, O., Rosas, I., Castro, T., Kuhlbusch, T., John, A. and Petzold, A.: Diagnosing black carbon trends in large urban areas using carbon monoxide measurements, J. Geophys. Res. Atmos., 107(21), doi:10.1029/2001JD000626, 2002.

Bond, T. C., Doherty, S. J., Fahey, D. W., Forster, P. M., Berntsen, T., DeAngelo, B. J., Flanner, M. G., Ghan, S., Kärcher, B., Koch, D., Kinne, S., Kondo, Y., Quinn, P. K., Sarofim, M. C., Schultz, M. G., Schulz, M., Venkataraman, C., Zhang, H., Zhang, S., Bellouin, N., Guttikunda, S. K., Hopke, P. K., Jacobson, M. Z., Kaiser, J. W., Klimont, Z., Lohmann, U., Schwarz, J. P., Shindell, D., Storelvmo, T., Warren, S. G. and Zender, C. S.: Bounding the role of black carbon in the climate system: A scientific assessment, J. Geophys. Res. Atmos., 118(11), 5380–5552, doi:10.1002/jgrd.50171, 2013.

Cai, H., Burnham, A. and Wang, M.: Updated Emission Factors of Air Pollutants from Vehicle Operations in GREET TM Using MOVES, , (September), 2013.

Carslaw, D. C. . and Ropkins, K.: The openair manual open-source tools for analysing air pollution data, King's Coll. London, 27–28(January), 287, 2012.

Chung, C. E., Kim, S. W., Lee, M., Yoon, S. C. and Lee, S.: Carbonaceous aerosol AAE inferred from in-situ aerosol measurements at the Gosan ABC super site, and the implications for brown carbon aerosol, Atmos. Chem. Phys., 12(14), 6173–6184, doi:10.5194/acp-12-6173-2012, 2012.

Crutzen, P. J. and Andreae, M.: Biomass Burning in the Tropics : Impact on Atmospheric Chemistry and Biogeochemical Cycles Estimates of Worldwide Biomass Burning, Science (80-.)., 250(4988), 1669–1678, doi:10.1126/science.250.4988.1669, 1990.

Dickerson, R. R., Andreae, M. O., Campos, T., Mayol-Bracero, O. L., Neusuess, C. and Streets, D. G.: Analysis of black carbon and carbon monoxide observed over the Indian Ocean: Implications for emissions and photochemistry, J. Geophys. Res., 107(D19), doi:Artn 8017\rDoi 10.1029/2001jd000501, 2002.

Drinovec, L., Močnik, G., Zotter, P., Prévôt, A. S. H., Ruckstuhl, C., Coz, E., Rupakheti, M., Sciare, J., Müller, T., Wiedensohler, A. and Hansen, A. D. A.: The "dual-spot" Aethalometer: An improved measurement of aerosol black carbon with real-time loading compensation, Atmos. Meas. Tech., 8(5), 1965–1979, doi:10.5194/amt-8-1965-2015, 2015.

Dumka, U. C., Manchanda, R. K., Sinha, P. R., Sreenivasan, S., Moorthy, K. K. and Suresh Babu, S.: Temporal variability and radiative impact of black carbon aerosol over tropical urban station Hyderabad, J. Atmos. Solar-Terrestrial Phys., 105–106(April 2016), 81–90, doi:10.1016/j.jastp.2013.08.003, 2013.

Field, R. D., van der Werf, G. R., Fanin, T., Fetzer, E. J., Fuller, R., Jethva, H., Levy, R., Livesey, N. J., Luo, M., Torres, O. and Worden, H. M.: Indonesian fire activity and smoke

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Author 7/27/2018 8:27 PM
Deleted: Andela, N. and van der Werf, G. R.: Recent trends in African fires driven by cropland expansion and El Niño to La Niña transition, Nat. Clim. Chang., 4(9), 791–795, doi:10.1038/nclimate2313, 2014.

pollution in 2015 show persistent nonlinear sensitivity to El Niño-induced drought, Proc. Natl. Acad. Sci., 113(33), 9204–9209, doi:10.1073/pnas.1524888113, 2016.

Garg, S., Chandra, B. P., Sinha, V., Sarda-Esteve, R., Gros, V. and Sinha, B.: Limitation of the Use of the Absorption Angstrom Exponent for Source Apportionment of Equivalent Black Carbon: a Case Study from the North West Indo-Gangetic Plain, Environ. Sci. Technol., 50(2), 814–824, doi:10.1021/acs.est.5b03868, 2016.

Gasore, J.: Quantifying Emissions of Carbon Dioxide and Methane in Central and Eastern Africa Through High Frequency Measurements and Inverse Modeling, Massachusetts Institute of Technology., 2018.

Gatari, M. J. and Boman, J.: Black carbon and total carbon measurements at urban and rural sites in Kenya, East Africa, Atmos. Environ., 37(8), 1149–1154, doi:10.1016/S1352-2310(02)01001-4, 2003.

Harrison, R. M., Beddows, D. C. S., Hu, L. and Yin, J.: Comparison of methods for evaluation of wood smoke and estimation of UK ambient concentrations, Atmos. Chem. Phys., 12(17), 8271–8283, doi:10.5194/acp-12-8271-2012, 2012.

Hersey, S. P., Garland, R. M., Crosbie, E., Shingler, T., Sorooshian, A., Piketh, S. and Burger, R.: An overview of regional and local characteristics of aerosols in South Africa using satellite, ground, and modeling data, Atmos. Chem. Phys., 15(8), 4259–4278, doi:10.5194/acp-15-4259-2015, 2015.

Hsu, Y. K., Holsen, T. M. and Hopke, P. K.: Comparison of hybrid receptor models to locate PCB sources in Chicago, Atmos. Environ., 37(4), 545–562, doi:10.1016/S1352-2310(02)00886-5, 2003.

Kabashnikov, V. P., Chaikovsky, A. P., Kucsera, T. L. and Metelskaya, N. S.: Estimated accuracy of three common trajectory statistical methods, Atmos. Environ., 45(31), 5425–5430, doi:10.1016/j.atmosenv.2011.07.006, 2011.

Kalnay, E., Kanamitsu, M., Kistler, R., Collins, W., Deaven, D., Gandin, L., Iredell, M., Saha, S., White, G., Woollen, J., Zhu, Y., Chelliah, M., Ebisuzaki, W., Higgins, W., Janowiak, J., Mo, K. C., Ropelewski, C., Wang, J., Leetmaa, A., Reynolds, R., Jenne, R. and Joseph, D.: The NCEP/NCAR 40-year reanalysis project, Bull. Am. Meteorol. Soc., 77(3), 437–471, doi:10.1175/1520-0477(1996)077<0437:TNYRP>2.0.CO;2, 1996.

Kirchstetter, T. W. and Thatcher, T. L.: Contribution of organic carbon to wood smoke particulate matter absorption of solar radiation, Atmos. Chem. Phys., 12(14), 6067–6072, doi:10.5194/acp-12-6067-2012, 2012.

Koch, D., Schulz, M., Kinne, S., McNaughton, C., Spackman, J. R., Balkanski, Y., Bauer, S., Berntsen, T., Bond, T. C., Boucher, O., Chin, M., Clarke, A., De Luca, N., Dentener, F., Diehl, T., Dubovik, O., Easter, R., Fahey, D. W., Feichter, J., Fillmore, D., Freitag, S., Ghan, S., Ginoux, P., Gong, S., Horowitz, L., Iversen, T., Kirkevåg, A., Klimont, Z., Kondo, Y., Krol, M., Liu, X., Miller, R., Montanaro, V., Moteki, N., Myhre, G., Penner, J. E., Perlwitz, J., Pitari, G., Reddy, S., Sahu, L., Sakamoto, H., Schuster, G., Schwarz, J. P., Seland, Ø., Stier, P., Takegawa, N., Takemura, T., Textor, C., van Aardenne, J. a. and Zhao, Y.: Evaluation of black carbon estimations in global aerosol models, Atmos. Chem. Phys., 9(22), 9001–9026, doi:10.5194/acp-9-9001-2009, 2009.

Lack, D. A. and Langridge, J. M.: On the attribution of black and brown carbon light absorption using the aerosol angstrom exponent, Atmos. Chem. Phys., 13(20), 10535–10543, doi:10.5194/acp-13-10535-2013, 2013.

Lupu, A. and Maenhaut, W.: Application and comparison of two statistical trajectory

Author 7/27/2018 8:27 PM
Deleted: Food and Agriculture Organization of the United Nations: What woodfuels can do to mitigate climate change, FAO For. Pap., 98, 2010. ... [15]

Author 7/27/2018 8:27 PM
Deleted: IHME: GBD Compare Data Visualization, Inst. Heal. Metrics Eval. Seattle, WA IHME, Univ. Washingt., 2016.

Author 7/27/2018 8:27 PM
Deleted: Kinney, P. L., Gichuru, M. G., Volavka-Close, N., Ngo, N., Ndiba, P. K., Law, A., Gachanja, A., Gaita, S. M., Chillrud, S. N. and Sclar, E.: Traffic impacts on PM2.5 air quality in Nairobi, Kenya, Environ. Sci. Policy, 14(4), 369–378, doi:10.1016/j.envsci.2011.02.005, 2011.

Author 7/27/2018 8:27 PM
Deleted: Knippertz, P., Coe, H., Chiu, J. C., Evans, M. J., Fink, A. H., Kalthoff, N., Liousse, C., Mari, C., Allan, R. P., Brooks, B., Danour, S., Flamant, C., Jegede, O. O., Lohou, F. and Marsham, J. H.: The dacciwa project: Dynamics-aerosol-chemistry-cloud interactions in West Africa, Bull. Am. Meteorol. Soc., 96(9), 1451–1460, doi:10.1175/BAMS-D-14-00108.1, 2015.

Author 7/27/2018 8:27 PM
Deleted: Kuik, F., Lauer, A., Beukes, J. P., Zyl, P. G. Van, Josipovic, M., Vakkari, V., Laakso, L. and Feig, G. T.: The anthropogenic contribution to atmospheric black carbon concentrations in southern Africa : a WRF-Chem modeling study, , 8809–8830, doi:10.5194/acp-15-8809-2015, 2015.

Author 7/27/2018 8:27 PM
Deleted: Liousse, C., Assamoi, E., Criqui, P., Granier, C. and Rosset, R.: Explosive growth in African combustion emissions from 2005 to 2030, Environ. Res. Lett., 9(3), 35003, doi:10.1088/1748-9326/9/3/035003, 2014.

techniques for identification of source regions of atmospheric aerosol species, Atmos. Environ., 36(36–37), 5607–5618, doi:10.1016/S1352-2310(02)00697-0, 2002.

Mkoma, S. L., Maenhaut, W., Chi, X., Wang, W. and Raes, N.: Characterisation of PM10 atmospheric aerosols for the wet season 2005 at two sites in East Africa, Atmos. Environ., 43(3), 631–639, doi:10.1016/j.atmosenv.2008.10.008, 2009.

Moosmüller, H. and Chakrabarty, R. K.: Technical Note: Simple analytical relationships between Ångström coefficients of aerosol extinction, scattering, absorption, and single scattering albedo, Atmos. Chem. Phys., 11(20), 10677–10680, doi:10.5194/acp-11-10677-2011, 2011.

Niang, I., Ruppel, O. C., Abdrabo, M. A., Essel, A., Lennard, C., Padgham, J. and Urquhart, P.: Africa, Clim. Chang. 2014 Impacts, Adapt. Vulnerability - Contrib. Work. Gr. II to Fifth Assess. Rep. Intergov. Panel Clim. Chang., 1199–1265, doi:10.1017/CBO9781107415386.002, 2014.

Pan, X. L., Kanaya, Y., Wang, Z. F., Liu, Y., Pochanart, P., Akimoto, H., Sun, Y. L., Dong, H. B., Li, J., Irie, H. and Takigawa, M.: Correlation of black carbon aerosol and carbon monoxide in the high-altitude environment of Mt. Huang in Eastern China, Atmos. Chem. Phys., 11(18), 9735–9747, doi:10.5194/acp-11-9735-2011, 2011.

Prinn, R. G., Weiss, R. F., Fraser, P. J., Simmonds, P. G., Cunnold, D. M., Alyea, F. N., O'Doherty, S., Salameh, P., Miller, B. R., Huang, J., Wang, R. H. J., Hartley, D. E., Harth, C., Steele, L. P., Sturrock, G., Midgley, P. M. and McCulloch, A.: A history of chemically and radiatively important gases in air deduced from ALE/GAGE/AGAGE, J. Geophys. Res. Atmos., 105(D14), 17751–17792, doi:10.1029/2000JD900141, 2000.

Ramanathan, V. and Carmichael, G.: Global and regional climate changes due to black carbon, Nat. Geosci., 1, 221–227, doi:10.1038/ngeo156, 2008.

Real, E., Orlandi, E., Law, K. S., Fierli, F., Josset, D., Cairo, F., Schlager, H., Borrmann, S., Kunkel, D., Volk, C. M., McQuaid, J. B., Stewart, D. J., Lee, J., Lewis, A. C., Hopkins, J. R., Ravegnani, F., Ulanovski, A. and Liousse, C.: Cross-hemispheric transport of central African biomass burning pollutants: Implications for downwind ozone production, Atmos. Chem. Phys., 10(6), 3027–3046, doi:10.5194/acpd-9-17385-2009, 2010.

Russell, P. B., Bergstrom, R. W., Shinozuka, Y., Clarke, a. D., DeCarlo, P. F., Jimenez, J. L., Livingston, J. M., Redemann, J., Holben, B., Dubovik, O. and Strawa, A.: Absorption Angstrom Exponent in AERONET and related data as an indicator of aerosol composition, Atmos. Chem. Phys., 10, 1156–1169, doi:10.5194/acpd-9-21785-2009, 2010.

Saarikoski, S., Carbone, S., Decesari, S., Giulianelli, L., Angelini, F., Canagaratna, M., Ng, N. L., Trimborn, a., Facchini, M. C., Fuzzi, S., Hillamo, R. and Worsnop, D.: Chemical characterization of springtime submicrometer aerosol in Po Valley, Italy, Atmos. Chem. Phys., 12(18), 8401–8421, doi:10.5194/acp-12-8401-2012, 2012.

Safari, B. K. (University of R. and Gasore, J. (University of R.: Estimation of Global Solar Radiation in Rwanda Using Empirical Models, Asian J. Sci. Res., 2(2), 68–75, doi:10.3923/ajsr.2009.68.75, 2009.

Sahu, L. K., Kondo, Y., Moteki, N., Takegawa, N., Zhao, Y., Cubison, M. J., Jimenez, J. L., Vay, S., Diskin, G. S., Wisthaler, A., Mikoviny, T., Huey, L. G., Weinheimer, A. J. and Knapp, D. J.: Emission characteristics of black carbon in anthropogenic and biomass burning plumes over California during ARCTAS-CARB 2008, J. Geophys. Res. Atmos., 117(16), 1–20, doi:10.1029/2011JD017401, 2012.

Author 7/27/2018 8:27 PM
Deleted: Marais, E. A. and Wiedinmyer, C.: Air Quality Impact of Di ff use and Ine ffi cient Combustion Emissions in Africa (DICE-Africa), , doi:10.1021/acs.est.6b02602, 2016.

Author 7/27/2018 8:27 PM
Deleted: Ngo, N. S., Gatari, M., Yan, B., Chillrud, S. N., Bouhamam, K. and Kinney, P. L.: Occupational exposure to roadway emissions and inside informal settlements in sub-Saharan Africa: A pilot study in Nairobi, Kenya, Atmos. Environ., 111, 179–184, doi:10.1016/j.atmosenv.2015.04.008, 2015.

Author 7/27/2018 8:27 PM
Deleted: Rwanda, R. of: Energy Sector Strategic Plan: Republic of Rwanda Ministry of Infrastructure, Kigali, Rwanda., 2015.

Author 7/27/2018 8:27 PM
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Saleh, R., Hennigan, C. J., McMeeking, G. R., Chuang, W. K., Robinson, E. S., Coe, H., Donahue, N. M. and Robinson, A. L.: Absorptivity of brown carbon in fresh and photo-chemically aged biomass-burning emissions, Atmos. Chem. Phys., 13(15), 7683–7693, doi:10.5194/acp-13-7683-2013, 2013.

Saleh, R., Robinson, E. S., Tkacik, D. S., Ahern, A. T., Liu, S., Aiken, A. C., Sullivan, R. C., Presto, A. a., Dubey, M. K., Yokelson, R. J., Donahue, N. M. and Robinson, A. L.: Brownness of organics in aerosols from biomass burning linked to their black carbon content, Nat. Geosci., 7(September), 1–4, doi:10.1038/ngeo2220, 2014.

Sandradewi, J., Prévôt, A. S. H., Szidat, S., Perron, N., Alfarra, M. R., Lanz, V. A., Weingartner, E. and Baltensperger, U.: Using Aerosol Light Absorption Measurements for the Quantitative Determination of Wood Burning and Traffic Emission Contributions to Particulate Matter, Environ. Sci. Technol., 42(9), 3316–3323, doi:10.1021/es702253m, 2008.

Sauvage, B., Thouret, V., Cammas, J. P., Gheusi, F., Athier, G. and Nédélec, P.: Tropospheric ozone over Equatorial Africa: regional aspects from the MOZAIC data, Atmos. Chem. Phys., 5, 311–335, doi:10.5194/acpd-4-3285-2004, 2005.

Seibert, P., Kromp-Kolb, H., Baltensperger, U., Jost, D. T. and Schwikowski, M.: Trajectory Analysis of High-Alpine Air Pollution Data, in Air Pollution Modeling and Its Application: NATO: Challenges of Modern Society, edited by S.-E. (Riso N. L. Gryning and M. M. (Centre for E. S. of the M. Millan, pp. 595–596, Springer USA., 1994.

Thompson, A. M., Diab, R. D., Bodeker, G. E., Zunckel, M., Coetzee, G. J. R., Archer, C. B., Mcnamara, D. P., Pickering, K. E., Combrink, J., Fishman, J. and Nganga, D.: Ozone over southern Africa during SAFARI-92 / TRACE A, , 101(95), 1996.

van Vliet, E. D. S. and Kinney, P. L.: Impacts of roadway emissions on urban particulate matter concentrations in sub-Saharan Africa: new evidence from Nairobi, Kenya, Environ. Res. Lett., 2(4), 45028, doi:10.1088/1748-9326/2/4/045028, 2007.

Welp, L. R., Keeling, R. F., Weiss, R. F., Paplawsky, W. and Heckman, S.: Design and performance of a Nafion dryer for continuous operation at CO2and CH4 air monitoring sites, Atmos. Meas. Tech., 6(5), 1217–1226, doi:10.5194/amt-6-1217-2013, 2013.

WHO: Health Effects of Particulate Matter: Policy imlications for countries in eastern Europe, Caucasus and central Asia, World Heal. Organ., 15 [online] Available from: www.euro.who.int, 2013.

Yuan, J. F., Huang, X. F., Cao, L. M., Cui, J., Zhu, Q., Huang, C. N., Lan, Z. J. and He, L. Y.: Light absorption of brown carbon aerosol in the PRD region of China, Atmos. Chem. Phys., 16(3), 1433–1443, doi:10.5194/acp-16-1433-2016, 2016.

Zhang, L., Jin, L., Zhao, T., Yin, Y., Zhu, B., Shan, Y., Guo, X., Tan, C., Gao, J. and Wang, H.: Diurnal variation of surface ozone in mountainous areas: Case study of Mt. Huang, East China, Sci. Total Environ., 538, 583–590, doi:10.1016/j.scitotenv.2015.08.096, 2015.

Author 7/27/2018 8:27 PM

Deleted: Tiitta, P., Vakkari, V., Croteau, P., Beukes, J. P., Van Zyl, P. G., Josipovic, M., Venter, A. D., Jaars, K., Pienaar, J. J., Ng, N. L., Canagaratna, M. R., Jayne, J. T., Kermi, V. M., Kokkola, H., Kulmala, M., Laaksonen, A., Worsnop, D. R. and Laakso, L.: Chemical composition, main sources and temporal variability of PM1 aerosols in southern African grassland, Atmos. Chem. Phys., 14(4), 1909–1927, doi:10.5194/acp-14-1909-2014, 2014.

Author 7/27/2018 8:27 PM

Deleted: World Bank: World Development Report 2011: World Development Indicators, Fossil Fuel Energy Consumption., 2011.

Author 7/27/2018 8:27 PM

Deleted: Zhang, Y., Cooper, O. R., Gaudel, A., Thompson, A. M., Nédélec, P., Ogino, S. and West, J. J.: Tropospheric ozone change from 1980 to 2010 dominated by equatorward redistribution of emissions, Nat. Geosci., 9(December), 875–881, doi:10.1038/NGEO2827, 2016.

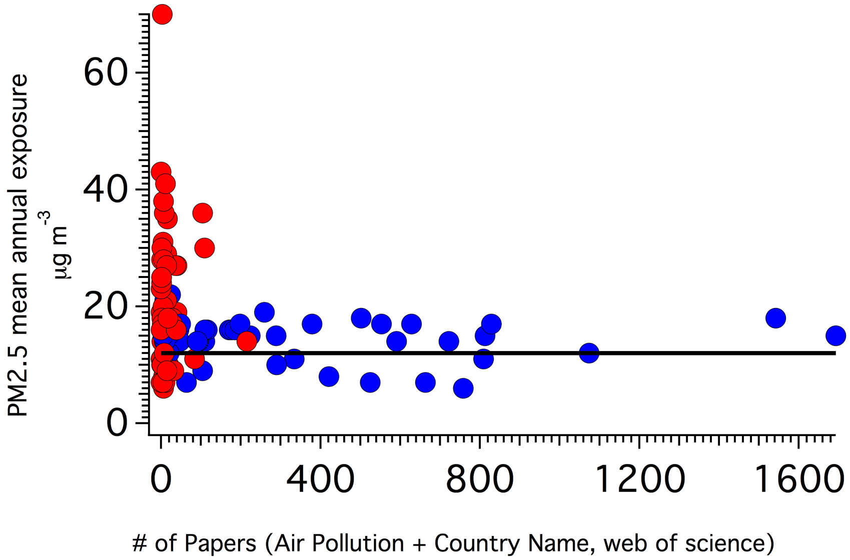

Figure 1: Africa (red) and Europe (blue), PM2.5 mean annual exposure (https://data.worldbank.org/indicator/en.atm.pm25.mc.m3) and paper count of country + air pollution.

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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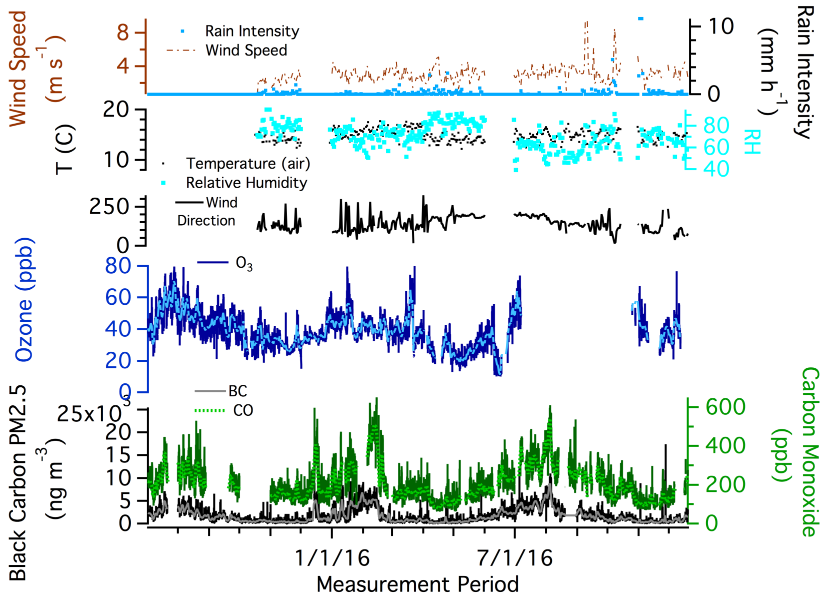

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Figure 3. From the top down up: (a) wind speed (red dotted) and rain intensity (blue dash) daily average values; (b) temperature (black) and relative humidity (light blue) values; (c) ozone (dark blue, light blue) (15 minute, daily); (d) black carbon (black, grey) and carbon monoxide (dark green, light green) (15 minute, daily) average concentrations.

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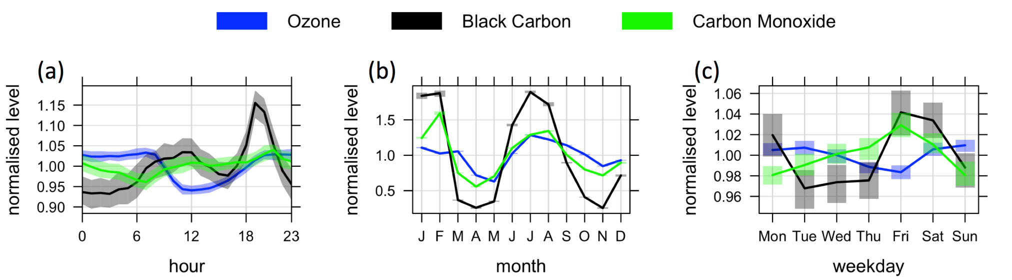

Figure 4. Normalized temporal variations of O_3 mixing ratios, CO mixing ratios, and BC concentrations: (a) diurnal (b) monthly concentrations, and (c) differences by day of the week. Shaded areas are 95% confidence intervals.

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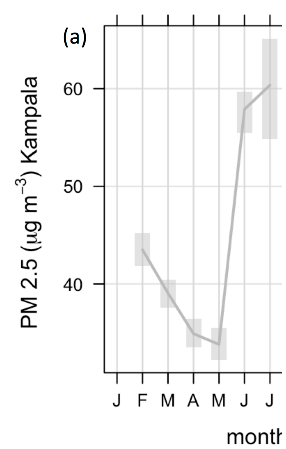

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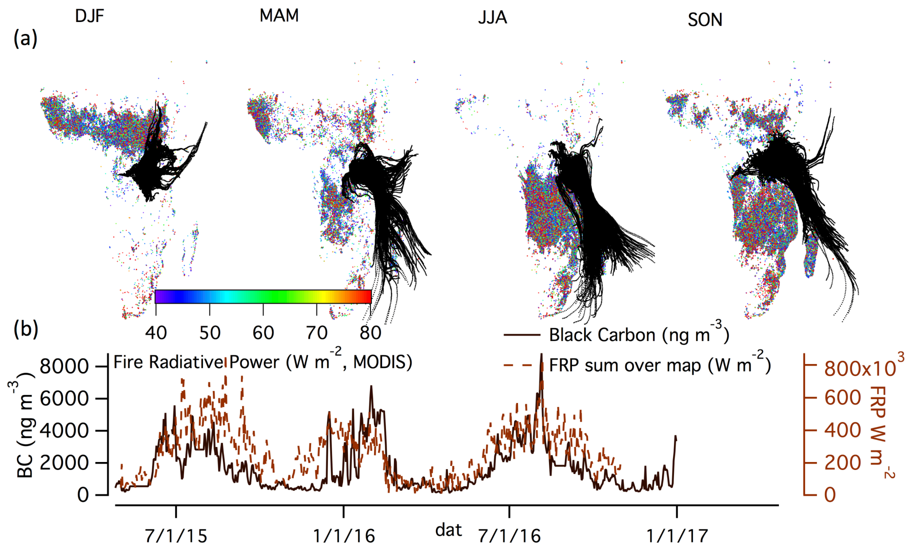

Figure 5. (a) Seasonal fire radiative power data acquired with the MODIS instrument and back trajectories of air masses (generated with the HYSPLIT model) reaching the Rwanda Climate Observatory for the period May 2015 to January 2017. Seasons in Rwanda are split into: short dry season, December-January-February (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).

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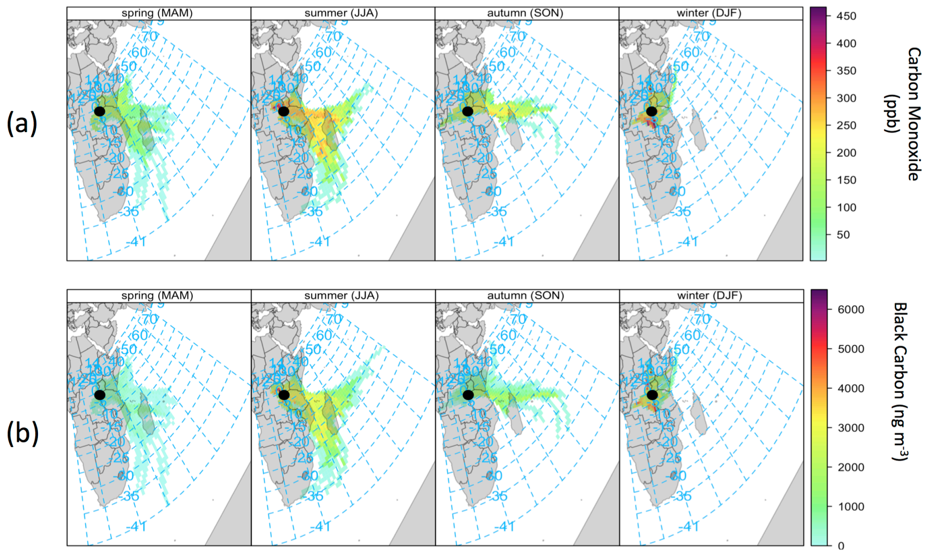

Figure6. Concentration-weighted back trajectories of (a) CO and (b) BC, separated by season, for measurements at the Rwanda Climate Observatory (black dot) for the period of July 2015-January 2017.

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Moved down [9]: (a) Urban and (b) rural maximum (dark grey/green) of annual averages (dark grey/green) and minimum (light grey/green) of annual averages (light grey/green) BC concentrations at various sites globally. The BC data for Rwanda is from one location (Mt. Mugogo, rural), while the data for other locations were from multiple locations, averaged over one year. The annual average BC concentrations for Rwanda were calculated for the data from April 1st to April 1st of the next year. There was BC data for two years measured at RCO. BC data source for other sites: https://www3.epa.gov/blackcarbon/2012report/Chapter5.pdf, compiled from multiple sources.

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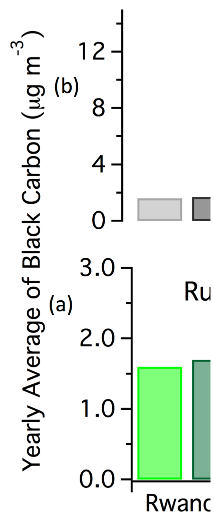

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Figure 6.

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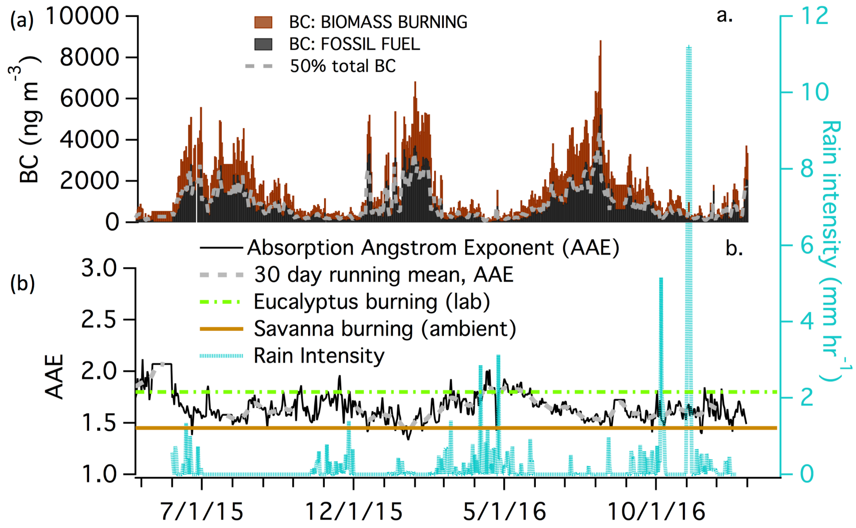

Figure 7. (a) Time series of contributions of fossil fuel combustion and biomass burning to BC concentrations observed at RCO. (b) Daily average absorption Angstrom exponent (AAE) measured at RCO (black line), rain intensity, and published AAE for Eucalyptus burning ((Yuan et al., 2016), laboratory studies,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

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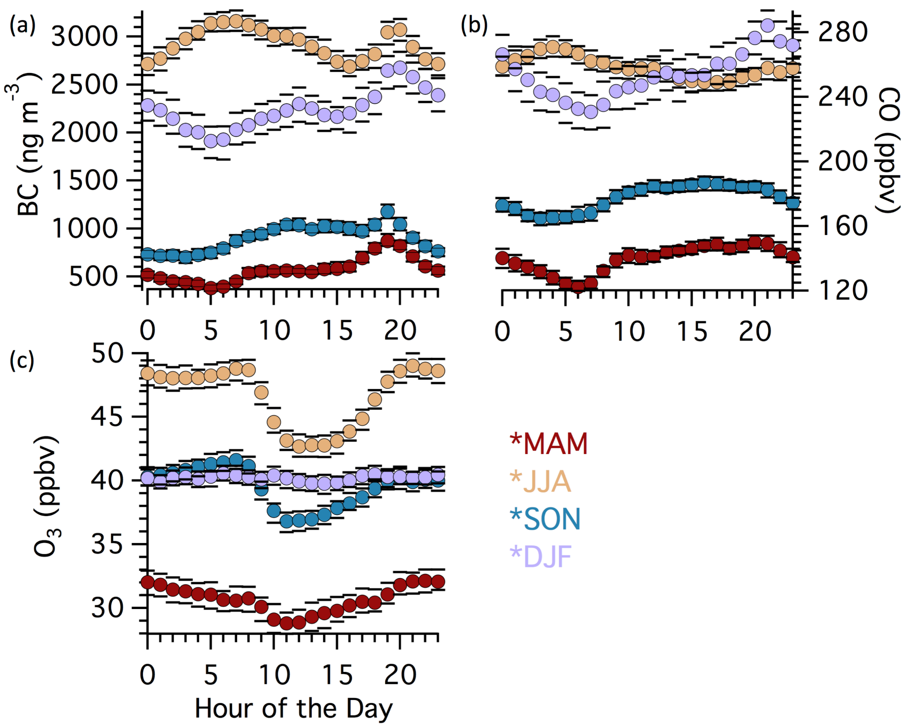

Figure 8. Seasonally separated diurnal profiles of (a) BC concentrations, (b) CO mixing ratios, and (c) O_3 mixing ratios, colored for each season. The circles represent mean concentrations and the lines represent 95% confidence intervals.

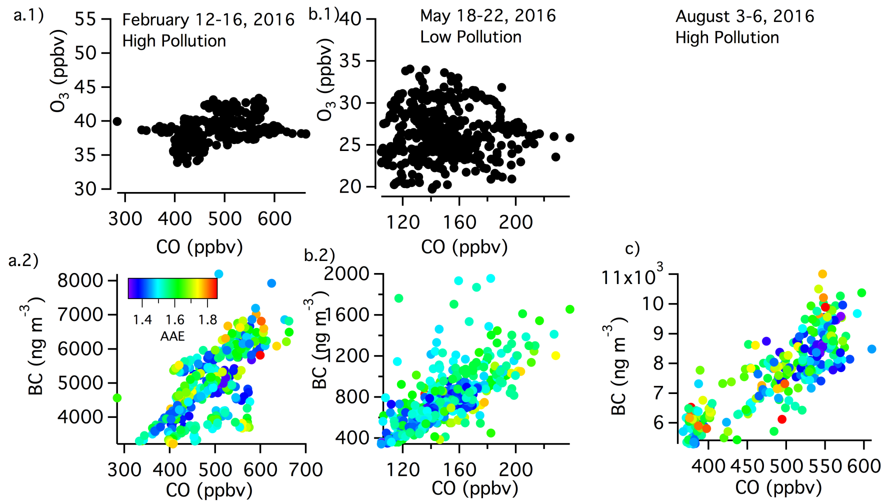

Figure 9: Polluted period in DJF (a), non-polluted period in MAM (b), and polluted period in JJA (c). Comparison of O3 and CO in a.1 and b.1, and comparison of BC and CO, color-coded by AAE, in a.2, b.2, and c for each respective period.

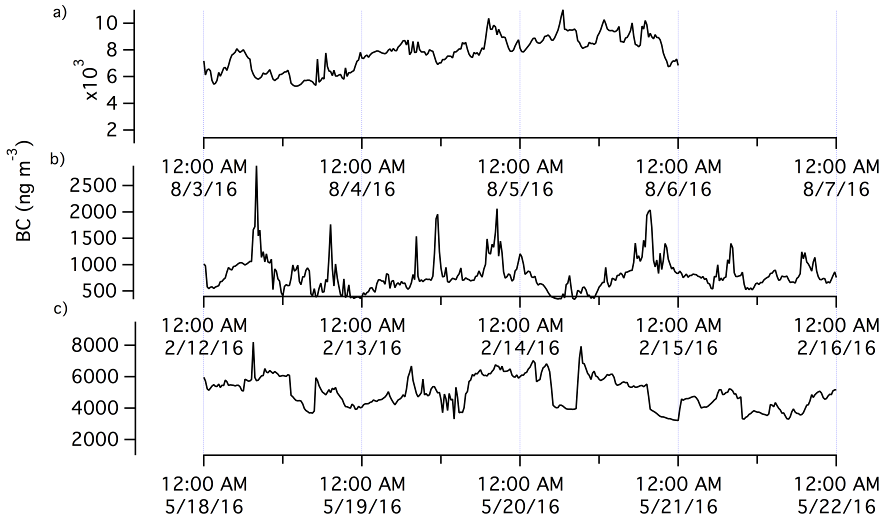

Figure 10: Case study of BC in a polluted period in August (a), a non-polluted period in May (b), and a polluted period in February (c).

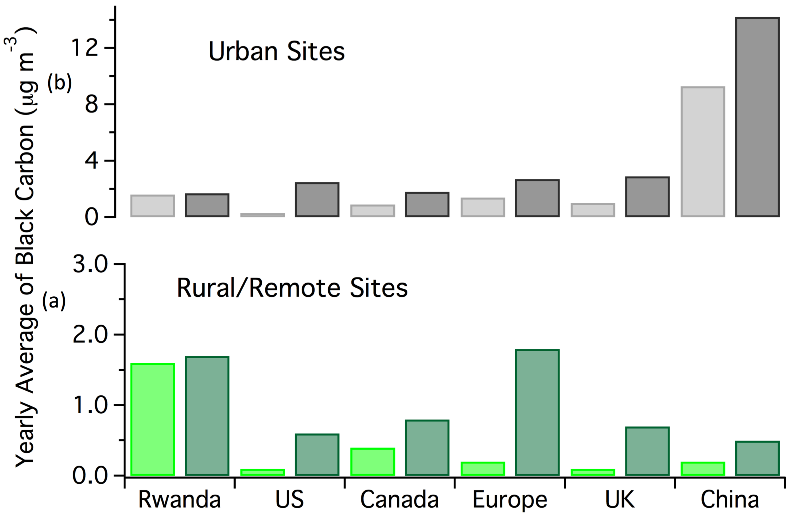

Figure 11. (a) Urban and (b) rural maximum (dark grey/green) of annual averages (dark grey/green) and minimum (light grey/green) of annual averages (light grey/green) BC concentrations at various sites globally. The BC data for Rwanda is from one location (Mt. Mugogo, rural), while the data for other locations were from multiple locations, averaged over one year. The annual average BC concentrations for Rwanda were calculated for the data from April 1st to April 1st of the next year. There was BC data for two years measured at RCO. BC data source for other sites: https://www3.epa.gov/blackcarbon/2012report/Chapter5.pdf, compiled from multiple sources.

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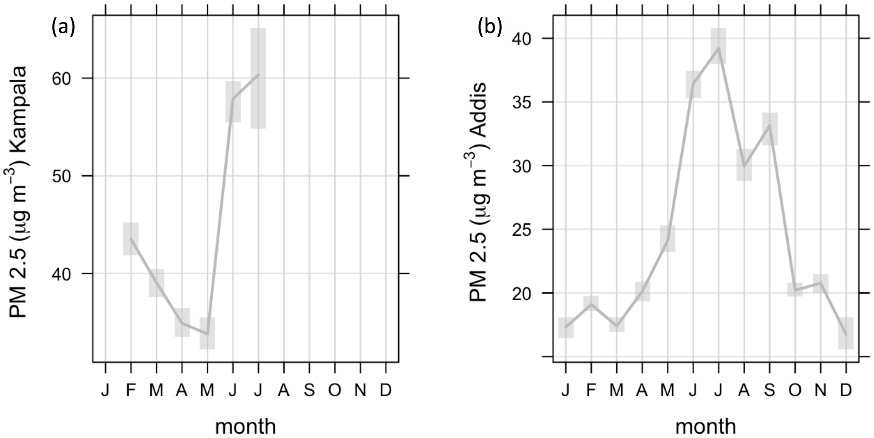

Figure 12: Monthly means of PM2.5 concentrations measured at the US Embassies in (**a**) Kampala, Uganda (as available) and (**b**) Addis Ababa, Ethiopia (right) from January-December 2016/2017 (as available). Shaded areas are 95% confidence intervals.

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