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## Impact of residential combustion on black carbon levels in Palapye, Botswana: Field measurements and GEOS-chem simulations

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## ABSTRACT

Over half of the population in Africa still rely on solid fuels such as wood, coal, dung, crop waste, and charcoal for household heating and cooking. Combustion of such fuels leads to high levels of PM<sub>2.5</sub> emissions, where a large fraction of PM<sub>2.5</sub> is composed of black carbon (BC) and organic carbon (OA). Additionally, there is a lack of continuous ground-based monitors in Africa to measure emissions and chemical composition, which is essential for monitoring changes in air quality. To better understand air pollution in Africa, we conducted a five-week field campaign in June and July of 2022 at Botswana International University of Science and Technology (BIUST) located in Palapye, Botswana and ran numerous GEOS-Chem simulations to understand which anthropogenic sources had the potential to impact the field-measured BC. BC field measurements were collected using a micro-aethalometer. Simulations were used to quantify the effect of four anthropogenic sources (energy, industry, residential, and transportation) on ambient BC in Southern Africa during June and July of 2018 and 2022. The average BC concentrations from field measurements at BIUST were 0.34  $\mu\text{g m}^{-3}$ . GEOS-Chem, simulation results showed that those residential emissions contributed 52 % and 49 % of the average ambient BC during both months in 2018 and 2022, respectively, at the observation site at BIUST. Compared to the other three combustion sources, residential emissions contributed the largest to the average ambient BC concentrations in this region.

### 1. Introduction

Air pollution causes a major burden on health for much of Africa's population (Atuyambe et al., 2024; Bililign et al., 2024; Isaxon et al., 2022; Mead et al., 2023) which is likely to get worse with the rapid unregulated industrialization, urbanization, and population growth in the region. Over 1.2 million of the 8 million premature deaths worldwide due to air pollution occur in Africa (HEI, 2024). According to one estimate, if no action is taken, outdoor air pollution alone could cause as many as 930,000 deaths across Africa in the year 2030 (UNEP, 2023).

Black carbon (BC) is a major component of particulate matter pollution (PM<sub>2.5</sub>, particulate matter with an aerodynamic diameter <2.5  $\mu\text{m}$ ) that has higher toxicity than overall PM<sub>2.5</sub> (Burnett et al., 2018; Janssen et al., 2011). BC commonly known as soot, is a

particle-phase pollutant with an atmospheric lifetime of a few days to weeks (Boucher and Reddy, 2008; IPCC and Intergovernmental Panel on Climate, 2023; Stier et al., 2007). BC is emitted from the combustion of fossil fuels (such as diesel engines) and biomass fuels (for residential heating/cooking, or from wildfires)(Bond et al., 2004; Lack et al., 2014; Ming et al., 2008; Petzold et al., 2013). Atmospheric BC is a strongly light-absorbing component of PM<sub>2.5</sub> that adversely affects air quality and human health. Exposure to PM<sub>2.5</sub>, including BC, is associated with respiratory and cardiovascular diseases, as well as premature mortality (Anenberg et al., 2011; Gordon et al., 2023; Pope and Dockery, 2006; Turner et al., 2020). BC plays a significant role in Earth's climate system by absorbing solar radiation and causing atmospheric warming, and indirectly influences cloud processes by acting as cloud condensation nuclei (CCN) and altering cloud properties and lifetimes, thus, changing

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precipitation patterns (Bond et al., 2013; Kristjánsson, 2002; Wang et al., 2011; Wilcox, 2012). When BC is deposited on snow and ice, alters the melting of snow and ice cover and their albedo (reflectivity) is reduced, accelerating the melting process (Flanner et al., 2007, 2009; Moloi et al., 2002).

Field campaigns, which aim to understand air quality and pollution sources through on-site atmospheric measurements over a set period, are most often conducted in the global North. In contrast, very few field campaigns have been conducted in Africa to study the impacts of various emissions sources across the continent (Flamant et al., 2018; Haywood et al., 2021; Knippertz et al., 2015; 2017; Petzold et al., 2013; Redemann et al., 2021). Examples of major field campaigns include the African Monsoon Multidisciplinary Analysis (AMMA) campaign over West Africa during July and August 2006; the Southern African Regional Science Initiative (SAFARI, 2000) during the dry season in southern Africa (Swap et al., 2002); and the 2016 DACCIWA campaign in Southern West Africa (SWA) (Flamant et al., 2018; Knippertz et al., 2015; 2017). The latter included ground and aircraft measurements to understand the chemical evolution of pollution plumes originating from the urban areas of Accra, Ghana and Lomé, Togo. Additional recent campaigns include the 2016–2018 US NASA ORACLES campaign based in coastal Namibia and São Tomé (Redemann et al., 2020) and the UK and the CLARIFY aircraft campaign based on Ascension Island (Haywood et al., 2020).

BC monitoring in African countries has been limited but is gradually increasing through targeted field campaigns and regional studies. Efforts include the Southern African Regional Science Initiative (SAFARI, 2000), which provided early insights into biomass burning impacts in southern Africa. Biomass burning in Africa—primarily from open fires such as savanna and agricultural burning—accounts for approximately 72 % of the global burned area and 52 % of total carbon emissions from biomass burning, including an estimated 60 % of global BC emissions (Ichoku, 2020; Ichoku et al., 2016; van der Werf et al., 2010). More recently, exposure to black carbon of students was reported using two real-time Portable Black Carbon Monitors (BC1060 -Aethalometer, Met One, and the USA) in Rwanda (Kalisa et al., 2023). Most recent studies used low-cost method to quantify BC in PM<sub>2.5</sub> by analyzing scattered red light from ambient particle deposits on glass fiber filters and estimating hourly ambient BC concentrations with filter tapes from beta attenuation monitors (BAMs) in three cities in sub-Saharan Africa (Addis Ababa, Accra, Abidjan) and in Pittsburg, USA (Anand et al., 2024). Despite these valuable contributions, continuous BC monitoring across much of sub-Saharan Africa remains sparse, and many regions lack long-term datasets necessary for understanding seasonal trends and source contributions. This study contributes to filling that gap by providing high-time-resolution BC data from a semi-rural location in southern Africa.

The only campaign we are aware of in the region of this study was a similar 2018 campaign at the same location in Palapye, Botswana (Lassman et al., 2020). The study by Lassman et al. involved a five-week air quality measurement campaign in Palapye, Botswana at BIUST which began in June 2018. The study tested the feasibility and reliability of low-cost air quality monitoring tools in a developing context, where traditional high-cost monitoring may be impractical like on the campus of BIUST. Lassman et al. (2020) study particularly focused on PM<sub>2.5</sub>, ammonia, and black carbon by using an Aerosol Mass and Optical Depth (AMOD) instrument, Radiello™ passive NH<sub>3</sub> adsorption cartridges, and microAeth AE51 aethalometer, respectively.

Botswana, a landlocked country in Southern Africa, has undergone significant economic development and urbanization in recent decades (Griffiths, 2024). However, this growth comes with severe environmental impacts that affect health. As Botswana's industries expand, sectors such as energy, transportation, and agriculture have become notable contributors to greenhouse gas emissions (Usiabulu 2024), and airborne PM, including BC. The increasing reliance on fossil fuels for power generation, coupled with a rising number of vehicles, has elevated carbon dioxide levels (Gür, 2022; Ortega-Ruiz et al., 2022).

Industries, particularly mining and manufacturing, release pollutants like methane and VOCs, affecting air quality and the broader environment (Friedrich and Obermeier, 1999; Zubair et al., 2021). Livestock farming and certain agricultural practices contribute to emissions of methane and nitrous oxide (Cui et al., 2024; Forabosco et al., 2017; Gerber et al., 2013; Laubach et al., 2024; Snyder et al., 2014). Understanding the sources and composition of emissions from all air pollution sources in Africa is critical for formulating effective mitigation strategies and regulations.

Here, we report the findings from the 2022 National Science Foundation (NSF) funded International Research Experience for Students (IRES) project. In the summer of 2022, a collaboration between North Carolina A&T State University (NCAT), Arizona State University, and Botswana International University of Science and Technology (BIUST) conducted a field campaign on the BIUST campus. In this study, we present the results of the 2022 field campaign measurements of BC concentrations, compare them with the 2018 campaign (Lassman et al., 2020), and use GEOS-chem model estimates over the Southern region of Africa to infer the major sources of BC that could be contributing to our BC measurements in the region. This study aims to investigate the sources, spatial variability, and temporal dynamics of BC concentrations in the region, with a particular focus on anthropogenic contributions. Specifically, the objectives are: (1) to determine whether anthropogenic activities are the dominant sources of BC and whether BC concentrations exhibit spatial heterogeneity; (2) to characterize the temporal patterns of BC concentrations; and (3) to evaluate the performance of GEOS-Chem simulations incorporating local emission inventories in reproducing field-measured BC concentrations, thereby assessing the accuracy of current inventories in representing local anthropogenic emissions.

## 2. Methods

### 2.1. Site description

The 2022 campaign measurements took place at the weather station (22.59502° S, 27.12680° E) on the campus of BIUST located in Palapye, Botswana, as shown in Fig. 1. The area where the weather station is located on the campus of BIUST is considered a rural area. Palapye, located 270 km northeast from the capital city of Gaborone, is considered the fastest growing village in Botswana with a population growth rate of 3.3 % from about 37,000 people in 2011 to 52,000 in 2022 as reported by the population census (Botswana, 2022; Statistics Botswana, 2014). This sampling site had the power requirements for sampling instruments, provided shelter for pumps and computers, was safe, and the samplers were far from large buildings that could obstruct air flow and impact sampling. The measurements were made during the southern hemisphere winter between June 22 and July 21, 2022. We also compared our data to a similar study where the microAeth AE51 aethalometer used in the Lassman et al. (2020) study was mounted on a wooden utility pole as part of a setup that included other instruments. Lassman et al. (2020) measurements were taken at a location southwest of the weather station from June 19th to July 20th in 2018 at BIUST.

BIUST campus is exposed to several local diffuse and point sources of air pollution, including those from the Palapye town, which is located 9 km north of campus. Major roads in the area experience a relatively large amount of traffic and all are two-lane roads that frequently have turning lanes at major intersections. Vehicles use a mix of fuels, either gasoline (unleaded petrol ULP 93 and 95) or diesel. Currently, there is no specific detailed public data available on the composition of the vehicle fleet in Palapye, Botswana, such as the exact proportion of petrol and diesel vehicles or the percentage of heavy-duty vehicles (HDVs). However, broader national-level data gives some insight into trends in vehicle types across Botswana. According to the Transport and Infrastructure Statistics Report for Botswana, passenger cars are the most common vehicle type (~73.8 % of registered motor vehicles), followed



**Fig. 1.** A) The location of the weather station on the BIUST campus where sampling took place (pink dot) at coordinates  $-22.59502^{\circ}$  S,  $27.12680^{\circ}$  E. B) The same location is indicated on a larger scale (right) with potential point sources: (1) Morupule State A coal-fired power plant, (2) Morupule Station B, (3) Morupule Coal mine, (4) Makoro Brick & Tile, and (5) a wastewater treatment plant. The town of Palapye is to the North of campus.

by vans (7.8 %). Most of the motor vehicles that were registered for the first time in 2021 were used vehicles (80.5 %). For instance, in 2021, there were notable volumes of passenger cars, but fewer HDVs and buses (16.8 %) compared to lighter vehicles, which generally dominate the fleet mix. In Botswana, vehicles undergo mechanical inspections to ensure road safety but do not currently face mandatory emissions testing, meaning air quality assessments often rely on studies measuring pollutants at the point of ambient exposure rather than by monitoring individual vehicle emissions. This situation could potentially affect the environmental impact of the vehicle fleet in regions like Palapye. Buildings are largely heated and cooled by wall-spanning dual-purpose units that are electrically powered and remotely controlled in individual rooms. Water heating is done electrically, but roof-mounted solar water heating units are also prevalent throughout the country (Seretse et al., 2018). Solid fuels (mainly wood and charcoal) are used for cooking and heating in the villages in the vicinity of the sampling site and emissions from those sources are the greatest at mealtimes (Ketlhoilwe and Kanene, 2018). While there was little wildfire activity in Botswana during this time period, there is often long-range wildfire smoke transport from neighboring countries to the south (South Africa), to the north (Angola and Zambia), and, to a lesser extent, the east (Zimbabwe and Mozambique) (Roberts et al., 2009). The prevailing wind is shown using an average wind-rose from 2012 to 2021 for June and July in Fig. S1.

Several point sources that are close to the sampling site are shown in Fig. 1. Close to campus, just to the northeast, is a wastewater treatment plant. There are two state-owned coal-fired power plants to the northwest - Morupule Station A (132 MW) and Station B (600 MW) (Essah and Ofetotse, 2014). These are situated near the coal mine and provide the bulk of Botswana's electricity. Their distribution network is part of the Southern African Power Pool. There are two other diesel power plants located elsewhere in the country that are only used during peak electrical demand. The Makoro Brick & Tile factory is towards the south and has its own smokestack that can be seen from campus. The Diffuse and Inefficient Combustion Emissions in Africa (DICE-Africa) is an emission inventory used to account for anthropogenic pollution in Africa in model studies. DICE-Africa is incorporated in the CEDS-GBD-MAPS however DICE-Africa inventory does not contain any current data.

## 2.2. MicroAeth AE51

In this study, equivalent black carbon (eBC) concentrations were measured using a microAeth® Model AE51 (AethLabs, San Francisco, CA, USA), a portable aethalometer that estimates BC mass concentrations based on aerosol light absorption at a wavelength of 880 nm. The instrument uses a T60 Teflon-coated borosilicate glass fiber filter tape and converts measured optical attenuation into mass concentration

using a mass absorption cross-section (MAC) of  $12.0 \text{ m}^2 \text{ g}^{-1}$  (Bond et al., 2013). The AE51 was operated at a flow rate of  $49 \text{ mL min}^{-1}$  and a sampling frequency of 1 min. It was housed in a weather station at the BIUST and operated continuously, 24 h per day, during the sampling period from 22 June to July 21, 2022. To ensure data quality, negative values resulting from brief instability in sensor response were removed, as these are not representative of environmental conditions.

Data correction methods were used to account for filter loading effects for 2022 data. Measurements were corrected using the equation  $\text{eBC}_{\text{corrected}} = \text{eBC}_{\text{non-corrected}} / (1 - k \cdot \text{ATN})$ , where ATN is the filter attenuation and  $k$  is the empirically-determined filter loading parameter (Drinovec et al., 2017). The closest analogous location was the rural background Payerne station, situated about  $\sim 1 \text{ km}$  SE of the small rural town of Payerne, Switzerland, and was surrounded by agricultural land (grassland and crops), forests, and small villages. A Summer value at 880 nm was used, where  $k = 0.0019$ .

For comparison, Lassman et al. (2020) also employed the AE51 to report eBC concentrations in 2018. In that study, the instrument operated at a higher flow rate of  $150 \text{ mL min}^{-1}$  and a 5-min sampling interval. Because the AE51 was powered by batteries, it was not run continuously; instead, it was set up intermittently throughout the day. After power cycling, the instrument often recorded negative values before stabilizing. These artifacts were acknowledged and excluded in the present analysis to maintain consistency and data integrity (Lassman et al., 2020; Petzold et al., 2013). Since attenuation values were not presented, a loading correction was not applied to this data.

Measurement of eBC using AE51 was recently evaluated (Miyakawa et al., 2020). Well-known uncertainties in filter-based methods used in AE51, are the sensitivity issue related to the particle loading effect, the air flow rate and sample spot size which is multiple-scattering effect and the positive artifact of light-scattering materials on the filter. Since the AE 51 detects light attenuation as particles accumulate on the filter, and while flow rate and sampling time indeed affect results, the relationship is not linear. A lower flow rate results in fewer particles accumulating per unit time, possibly leading to weaker light attenuation and underestimating eBC levels over short periods. We like to highlight that differences in eBC variability may also be influenced by atmospheric conditions, emission sources, or even instrumental noise in the 2018 and 2022 data. In our 2022 study, the power adaptor was used for the entire campaign, and we identify this as a limitation. The presence of instrumental noise regarding optics and electronics can lead to errors as well. Lee (2019) has proven that the direct connection of the AE51 to a power outlet continual usage of the power adaptor intensified the noise in the instrument. Even though corrections are required, many studies associated with mobile monitoring of eBC and personal exposure studies often report no correction for filter loading of the microAeth® AE51

(Buonanno et al., 2013; Rivas et al., 2016). We didn't report these errors here.

### 2.3. GEOS-chem set-up

We use GEOS-Chem version 12.6.0 (Bey et al., 2001; Leibensperger et al., 2012; Park et al., 2004) aerosol-only simulation with offline oxidation fields to simulate ambient BC in the Southernmost region of Africa. BC was assumed to be entirely in the fine mode (diameters smaller than  $2.5 \mu\text{m}$ ). Fourteen simulations were conducted for this study as outlined in Table S1. The simulations were driven by the Modern-Era Retrospective Analysis for Research and Applications, Version 2 (MERRA-2) meteorological fields from the Goddard Earth Observing System (GEOS) of the NASA Global Modeling and Assimilation Office (GMAO, <https://gmao.gsfc.nasa.gov>). First, we ran two global simulations at a horizontal resolution of  $4 \times 5^\circ$  with 47 vertical layers for the summers of 2018 and 2022 to obtain the boundary conditions for our nested simulation, with one month spin up. The advection timescale in/out of the regional domain is much less than 1-month, major BC sources are within 1000 km of Palapye, and the aerosol mass lifetime in the atmosphere is about 1 week. All our emissions sensitivity simulations with emissions from specific sectors off were within the regional domain, so 1 month adequate for adjusting to simulation changes within the region. The 12 nested simulations were conducted over the Sub-Saharan region of Africa at a resolution of  $0.5 \times 0.625^\circ$  with 47 vertical layers. We acknowledge that our spatial resolution of the nested GEOS-Chem simulations likely limits its ability to capture localized and spatially heterogeneous sources, such as village-scale biomass burning, which may contribute to the discrepancies observed between modeled and measured BC concentrations. As opposed to models with online fluid dynamics (e.g., climate models and weather models with chemistry), GEOS-Chem runs on offline meteorological fields. The native resolution of the MERRA-2 reanalysis meteorology product is  $0.5^\circ \times 0.625^\circ$ , which sets the spatial resolution of the finest resolution domain (the coarser global domain is based on a spatial averaging of the MERRA-2 product). While it is typical for regional GEOS-Chem simulations to be run at this resolution (Gong et al., 2023; Kodros et al., 2016; Wainwright et al., 2012), we understand that we will not resolve sources and spatial gradients finer than  $\sim 50$  km. This resolution likely limits its ability to capture localized and spatially heterogeneous sources, such as village-scale biomass burning, which may contribute to the discrepancies observed between modeled and measured BC concentrations. This resolution jump does mean that gradients finer than  $\sim 400$  km are not resolved at the domain edges. Most sources affecting Palapye, Botswana are either from within the nested domain, or we do not expect them to have fine spatial gradients. The east, west, and south borders of the nested domain are over the ocean. The north border is at the equator. The majority of African biomass burning in the boreal summer is in the southern hemisphere (the period of our campaigns), and we expect little cross-equator transport of pollution to Botswana.

For anthropogenic emissions from different sources (ENE: energy, IND: industry, RCO: residential and commercial, and TRA: transportation), the Community Emissions Data System GBD-Major Air Pollution Sources (CEDS-GBD\_MAPS) inventory (McDuffie et al., 2020) was used. Based on our previous comparison of open biomass burning inventories in Africa, in Gordon et al. (2023), here we used the Quick Fire Emission Data set (QFED) (Koster et al., 2018). Sea salt emissions were estimated using the Jaeglé et al. (2011) method, and the Mineral Dust Entrainment and Deposition model was used to estimate dust emissions (Zeng et al., 2019). Secondary organic aerosol formation was not modeled.

The first two nested simulations that were conducted were the base-case simulations. All emissions across the globe were turned on for each base-case simulation for 2018 and 2022 during the observation periods. The remaining ten sensitivity scenario simulations were used to estimate

the impacts of each of four anthropogenic sectors (i.e., ENE, IND, TRA, RCO), as defined by McDuffie et al. (2020), for Southern Africa (Table S1). The southern region of Africa includes the following countries: Botswana, Eswatini (Swaziland), Lesotho, Namibia, South Africa, and Zimbabwe, according to the Global Burden of Disease (GBD). In each of the sensitivity scenarios simulations, one of the four sectors was turned off across the region only. The eBC concentrations in the sensitivity simulations were then subtracted from the base-case simulation to quantify the eBC concentration contribution from each anthropogenic emissions sector in the Southern region of Africa during June and July of 2018 and 2022. As BC is a primary species only (not formed in the atmosphere), and dry- and wet-deposition sinks are first-order losses in these model simulations, the differences between the base-case simulation and the various sensitivity simulations are exact estimates of the modeled contribution of each source sector in the region to BC concentrations in the study region.

## 3. Results and discussion

### 3.1. Measured BC from MicroAeth AE51

Fig. 2 shows the difference in daily eBC concentrations from the Lassman et al. (2020) 2018 data and our 2022 data. There is a 76 % difference between average June and July eBC concentrations in 2018. In the Lassman et al. (2020) study, June 2018 eBC concentration varied between  $0.40$  and  $0.95 \mu\text{g m}^{-3}$ , while July tapered off from a high of  $0.48$  to lows of  $0.04 \mu\text{g m}^{-3}$ . In contrast, higher eBC concentrations occurred during the month of July when compared to June in 2022. The 2022 eBC concentrations peak at  $\sim 0.6 \mu\text{g m}^{-3}$  during the second to last week of June and the second week of July. Microaeth data from 2018 was far less variable, possibly due to greater time averaging. We found an average eBC concentration of  $0.36 \mu\text{g m}^{-3}$  from the 2022 field measurement at BIUST, which was 44 % lower than the average in Lassman et al. (2020).

### 3.2. GEOS-chem BC concentration over Southern Africa

Fig. 3 shows the average BC surface concentration from our base-case scenarios for the sampling period in 2018 and 2022. The base-case BC concentrations are elevated because the observation period is during the biomass burning season (Ramo et al., 2021). The highest BC concentrations appear to be in the southern Democratic Republic of the Congo (DRC), northern Angola, and eastern South Africa in both years. For context, modeled BC concentrations over northern Zambia and southern DRC in 2022 reach values exceeding  $3 \mu\text{g m}^{-3}$  (Fig. 3), which is comparable to or higher than values reported for other biomass burning regions. For instance, studies have shown peak seasonal BC concentrations of  $\sim 2\text{--}4 \mu\text{g m}^{-3}$  in central Africa (Holanda et al., 2020) and  $\sim 1\text{--}2 \mu\text{g m}^{-3}$  in parts of the Amazon and Southeast Asia during their respective fire seasons e.g., (Archer-Nicholls et al., 2015; Bond et al., 2013). From GEOS-Chem simulations, our base-cases June–July average BC surface concentration at the observation site (see section 2.1) was  $0.26 \mu\text{g m}^{-3}$  in 2018 and  $0.30 \mu\text{g m}^{-3}$  in 2022. When comparing our 2022 field measured BC to the simulated base-case BC concentrations, the GEOS-Chem simulations underestimated average surface BC concentrations with a normalized mean bias NMB of  $-0.23 \%$ .

In Fig. 4, the hourly field eBC concentrations from 2022 and GEOS-Chem simulated BC concentrations from each nested simulation are presented to understand the contribution of each source. In comparison to the model, the highest eBC concentrations occurred during the evening peaked at  $\sim 1.2 \mu\text{g m}^{-3}$ .

Even though corrections are required, many studies associated with mobile monitoring of eBC and personal exposure studies often report no correction for filter loading of the microAeth® AE51 (Buonanno et al., 2013; Rivas et al., 2016).

To further assess model performance, Fig. S2 compares the Base-Case

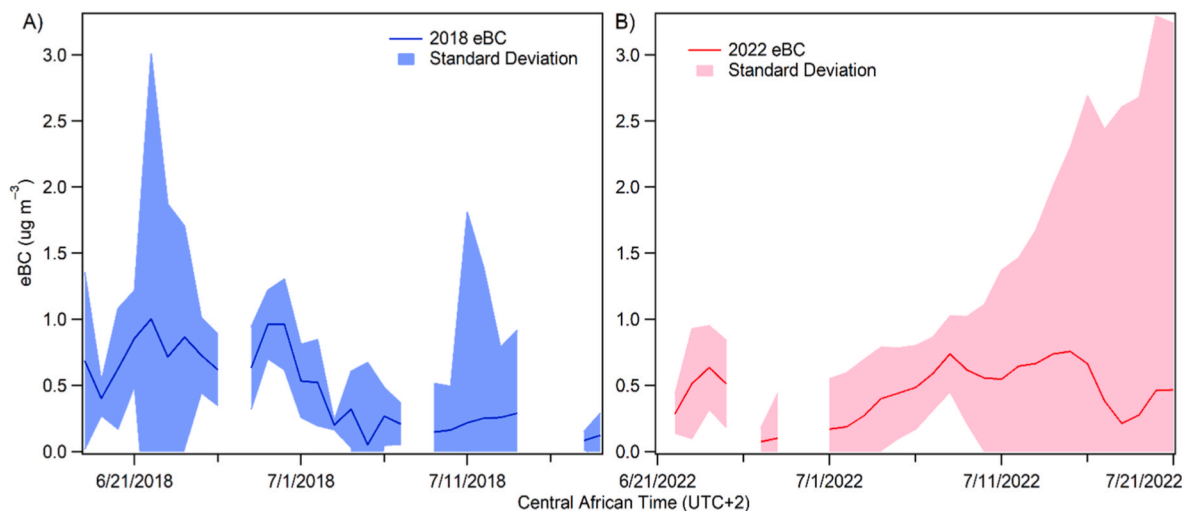


Fig. 2. A) Daily mean eBC concentrations from Lassman et al. (2020) during 2018 and B) our 2022 field study. Along with  $1\sigma$  of each measurement in blue and red, respectively.

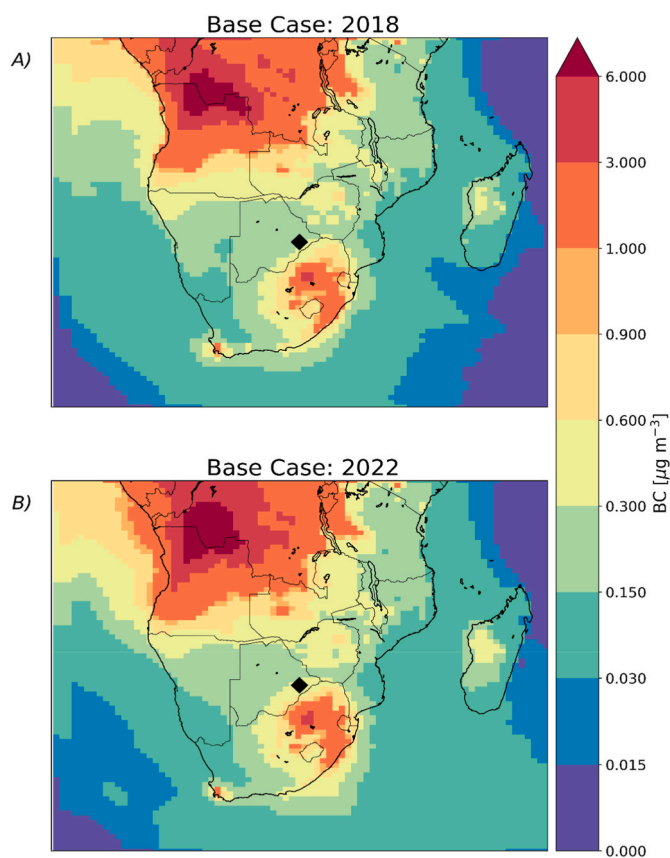


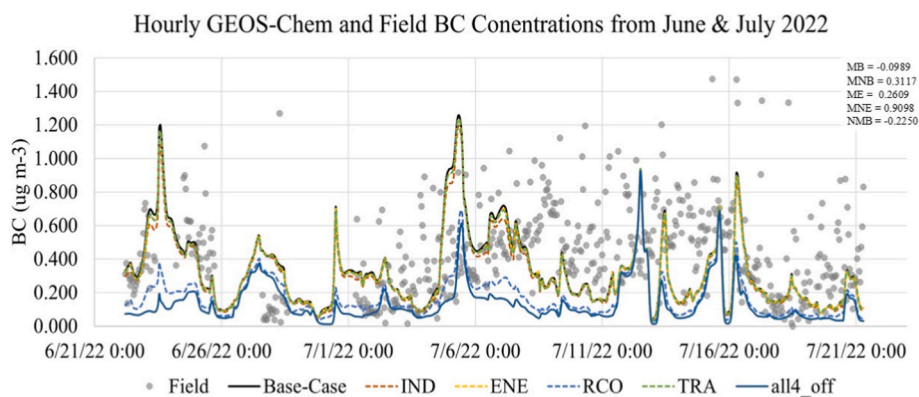
Fig. 3. GEOS-Chem BC concentration from June and July of 2018 and 2022 over Southern Africa. Base-case scenario (all emissions on) from A) 2018 and B) 2022 for both June and July of each year. The black diamond where the weather station is located on the BIUST campus where field BC measurements were collected.

simulation with the observed daily mean BC concentrations and associated standard deviation. The observed data exhibit substantial day-to-day variability, with standard deviations frequently exceeding  $1.0 \mu\text{g m}^{-3}$ . In contrast, the model output remains comparatively smooth and fails to reproduce the observed spread in concentrations. This underrepresentation of variability indicates a limited ability to resolve short-

lived, high-intensity emissions or sharp diurnal transitions in near-surface BC levels. The observed discrepancies are likely the result of several interacting factors.

First, the coarse spatial resolution of the GEOS-Chem nested domain ( $0.5^\circ \times 0.625^\circ$ ) restricts its capacity to resolve local emissions. Sub-grid sources such as cooking with solid fuels, informal waste burning, and traffic-related emissions near the monitoring site are averaged out at this resolution, contributing to the underestimation of variability and concentration levels in the model output. Second, limitations in the emissions inventories themselves likely contribute to model-observation mismatch. Inventories such as the Community Emissions Data System (CEDS) and the Global Fire Emissions Database (GFED) are known to underrepresent informal, unregulated, or seasonally variable sources—particularly in regions like Southern Africa where localized combustion activities are common but poorly documented. Third, the model's reliance on meteorological inputs from MERRA-2 introduces uncertainty in the representation of local atmospheric dynamics. Processes such as shallow nocturnal boundary layers, morning transition periods, and local recirculation patterns strongly influence BC accumulation but are difficult to capture at the resolution and physical detail provided by global reanalysis products.

From the comparisons between GEOS-Chem BC and field eBC, during the latter part of the campaign where more field measurements were taken the model seems to capture the local variations in BC concentrations. SI Fig. S3 show the correlation between daily averaged BC concentrations from GEOS-chem and 2022 field measurements with  $R^2 = 0.1633$ , Pearson coefficient =  $-0.40411$ , Spearman rho =  $-0.42247$ , and cos theta =  $0.66816$ . We have also evaluated the model using statistical metrics: mean bias (MB =  $-0.0989$ ), mean normalized bias (MNB =  $0.3117$ ), mean error (ME =  $0.2609$ ), mean normalized error (MNE =  $0.9098$ ), and normalized mean bias (NMB =  $-0.2250$ ). We highlight that the relatively poor performance of the model is a limitation of our study. When comparing each sensitivity simulation, BC concentrations obtained by removing the simulated contributions of energy and transportation sectors closely follow the base-case BC concentrations during the campaign. If we examine a specific feature in the time series, such as the spike in BC on 6/30/22, we see that there is an insignificant drop when removing industrial, energy, and transportation sources, while removing residential sources does not bring BC to zero. The remaining  $0.2 \mu\text{g m}^{-3}$  of BC may be from open biomass burning smoke transported from South Africa (See SI Fig. S4 & S5 obtained by using a NOAA HYSPLIT model backward trajectories). On the other hand, the largest contribution of BC concentrations over the observation site during this period came from residential sector.

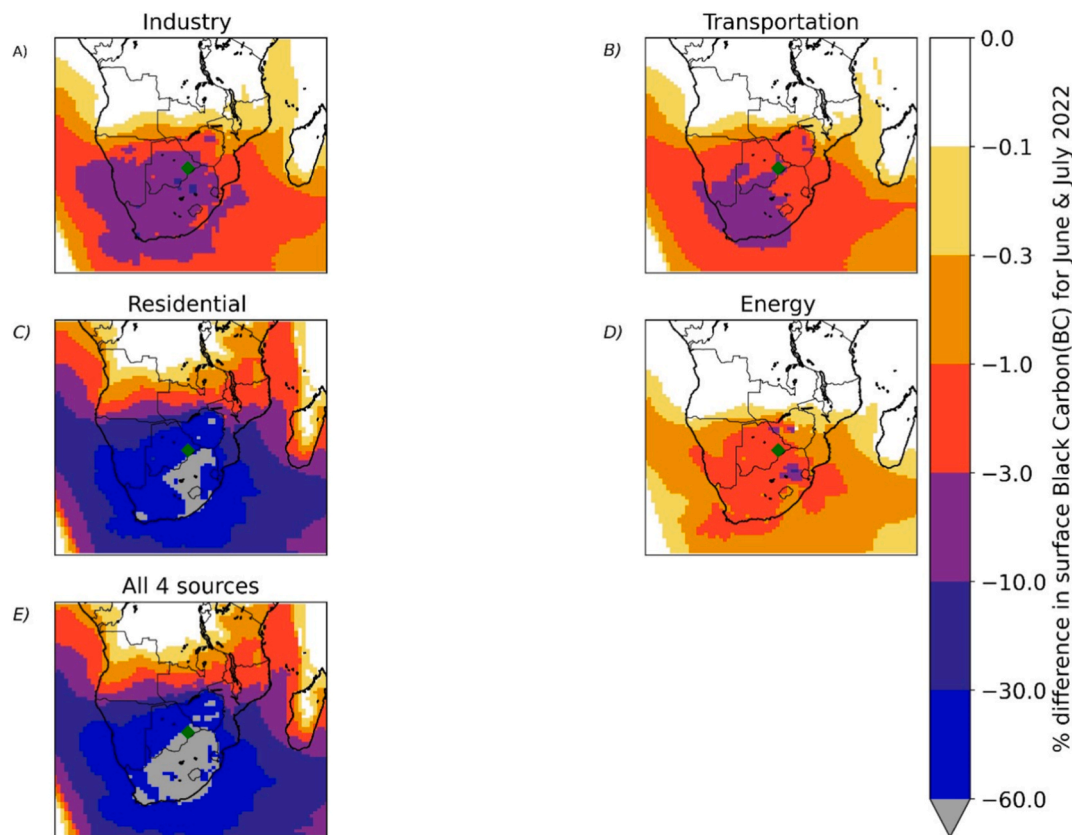


**Fig. 4.** Time series hourly eBC measurements in 2022 from the field (grey circles), BC from all GEOS-Chem anthropogenic sources (e.g., base-case (black)), and when specific GEOS-Chem anthropogenic sectors are turned off (ENE: energy, IND: industry, RCO: residential and commercial, TRA: transportation, and all4\_off: four anthropogenic sectors).

The contribution of energy, industry, residential, commercial, and transportation emissions to averaged surface BC from southern Africa is shown in SI Fig. S6 for 2018 and Fig. 5 for 2022. We evaluated the GEOS-Chem BC simulation outputs at the same latitude and longitude as the weather station to understand the potential impacts of these four anthropogenic sources on our measured BC concentrations. From the sensitivity simulations at the observational site, these four anthropogenic sources investigated contributed 64 % of surface BC concentration during June and July of 2018 and 60 % during 2022. The remaining 36–40 % of the averaged BC concentrations are from other sources, such as open biomass burning or long-range transport. These sectoral source attribution results rely on sensitivity simulations from our model, which,

despite being informative, have inherent limitations. The model's spatial resolution and the location of the sampling site at BIUST may bias the source contribution estimates by underrepresenting emissions from localized sources, such as nearby coal mining operations. Therefore, the dominant residential and commercial source influence indicated by the model should be interpreted with caution. We acknowledge that the complex emission environment and possible transport from adjacent source areas can complicate source isolation. Additional measurements at multiple sites and higher-resolution modeling, complemented by receptor-based source apportionment techniques, would help to more robustly validate and refine these sectoral contributions.

The contribution of each of the four anthropogenic sources at BIUST



**Fig. 5.** Fraction of BC attributed to A) Industry, B) Transportation, C) Residential and commercial, D) Energy, and E) all for sources in Southern Africa. Each panel (A–E) represents an individual simulation where each emission source was turned off by the region of Southern Africa for both June and July of 2022. The green diamond where the weather station is located on BIUST campus where field BC was collected.

is displayed in Fig. 6. Of the four sources, residential emissions were the largest contributor to average BC surface concentration (52 % and 49 %) at the site in both 2018 and 2022. Of the four sources investigated here, energy sources contributed the least to average ambient BC. Electricity generated in Botswana is dominated by coal power and the power stations are located northwest of the weather station (Maswabi et al., 2021). Also, Botswana depends on imported energy while less than ~20 % of the population is estimated to receive electricity from the national grid (Ofetotse and E, 2012).

According to Petkova et al. (2013), some of the major anthropogenic air pollution sources in African cities include emissions from waste and biomass burning for household and commercial needs, industrial activities, and emissions from vehicles. Industrial emissions are the second largest contributor to average ambient BC at the site for both 2018 and 2022. This source contributed about 7 % of surface BC concentration. Vehicle ownership is rapidly increasing in many African countries and is of particular concern for air quality in urban areas. Many of the newly imported vehicles are old and poorly maintained and there are very few emission standards or regulatory standards that are rarely enforced (Lagarde, 2007). The increase of such vehicles and traffic has the potential to lead to traffic-related emissions in this region (Wiston, 2017), although transportation only contributed 2 % of surface BC concentrations at the measurement site. There appears to be limited information specifically focused on source apportionment for air pollutants in Palapye, Botswana. However, one significant study carried out by Lassman et al. (2020) did focus on identifying and monitoring air quality in Palapye. The findings highlighted contributions from coal combustion at the Morupule Power Station, as well as biomass burning, which are major sources in the region, affecting PM<sub>2.5</sub> levels. However, the study primarily emphasized monitoring rather than providing detailed source apportionment analyses that distinguish precise contributions from different sources.

Here in our study, we utilized a portable eBC monitor such as the microAeth AE51 to conduct spatial and temporal assessment to study the varying levels of anthropogenic activity at the observation site at BIUST in Palapye. We implemented the GEOS-Chem model, inputting detailed local emission sources (including energy, industrial, transportation, and residential combustion) to simulate and analyze eBC levels at the observation site at BIUST in Palapye. These hypotheses and methodologies helped structure our study on the impact of anthropogenic emissions on black carbon levels in Palapye, Botswana, providing insights into local air quality.

#### 4. Conclusions

In this study, we report the real time eBC concentrations measured during the NSF-IRES field study conducted on the campus of BIUST in Palapye, Botswana in 2018 and 2022. Field measurements were compared with GEOS-Chem model output over the observation site, and the results show that residential emissions may be the main source BC

concentrations collected by the microAeth AE51 which is also corroborated by the Lassman et al. (2020) study conducted at the same site. We found that there was a 26 % decrease in average ambient BC on the campus of BIUST between the years of 2018 and 2022 during the southern hemisphere winter months (June and July), though longer-term sampling would be necessary to establish trends. The study compares eBC concentrations from Lassman et al. (2020) in 2018 and newly collected data from 2022, highlighting notable differences in seasonal peaks and overall averages. Our key findings in 2018, eBC peaked around late June and early July, while in 2022, the peaks were observed during mid to late June and early July. The average eBC concentration in 2022 was 44 % lower than in 2018. 2018 data showed less variability, likely due to greater time averaging. The proximity of the residential areas to the observation site suggested potential local sources of eBC. Comparing the time series of GEOS-Chem BC simulations vs field surface eBC concentrations has shown that the field site may be poorly representative of local or regional emissions, or vice versa.

Sensitivity simulations indicated that residential emissions were the largest contributor to surface BC concentrations in 2018 and 2022 (49–52 %), followed closely by other sources (36–40 %), industrial sources a distant third (7–8 %), and followed by transportation and energy sources. This reflects the limited reach of coal power stations and Botswana's reliance on imported energy. Focusing on residential contributions to BC would be the most effective at lowering BC exposure. Overall, the study underscores the importance of identifying and quantifying local and regional sources of BC to better understand and mitigate air pollution, particularly in rapidly urbanizing areas of Southern Africa. The results also highlight the need for improved accuracy in BC measurement and modeling to inform effective environmental and public health policies.

Africa is already one of the largest global source regions for carbonaceous emissions and dust, and it is undergoing a period of rapid urbanization that has the potential to significantly increase its anthropogenic emissions. To date, there has been relatively little research to address this growing issue that will affect both the growing population in the region and the composition of the global atmosphere. While this study is limited to a single country in Southern Africa, we anticipate that it will provide a framework for future studies in other African countries that help fill the existing data gaps.

#### CRedit authorship contribution statement

**Janica N.D. Gordon:** Writing – original draft, Investigation, Formal analysis, Data curation. **Kelsey R. Billsback:** Writing – review & editing, Supervision, Formal analysis, Conceptualization. **Marc N. Fiddler:** Writing – review & editing, Validation, Project administration, Formal analysis. **Jeffrey R. Pierce:** Writing – review & editing, Supervision, Methodology, Funding acquisition, Conceptualization. **Gizaw Mengistu Tsidu:** Writing – review & editing, Resources, Data curation. **Solomon Bililign:** Writing – review & editing, Supervision, Project

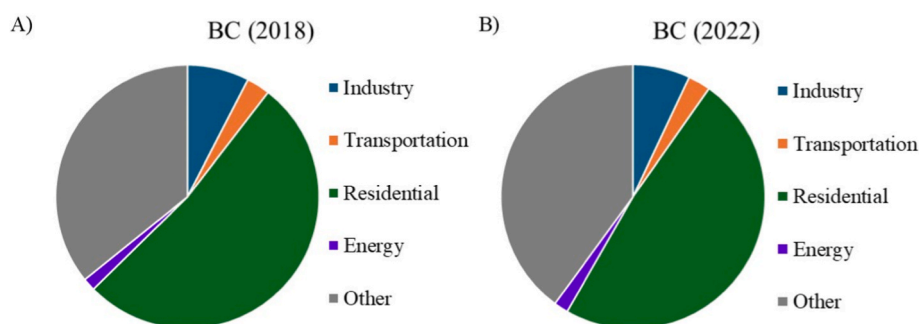


Fig. 6. Percentage difference in monthly-average surface ambient BC from GEOS-Chem for June and July of A) 2018 and B) 2022 at BIUST, where values are percent changes in units of  $\mu\text{g m}^{-3}$  for industry (blue), transportation (orange), residential (green), energy (purple), and other sources (grey).

administration, Funding acquisition, Conceptualization.

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## Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Solomon Billig reports financial support was provided by North Carolina Agricultural and Technical State University. Janica Gordon reports was provided by National Science Foundation. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.apr.2025.102707>.

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