nature portfolio

Peer Review File

Black Carbon Emissions Generally Underestimated in the Global South as Revealed by Globally Distributed Measurements

Corresponding Author: Ms Yuxuan Ren

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Attachments originally included by the reviewers as part of their assessment can be found at the end of this file.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)
See attached file

Reviewer #2

(Remarks to the Author)
BLACK CARBON EMISSIONS GENERALLY UNDERESTIMATED IN THE GLOBAL SOUTH AS REVEALED BY GLOBALLY DISTRIBUTED MEASUREMENTS
(Manuscript Number: ANCOMMS-25-10179)

COMMENTS FOR AUTHORS
Black carbon has great climatic importance because it is the aerosol that absorbs solar radiation most efficiently, consequently, the air that surrounds it increases its temperature. BC is also associated with adverse health impacts. In this study, the authors evaluate global BC emission inventories by applying a new dataset of BC mass measurements from the Surface Particle Network (SPARTAN) and complementary measurement networks. To achieve this goal, they use the global chemical transport model (GEOS-Chem) in its high-performance configuration (GCHP) for high-resolution simulations (relating BC emissions to their ambient concentrations) to compare them with the measurements. The authors find that simulations using the Community Emissions Data System (CEDS) emissions inventory show skill ($r^2 = 0.73$) in representing variability in SPARTAN measurements mainly in developed regions with low BC concentrations, but show a pronounced discrepancy ($r^2 = 0.00019$) in regions with high BC concentrations in the Global South; that is, the model used underestimates BC concentrations at most sites in the Global South (normalized mean bias of -38%). Furthermore, they obtain similar results when using alternative inventories (EDGARD, HTAP).

General comment
As a general comment, I consider that the authors of this study have omitted, in the discussion of the results and/or as complementary measurements, the efforts made by some South American countries to measure BC in urban areas, such as: Brazil, Colombia, Peru, Chile and Ecuador:
(https://doi.org/10.1016/j.atmosenv.2018.05.042;
https://doi.org/10.1016/j.jes.2022.12.025;
https://doi.org/10.1016/j.scitotenv.2022.156332;
https://doi.org/10.1016/j.scitotenv.2020.142736;
https://doi.org/10.4209/aaqr.210110;
https://doi.org/10.1080/10962247.2014.890141;
https://doi.org/10.1016/j.atmosenv.2020.117546).

Specifically, in the study https://doi.org/10.3390/rs15194702 , compares surface black carbon measurements (obtained with a Photoacoustic Extinctiometer (PAX) at 870 nm), with BC values obtained through the MERRA-2 reanalysis, in a city in

Peru bordering Chile, located at the head of the Atacama Desert. The authors found that MERRA-2 underestimates BC emissions from local sources.

The authors of this manuscript are aware that the (coarse) resolution of the simulations using models causes an artificial dilution within the grid cells, which limits the capacity of the models to capture fine-scale patterns; for this reason, they present an improvement with the GCHP.
Despite recognizing this effort by the authors, I consider that this study requires incorporating the results obtained in South American cities, those mentioned and others that allow for a better discussion, especially if, for the southern hemisphere, the sites considered are only 5 of the SPARTAN network and the complementary sites are only three from South America: urban (Bolivia), semi-urban (Argentina), and rural (Peru).

Reviewer #3

(Remarks to the Author)
Black carbon emissions generally underestimated in the Global South as revealed by globally distributed measurements by Y. Ren et al.

This paper uses black carbon (BC) mass concentrations (MBC) near the surface measured by SPARTAN in various regions of the world in 2019-2023. The MBC data are compared with the MBC values calculated by GEOS-Chem in its high-performance configuration (GCHO). CEDS, EDGAR, and HTAP BC emission inventories are used as inputs to the model. The model calculated MBC underestimate those observed at most Global South sites by about 38% on average. This indicates the necessities further investigations of BC emissions from developing regions.

Major comments

The attempt to evaluate the uncertainties of emission inventories in the global South by comparisons of MBC measured by SPARTAN and model calculations is interesting in principle. The motivation is valuable, if the SPARTAN provides MBC data with accuracies, sufficient enough to support the conclusion. However, the conclusion that the current emission inventories are underestimated is not supported, as detailed below. Therefore, I do not recommend the publication of this paper in Nature Communications.

1) The discussion in Section of “Uncertainties” of mass absorption cross section (MAC) is not systematic. First, MAC of HIPS is discussed in relation to that of Aethalometer. However, according to Singh (2024), Ohata et al. (2021) and number of other papers cited there, absorption coefficient of BC (babs) for AE is calculated as

babs (λ) = b0 (λ)/C0 R (1)

References
a) Mohit Singh, et al. (2024), Aerosol Sci. Technol., doi:10.1080/02786826.2024.2316173, https://doi.org/10.1080/02786826.2024.2316173.
b) Ohata, S. et al. (2021), Atmos. Meas. Tech. 14 (10): 672348. Doi:10.5194/amt-14-6723-2021.

As discussed in these papers, C0 is often set to be 3.5 for AE31 and AE33. And MAC is estimated as

MAC (λ) = b0 (λ)/(C0 MBC) = babs/MBC (2)

MBC = babs (λ) /MAC (λ) (3)

If different C0 is used, MAC (λ) varies accordingly. Namely, the absolute value of MAC (λ) depends on C0 chosen. It is necessary to clarify how you defined C0 and MAC for HIPS of SPARTAN. Otherwise, it is not possible to compare absolute value of MAC of HIPS with that of AE.

2) It is mentioned that emission inventories of BC are consistent with the SPARTAN data in Europe ($r2 = 0.93$, slope = 1.2). Singh et al. (2024) showed that MAC of AE was stable to within about 10% in Europe. In this regard, the $r2 = 0.93$ (Line 162) is consistent with stable MAC measured by Singh et al. (2024).

3) In this paper it is assumed that MAC is constant throughout the world, due to the lack of measurements of MAC in different regions of the world. However, this assumption has to be proven by direct measurements of MAC.
As discussed in this paper, BC emissions in Global South are dominated by diffuse and inefficient combustion sources, …. (Line 190-205).
MAC of BC strongly influenced by these sources can be different from that in more developed regions, including Europe.
This paper concludes that emission estimates of BC in Global South is underestimated by about 40% (Figure 2). But in turn, this result can be interpreted that MAC used for SPARTAN in Global South is underestimated by 40%.
The serious problem of SPARTAN for MBC measurements is that MAC was not determined by simultaneous measurements of MBC in different regions. In order to estimate MBC with high accuracy from equation (3), MAC (λ) values in pertinent regions have to be determined first. Otherwise, the logic of estimating MBC from babs becomes circular.

4) For the blue markers in Figure 2 (i.e., the data mostly from North America and East Asia), the correlation coefficient

between model (GEOS-Chem) and observation (HIPS) is high, but the slope is 2.2 (Figure 2). Depending on the assumed MAC values for HIPS, this slope can reach 2.9 (Figure S6). These results could also be interpreted as “the relative distribution of BC emissions in these regions is somewhat correct, but the absolute values are overestimated”.
We suggest reporting the mean or median model/observation MBC ratios, in addition to the slope of the model/observation MBC correlation.

5) For the red markers in Figure 2 (i.e., the data mostly from Africa and South Asia), the high-concentration observations from Dhaka and Addis Ababa seem to have a significant impact on the correlation and slope, and thus on the conclusions of this study. It would be desirable to have more explanation of the validity of the HIPS measurements at these observatories.
The HIPS technical paper (White et al., 2016; reference No. 44 of the manuscript) mentions the possibility of dust (Fe) as well as BC affecting the measurements. Is it possible that dust is strongly affecting the measurements, especially at these two observatories?

Minor comments

Figure 2.
a) The line corresponding to the equation marked in red is not shown. Ther is no need to show 1:1 relation line. And explanations for the equation marked in blue should be given.

b) It is written that the blue markers in Figure 2 are for the data mostly from North America and East Asia. On the other hand, one, it is written that in Europe r2 = 0.93 and slope = 1.2 (Lines 160-163). There are no explanations on the difference between Europe and North America (blue markers in Figure 2). This is inconsistent and confusing.

Reviewer #4

(Remarks to the Author)
I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)
The authors have thoroughly addressed all of my previous comments and concerns. Their responses were clear, detailed, and supported by appropriate revisions in the manuscript. I am satisfied with the changes made and believe the manuscript has been significantly improved as a result. I have no further concerns.

Reviewer #2

(Remarks to the Author)
BLACK CARBON EMISSIONS GENERALLY UNDERESTIMATED IN THE GLOBAL SOUTH AS REVEALED BY GLOBALLY DISTRIBUTED MEASUREMENTS
(Manuscript Number: NCOMMS-25-10179A)
GENERAL COMMENTS FOR AUTHORS / EDITOR
The revised manuscript has incorporated the comments/suggestions made to the authors, and therefore, from our perspective, its content has improved.
To improve regional coverage, they include a sensitivity study with a more flexible sampling duration criterion from six to two months, to allow for the inclusion of additional BC concentration studies (conducted in South American countries) that provide valuable information (initially omitted). Using these data, they have re-evaluated BC emissions and present the new results in the revised Supplementary Information (Text S2 and Table S5).
They also cite the recommended references in the revised manuscript and in the Supplement, as appropriate.
The authors, having taken my comments/suggestions into account, consider the manuscript ready for publication.
Finally, I would like to thank the journal's editor for considering me as a reviewer for this manuscript.

Reviewer #3

(Remarks to the Author)
The authors revised the submitted manuscript fully taking into account of the comments from me and the co-reviewer.

Therefore, this paper should be published in Nature Communications.

Reviewer #4

(Remarks to the Author)
I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

We sincerely appreciate the reviewers for their constructive comments. In the revision and reply to comments, we now better clarify the reliability of SPARTAN and complementary BC measurements and thus the robustness of evaluating emissions. Specific rebuttals to each comment from reviewers are given below in blue. All line numbers refer to the revised manuscript (without tracked changes) unless explicitly stated.

Referees' comments:

Referee #1 (Remarks to the Author):

This manuscript presents a dataset of globally distributed black carbon (BC) mass measurements obtained from the Surface Particulate Matter Network (SPARTAN) and complementary measurement networks to evaluate contemporary BC emission inventories. The authors employ the GEOS-Chem global chemical transport model in its high-performance configuration (GCHP) for high-resolution simulations to link BC emissions with ambient concentrations, facilitating comparison with the observational dataset. The results indicate that the model systematically underestimates BC concentrations at most Global South sites, with a normalized mean bias of -38%.

While previous studies have relied primarily on different models and satellite-based data, this manuscript underscores the necessity of long-term surface monitoring, particularly in the Global South. Prior studies that utilized satellite estimates often reported annual mean values, but such estimates should be interpreted with caution due to the increased error in regions lacking ground-based BC measurements. This study integrates ground-based measurements with modeling approaches/Simulations to demonstrate that BC emissions tend to be underestimated in the Global South compared to the Global North.

Although the manuscript is relevant to the scope of the journal, the novelty of the data presented is somewhat limited. The study does not introduce fundamentally new findings beyond those reported in previous characterizations of BC emissions, particularly in the Global South. However, the value of this work lies in the scarcity of comparable datasets from this region and its integration of ground-based observations. Given the significance of the study and the robustness of the analysis, I recommend the manuscript for publication. The scarcity of BC measurements in the Global South, particularly in Africa, makes this study highly relevant to the scientific community.

Reply: We thank the referee for affirming the significance and robustness of this work. Indeed the novelty of this work reflects the opportunity to rigorously evaluate BC emission inventories across the Global South to elucidate their ability to represent novel coordinated measurements.

General Comments for Improvement

A key strength of this study is the acquisition of a comprehensive dataset of BC concentrations from globally distributed SPARTAN sites. However, the authors have utilized datasets that are inherently challenging to compare due to differences in instrumentation, sampling objectives, sampling heights (e.g., rooftop placement), and sampling periods.

Reply: Within SPARTAN, we maintain rigorous consistency in instrumentation and sampling objectives across all globally distributed sites. While sampling heights inevitably vary, rooftop placements are carefully selected to enhance spatial representativeness and better represent urban background conditions. Similarly, despite slight differences in exact sampling periods, most sites

have multi-year sampling records covering all four seasons. Therefore, using annual mean BC concentrations at SPARTAN sites provides a robust approach for evaluating emissions. We add the following sentence to the revised manuscript.

Line 369-373: "Table S3 provides specific location details for globally distributed SPARTAN sites. High population density and poorly sampled regions are two key factors in site selection. Rooftop placement enhances spatial fetch, better represents urban background, and offers instrument security."

For complementary measurements, we acknowledge the greater variability in instrumentation, sampling objectives, and sampling schemes across different networks and studies. To improve comparability, we have applied a rigorous data screening process where possible. Additionally, we recognize the inherent challenges of comparing complementary measurements and explicitly address these uncertainties in the revised manuscript.

Line 359-362: "Additionally, variations in instrumentation, sampling objectives, and methodologies among other individual studies create challenges for intercomparison and model evaluation for intercomparison and model evaluation."

• The authors consistently refer to "developing countries" throughout the manuscript. It would be more appropriate to use "low- and middle-income countries" (LMICs) to ensure inclusivity. Several arguments have been made against the use of the term "developing world," and adopting LMICs would enhance precision and consistency with contemporary discourse.

Reply: We now use "LMICs" instead of "developing countries" everywhere in the revised manuscript wherever it applies.

• The authors should provide further discussion on the source contributions of BC emissions in the datasets used. For example, BC emissions in Africa arise from diverse sources, including wildfires, biomass combustion, and vehicular traffic, with the relative contributions varying across countries. These differences should be explicitly highlighted and discussed in the manuscript.

Reply: We add a paragraph on the source contributions of BC emissions, with a particular focus on LMICs. We also include BC emissions by source sector across regions in Figure S8 and provide additional details on source sectors of widely used inventories in Table S4 in the revised Supplementary Information. The content of Table S4 is not shown here for conciseness.

Line 424: "Sector definitions for CEDS v2, EDGAR v6.1, and HTAP v3 are detailed in Table S4."

Line 425-434: "Despite variations among global inventories, an overall assessment of the 2019 BC emissions indicates that residential, transportation, and industrial sectors are the dominant anthropogenic sources, while energy and waste contribute relatively smaller fractions (Figure S8). The relative contribution of these sectors also exhibits considerable regional variations. For example, the residential sector plays a larger role across South Asia and Sub-Saharan Africa, while transportation accounts for a relatively greater share in North America, North Africa, and the Middle East. In addition to anthropogenic sources, biomass burning contributes significantly to total BC emissions in Central Sub-Saharan Africa, Tropical and Andean Latin America, and Australasia."

Supplement Line 267-269: "

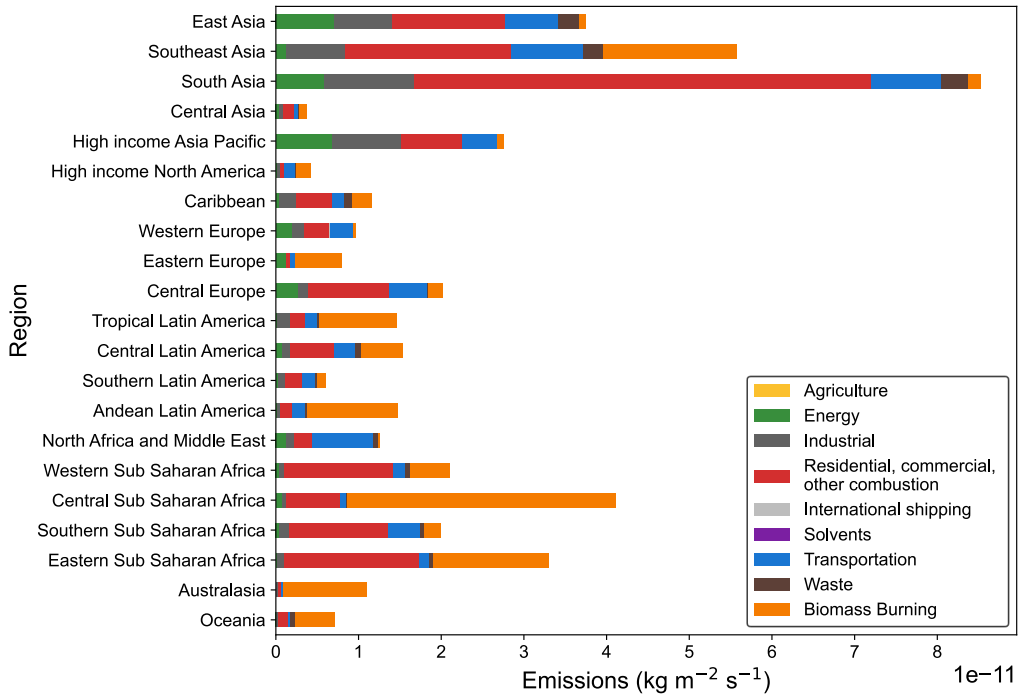

Figure S8. BC emissions by source sector across 21 global regions in 2019 from CEDS v2 and GFED v4.1s emission inventory."

• The study includes 2,257 filter samples collected from 22 SPARTAN sites between 2019 and 2023. This period encompasses the COVID-19 lockdowns, during which human activities, particularly those contributing to BC emissions, were significantly reduced (by approximately 80% in some regions). The authors should clarify whether the data collected during this period were included in their comparisons and discuss the potential impact of pandemic-related emission reductions on their findings.

Reply: We explore this issue by considering filter samples collected in 2019 and from July 2021 onward as unaffected by COVID-19 lockdowns. The subset of data unaffected by lockdowns consists of 2,080 filter samples from 22 sites. To assess the impact of the lockdown period, we compare the annual mean BC concentrations between this subset and the full dataset across sites, finding a normalized mean bias of -3.2%. This suggests that the small proportion of samples collected during lockdowns has minimal impact on our comparisons. We add the following sentence to the revised manuscript.

Line 317-321: "We examine the potential effect of COVID-19 lockdowns on our analyses. Exclusion of the period Jan 2020 – Jul 2021 that may have been affected by COVID-19 lockdowns would reduce the total number of samples by 7.8%, and would introduce a NMB versus the full dataset of only -3.2% without affecting our conclusions. Thus we err on the side of inclusion and present the full dataset for completeness."

• The manuscript references BC measurements in Africa but includes data from only four sites across three African countries. These limited sampling locations may not provide a sufficiently representative characterization of BC emissions across the continent or Sub-Saharan Africa. The authors should acknowledge this as a limitation of the study and discuss its implications.

Reply: We now explicitly acknowledge this limitation and discuss its implications in the revised manuscript.

Line 358-360: "Despite SPARTAN's efforts to provide long-term, reliable measurements across globally distributed sites, additional geographic coverage in Africa and South America remains desirable."

Line 362-364: "This highlights the need for expanded measurement networks across additional locations to improve our understanding of BC concentrations and emissions, particularly in the Global South."

• In Figure 1, New Zealand appears to be missing from the map. The authors should clarify whether this omission was intentional or an oversight.

Reply: We appreciate the reviewer for pointing this out. New Zealand is unintentionally cropped, and we update Figure 1 to ensure its inclusion.

Referee #2 (Remarks to the Author):

BLACK CARBON EMISSIONS GENERALLY UNDERESTIMATED IN THE GLOBAL SOUTH AS REVEALED BY GLOBALLY DISTRIBUTED MEASUREMENTS (Manuscript Number: ANCOMMS-25-10179)

COMMENTS FOR AUTHORS

Black carbon has great climatic importance because it is the aerosol that absorbs solar radiation most efficiently, consequently, the air that surrounds it increases its temperature. BC is also associated with adverse health impacts. In this study, the authors evaluate global BC emission inventories by applying a new dataset of BC mass measurements from the Surface Particle Network (SPARTAN) and complementary measurement networks. To achieve this goal, they use the global chemical transport model (GEOS-Chem) in its high-performance configuration (GCHP) for high-resolution simulations (relating BC emissions to their ambient concentrations) to compare them with the measurements. The authors find that simulations using the Community Emissions Data System (CEDS) emissions inventory show skill ($r2 = 0.73$) in representing variability in SPARTAN measurements mainly in developed regions with low BC concentrations, but show a pronounced discrepancy ($r2 = 0.00019$) in regions with high BC concentrations in the Global South; that is, the model used underestimates BC concentrations at most sites in the Global South (normalized mean bias of -38%). Furthermore, they obtain similar results when using alternative inventories (EDGARD, HTAP).

General comment

As a general comment, I consider that the authors of this study have omitted, in the discussion of the results and/or as complementary measurements, the efforts made by some South American countries to measure BC in urban areas, such as: Brazil, Colombia, Peru, Chile and Ecuador: (https://doi.org/10.1016/j.atmosenv.2018.05.042;
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https://doi.org/10.1080/10962247.2014.890141;

https://doi.org/10.1016/j.atmosenv.2020.117546).
Specifically, in the study https://doi.org/10.3390/rs15194702, compares surface black carbon measurements (obtained with a Photoacoustic Extinctiometer (PAX) at 870 nm), with BC values obtained through the MERRA-2 reanalysis, in a city in Peru bordering Chile, located at the head of the Atacama Desert. The authors found that MERRA-2 underestimates BC emissions from local sources.

Reply: We thank the reviewer for highlighting relevant BC measurement efforts in South America. In our selection of complementary measurements, we have applied careful screening criteria to ensure data reliability for emission evaluation: continuous measurements at background sites for at least six months adjacent to 2019. While this approach maximizes individual site representativeness, it also limits the number of available studies, thereby restricting overall coverage in South America. To improve regional coverage, we now include a sensitivity study with relaxed sampling length criterion from six months to two months, allowing the inclusion of additional studies that provide valuable insights. Using this expanded complementary dataset, we reevaluate BC emissions alongside the original evaluation and present new results in the revised Supplementary Information, including Text S2 and Figure S9. We also update Table S5 accordingly and add clarifying sentences to differentiate between the original and relaxed complementary datasets where applicable. We now cite the recommended references wherever appropriate in the revised manuscript and Supplement. Figure S9 and Table S5 are not shown here for conciseness.

Line 185-187: "Evaluation using complementary measurements also suggests a model-measurement discrepancy in the Global South, with additional details discussed in Text S2 of the Supplementary Information."

Line 475-477: "To improve regional coverage, we further relax the sampling length criterion to two months for an additional complementary dataset alongside the original. Both datasets are used for model evaluation."

Supplement Line 86-110: **"Text S2. Evaluation Using Complementary Measurements**

In addition to the robust evaluation based on long-term measurements from globally distributed SPARTAN sites, we extend our analysis by incorporating data from other available networks and individual studies ("Methods" section). Figure 1 and Figure S9 present evaluation results using the original and the relaxed data screening criteria, which require a minimum sampling duration of six months and two months, respectively. The evaluation with complementary measurements generally supports similar conclusions as those derived from SPARTAN, showing overall consistency in developed regions while highlighting discrepancies in the Global South. Notably, with the original data screening criteria, the median simulated-to-measured ratio is 0.44 across five sites in South America and 0.25 across six sites in Africa. While emission evaluations in the Global South remain limited, several site-specific studies also indicate underestimation in simulations. For example, simulated BC concentrations using three different emission inventories were consistently lower than measured values at a rural site in India, with a median simulated-to-measured ratio of 0.59[1]. Similarly, in Tacna, Peru, simulated BC concentrations were found to be an order of magnitude lower than ground-based measurements[2]. These pronounced discrepancies reveal the challenges in accurately representing BC emissions in existing inventories across the Global South.

Unlike SPARTAN, which maintains consistency across all globally distributed sites and ensures robustness for evaluation, complementary datasets are compiled from diverse sources and exhibit inherent variability in instrumentation, sampling objectives, and sampling schemes. Despite applying a careful data screening process, we acknowledge the uncertainties associated with these variations and, therefore, primarily focus our discussion on SPARTAN data."

The authors of this manuscript are aware that the (coarse) resolution of the simulations using models causes an artificial dilution within the grid cells, which limits the capacity of the models to capture fine-scale patterns; for this reason, they present an improvement with the GCHP.

Reply: We thank the referee for recognizing our efforts in addressing the representativeness bias.

Despite recognizing this effort by the authors, I consider that this study requires incorporating the results obtained in South American cities, those mentioned and others that allow for a better discussion, especially if, for the southern hemisphere, the sites considered are only 5 of the SPARTAN network and the complementary sites are only three from South America: urban (Bolivia), semi-urban (Argentina), and rural (Peru).

Reply: We now include more sites in South America, with five based on the original screen criteria and seven based on the relaxed criteria, as discussed above. Additionally, we explicitly recognize this limitation in the revised manuscript.

Line 358-360: "Despite SPARTAN's efforts to provide long-term, reliable measurements across globally distributed sites, additional geographic coverage in Africa and South America remains desirable."

Referee #3 (Remarks to the Author):

Black carbon emissions generally underestimated in the Global South as revealed by globally distributed measurements by Y. Ren et al.

This paper uses black carbon (BC) mass concentrations (MBC) near the surface measured by SPARTAN in various regions of the world in 2019-2023. The MBC data are compared with the MBC values calculated by GEOS-Chem in its high-performance configuration (GCHO). CEDS, EDGAR, and HTAP BC emission inventories are used as inputs to the model. The model calculated MBC underestimate those observed at most Global South sites by about 38% on average. This indicates the necessities further investigations of BC emissions from developing regions.

Major comments

The attempt to evaluate the uncertainties of emission inventories in the global South by comparisons of MBC measured by SPARTAN and model calculations is interesting in principle. The motivation is valuable, if the SPARTAN provides MBC data with accuracies, sufficient enough to support the conclusion. However, the conclusion that the current emission inventories are underestimated is not supported, as detailed below. Therefore, I do not recommend the publication of this paper in Nature Communications.

Reply: We thank the reviewer for affirming the significance of this endeavor, however, we respectfully disagree with the assertion that BC mass data provided by SPARTAN are inaccurate and affirm that these data are robust enough to support our current conclusions. We systematically

reinforce the reliability of SPARTAN data and provide a more detailed analysis of associated uncertainties. Please find our responses to specific comments below.

1) The discussion in Section of "Uncertainties" of mass absorption cross section (MAC) is not systematic. First, MAC of HIPS is discussed in relation to that of Aethalometer. However, according to Singh (2024), Ohata et al. (2021) and number of other papers cited there, absorption coefficient of BC (babs) for AE is calculated as

babs (λ) = b0 (λ)/C0 R (1)

References

a) Mohit Singh, et al. (2024), Aerosol Sci. Technol., doi:10.1080/02786826.2024.2316173, https://doi.org/10.1080/02786826.2024.2316173.

b) Ohata, S. et al. (2021), Atmos. Meas. Tech. 14 (10): 672348. Doi:10.5194/amt-14-6723-2021.

As discussed in these papers, C0 is often set to be 3.5 for AE31 and AE33. And MAC is estimated as

MAC (λ) = b0 (λ)/(C0 MBC) = babs/MBC (2)

MBC = babs (λ) /MAC (λ) (3)

If different C0 is used, MAC (λ) varies accordingly. Namely, the absolute value of MAC (λ) depends on C0 chosen. It is necessary to clarify how you defined C0 and MAC for HIPS of SPARTAN. Otherwise, it is not possible to compare absolute value of MAC of HIPS with that of AE.

Reply: Thank you for identifying this potential source of confusion. Below we address the concern by explaining the similarities and differences between aethalometers and HIPS, and the attributes of HIPS that enable its robust characterization of BC. For aethalometers, the absorption coefficient (b_{abs}) is corrected for multiple scattering effects (C_0) (i.e., optical interactions between absorbing particles and multiple-scattering filter substrate) and filter loading effects (R) (i.e., nonlinear decrease in attenuation as more absorbing particles accumulate on the filter). In contrast, HIPS rigorously accounts for these effects through simultaneous transmittance and reflectance measurements, a theory-based calibration model derived from field blanks, and the use of optically thin PTFE filters (White et al., 2016).

1. Simultaneous measurements of transmitted and reflected light. Aethalometers determine b_0 by measuring the attenuation of light transmitted through particles accumulating on the filter (Equation 4). In contrast, HIPS independently measures both transmittance (t) using an integrating plate and reflectance (r) using a sphere positioned on the unexposed side of the filter (Equation 5). The reflectance measurement accounts for pre-sampling transmittance (I_0) and additionally supplies a correction for backscattering by sampled particles, thus accounting for filter loading effects.

$$b_0(AE) = \frac{A}{V}\ln\left(\frac{I_0}{I}\right) \quad (4)$$

$$b_{abs}(\text{HIPS}) = \frac{A}{V}\ln\left(\frac{1-r}{t}\right) \quad (5)$$

2. Theory-based calibration model derived from field blanks. Rather than applying fixed factors for correction, HIPS converts b_0 to b_{abs} using a theory-based calibration model with parameters determined from field blank measurements (White et al., 2016). Briefly, a linear

regression of field blanks provides an intercept (a_0) and slope (a_1), which transform raw reflectance (R) and transmittance (T) values into field blank corrected values $r = -a_1R/a_0$ and $t = T/a_0$. The b_0 and b_{abs} are then calculated as:

$$b_0(\text{HIPS}) = \frac{A}{V}\ln\left(\frac{1-R}{T}\right) \quad (4)$$

$$b_{abs}(\text{HIPS}) = \ln\frac{A}{V}\left(\frac{a_0+a_1R}{T}\right) \quad (5)$$

This correction ensures that field blanks exhibit zero absorption, effectively mitigating multiple scattering effects. To ensure accuracy, at least 40 field blanks from the same filter batch are used for calibration.

3. Use of optically thin PTFE filters. Aethalometers typically use optically thick quartz- or glass-fiber filters, which can adsorb ambient organic gases at chemically active surface sites. In contrast, PTFE filters used in SPARTAN have considerably lower surface areas for physical adsorption and show minimal absorption of gas-phase organic carbon. Additionally, PTFE filters introduce much less extraneous scattering to the analyzed matrix of collected particles and the filter substrate.

Following these corrections, b_{abs} is converted to BC mass (M_{BC}) using the mass absorption cross section (MAC). The definition of MAC for HIPS is conceptually the same as for aethalometers, as noted in Equation 2 by the reviewer, where b_{abs} is determined by HIPS and M_{BC} is elemental carbon (EC) measured using IMPROVE thermal optical reflectance (TOR) method (Chow et al., 2001; 2007). Since both HIPS and aethalometers define MAC based on b_{abs} and a reference mass, the comparison used in this study is appropriate.

We now systematically present the HIPS methodology and explicitly define the corrections for multiple scattering effects and filter loading effects in Text S5 (Supplement Line 141-174). For conciseness, the content of Text S5 is not shown here.

Reference:

Chow, J. C., Watson, J. G., Crow, D., Lowenthal, D. H. & Merrifield, T. Comparison of IMPROVE and NIOSH carbon measurements. *Aerosol Sci. Tech.* **34,** 23–34 (2001).

Chow, J. C. et al. The IMPROVE-A temperature protocol for thermal/optical carbon analysis: maintaining consistency with a long-term database. *J. Air & Waste Manage. Assoc.* **57,** 1014–1023 (2007).

White, W. H., Trzepla, K., Hyslop, N. P. & Schichtel, B. A. A critical review of filter transmittance measurements for aerosol light absorption, and calibration for a decade of monitoring on PTFE membranes. *Aerosol Sci. Tech.* **50,** 984–1002 (2016).

2) It is mentioned that emission inventories of BC are consistent with the SPARTAN data in Europe (r^2 = 0.93, slope = 1.2). Singh et al. (2024) showed that MAC of AE was stable to within about 10% in Europe. In this regard, the r^2 = 0.93 (Line 162) is consistent with stable MAC measured by Singh et al. (2024).

Reply: We thank the reviewer for pointing this out! We add a citation to Singh et al. (2024). The stability of MAC in Europe is addressed alongside other regions in our response to Comment 3.

3) In this paper it is assumed that MAC is constant throughout the world, due to the lack of measurements of MAC in different regions of the world. However, this assumption has to be

proven by direct measurements of MAC. As discussed in this paper, BC emissions in Global South are dominated by diffuse and inefficient combustion sources, ….(Line 190-205). MAC of BC strongly influenced by these sources can be different from that in more developed regions, including Europe. This paper concludes that emission estimates of BC in Global South is underestimated by about 40% (Figure 2). But in turn, this result can be interpreted that MAC used for SPARTAN in Global South is underestimated by 40%.

The serious problem of SPARTAN for MBC measurements is that MAC was not determined by simultaneous measurements of MBC in different regions. In order to estimate MBC with high accuracy from equation (3), MAC (λ) values in pertinent regions have to be determined first. Otherwise, the logic of estimating MBC from babs becomes circular.

Reply: We address the concern about the lack of simultaneous MAC measurements and its potential implications for our conclusions as follows:

1. Regional MAC variations cannot explain the identified underestimations. While we recognize that MAC varies with aerosol composition, mixing state, and morphology, regional differences are relatively limited and unlikely to account for the underestimation of BC emissions identified in this study. Previous studies indicate that MAC variation in developed regions is limited, with absolute values close to 10 m^2/g used in SPARTAN. For example, as kindly noted by the reviewer in Comment 2, Singh et al. (2024) found a stable MAC value of 10.9 m^2/g with only 11% spatial variability across four Arctic sites, which further aligns within 10% with prior studies at northern mid-latitudes and Arctic sites. In the Global South, despite the limited number of studies, available data generally report values close to 10 m^2/g or within the 7 m^2/g to 13 m^2/g uncertainty range outlined in our study. Additionally, MAC values associated with inefficient combustion sources are often reported to be even lower than 10 m^2/g. This suggests that MAC uncertainty may, in some cases, amplify the underestimation of BC emissions rather than account for it.
   Conversely, if we were to fully attribute the identified underestimates to MAC variations, it would result in unreasonably high MAC values at Global South sites. For example, the simulated-to-measured BC ratios in Dhaka (Bangladesh), Addis Ababa (Ethiopia), and Ilorin (Nigeria) are 0.25, 0.31, and 0.48, respectively. If we assume the simulations represent the true BC mass and adjust MAC accordingly, the "adjusted" MAC values would be 40 m^2/g for Dhaka, 32 m^2/g for Addis Ababa, and 21 m^2/g for Ilorin. These values are not physically plausible for urban background sites, reinforcing that MAC uncertainties alone cannot explain the observed underestimation in emissions.
2. Direct and independent MAC measurements are important but out of the scope of this study. While direct MAC measurements are highly valuable for characterizing aerosol optical properties, obtaining such measurements requires simultaneous instrumentation for both b_{abs} (e.g., aethalometer) and reference BC mass (e.g., continuous soot monitoring system, COSMOS). A full exploration of exact MAC values and aerosol optical properties is not the focus of this study. Additionally, deploying and maintaining additional reference instruments at all 22 globally distributed SPARTAN sites for years is infeasible at this time.

Accordingly, we include a compilation of previously measured MAC values in Table S2 (Supplement Line 184-191) and reorganize the discussion on MAC uncertainty in the revised manuscript. We begin with MAC values for freshly emitted BC, followed by an analysis of its regional variability, and include the associated uncertainties and their impact on our conclusions. Additionally, we emphasize the importance of future studies focused on direct MAC

measurements, particularly in the Global South. For conciseness, the content of Table S2 is not shown here.

Line 273-275: "The optical measurements depend on the mass absorption cross section (MAC), which varies with aerosol composition, mixing state, and morphology to estimate BC concentration."

Line 279-299: "Some other studies use MAC values that deviate from the traditional 10 m^2/g, which are described below and for consistency are adjusted to 633 nm by assuming an inverse wavelength dependence. We summarize recent laboratory and field-measured MAC values across different regions and combustion sources in Table S2. For freshly emitted BC, Bond and Bergstrom[52] suggested a MAC of 6.52 ± 1.05 m^2/g, and Liu et al.[53] recommended a MAC of 6.95 ± 0.608 m^2/g. Once released into the atmosphere, aerosol undergoes processes of condensation, aggregation, and aging, which may cause the MAC value to increase due to coating or decrease due to particle coagulation and aggregate collapse. In developed regions, MAC variation is limited, with absolute values generally close to 10 m^2/g. Singh et al.[54] found a stable MAC value of 10.9 m^2/g with 11% spatial variability across four Arctic sites, which further aligns within 10% with prior studies at northern mid-latitudes and Arctic sites. Similarly, White et al.[44] found a coefficient of 10.2 m^2/g for b_{abs}/M_{EC} across 110 IMPROVE sites in the US. In the Global South, despite the limited number of studies, available data generally report MAC values with an uncertainty range of 7 m^2/g to 13 m^2/g, with MAC values from inefficient combustion sources often reported to be lower than 10 m^2/g. For example, average MAC values from residential biofuel stoves, diesel trucks, and non-road mobile machinery are reported to be 8.03, 7.25, and 9.99, respectively[55,56]. These lower MAC values would strengthen our conclusion about underestimates in BC emissions in the Global South as discussed further below."

Line 300-302: "Given the limited regional variation, we apply the best available MAC value of 10 m^2/g and conduct a sensitivity test using an uncertainty range of 7 m^2/g to 13 m^2/g (Figure S6)."

Line 304-309: "Alternatively, applying a MAC of 7 m^2/g in SPARTAN would improve the model-measurement slope in developed regions (r^2 = 0.73; slope = 1.5) and partially address the model overestimation in Beijing, by reducing the simulated-to-measured ratio from 7.4 to 5.2, but would increase the model-measurement bias in the Global South such as in Dhaka (0.17), Addis Ababa (0.22), Ilorin (0.34), and Mexico City (0.40), thus strengthening the conclusions of this study."

Line 312-316: "Unrealistic MAC values of 40 m^2/g for Dhaka, 32 m^2/g for Addis Ababa, and 21 m^2/g for Ilorin would be needed to achieve unity simulated-to-measured BC ratios. Thus, despite uncertainties surrounding the exact MAC value, the overall conclusion remains that BC emissions are generally underestimated in the Global South."

4) For the blue markers in Figure 2 (i.e., the data mostly from North America and East Asia), the correlation coefficient between model (GEOS-Chem) and observation (HIPS) is high, but the slope is 2.2 (Figure 2). Depending on the assumed MAC values for HIPS, this slope can reach 2.9 (Figure S6). These results could also be interpreted as "the relative distribution of BC emissions in these regions is somewhat correct, but the absolute values are overestimated".

We suggest reporting the mean or median model/observation MBC ratios, in addition to the slope of the model/observation MBC correlation.

Reply: To improve clarity, we change the phrase "consistency (r^2 = 0.73) in the spatial variation" to "consistency (r^2 = 0.73) in the relative spatial distribution" (Line 158) and explicitly address the

potential overestimate reflected by the slope. We also add the mean, median, and standard error of simulated-to-measured ratios to the revised manuscript.

Line 159-162: "The simulated-to-measured ratios across these SPARTAN sites are 1.45 ± 0.29 (mean ± standard error) with a median of 1.52. Both the ratio and slope exceed unity, primarily reflecting a simulation overestimate in East Asia that may arise from the recent adoption of BC control technologies[32]."

Line 182-183: "The simulated-to-measured ratios across these Global South sites are 0.67 ± 0.09 (mean ± standard error) with a median of 0.66."

5) For the red markers in Figure 2 (i.e., the data mostly from Africa and South Asia), the high-concentration observations from Dhaka and Addis Ababa seem to have a significant impact on the correlation and slope, and thus on the conclusions of this study. It would be desirable to have more explanation of the validity of the HIPS measurements at these observatories.

The HIPS technical paper (White et al., 2016; reference No. 44 of the manuscript) mentions the possibility of dust (Fe) as well as BC affecting the measurements. Is it possible that dust is strongly affecting the measurements, especially at these two observatories?

Reply: This is an interesting point, however it doesn't alter our conclusions as described below. Indeed, the absorption coefficient (b_{abs}) is influenced by Fe in the sampled aerosol in addition to BC, leading to potential dust interference in any absorption-based measurement, including HIPS and aethalometers. To address this, we apply the Fe correction approach from White et al. (2016) and use adjusted BC values for comparison with simulations. We find that this correction has a negligible impact on the overall conclusions on emission evaluations. For Dhaka and Addis Ababa specifically, the original mean BC concentrations are 5.6 μg/m^3 and 4.8 μg/m^3, respectively, while the corrected values are 5.2 μg/m^3 and 4.5 μg/m^3 — still considerably higher than the simulated concentrations of 1.4 μg/m^3 and 1.5 μg/m^3.

Accordingly, we include a discussion on dust interference in the "Uncertainties" section in the revised manuscript and provide further details on the correction method in Text S4 in the Supplement.

Line 322-336: "Dust, in addition to BC, contributes to absorption and can interfere with BC determination in any absorption-based measurement. Among major dust elements, iron (Fe) serves as a tracer for absorbing dust and has a distinct association with absorption[15,44], independent of BC. To account for this, we apply a dust correction by subtracting Fe's contribution to absorption and use the adjusted BC concentrations for comparison with simulations (Text S4). While this correction leads to a slight decrease in BC concentrations (with a NMB of -9.7%), its impact on emission evaluations is negligible. Consistency remains high for primarily developed regions, while discrepancies persist across most Global South sites, with r^2 values changing only slightly from 0.71 to 0.74 and 0.00019 to 0.00035, respectively (Figure S7). Although the total Fe content used for correction is an imperfect indicator of absorbing dust, as it includes both (hydr)oxides responsible for absorption and structural Fe in non-absorbing clays, the minimal impact of this correction confirms that dust interference in BC determination is negligible in this study, reinforcing our conclusion that BC emissions are generally underestimated in the Global South."

Supplement Line 128-140: "**Text S4. Fe Correction**

In addition to multiple methods for measuring aerosol absorption, SPARTAN filters are analyzed for elemental composition by X-ray fluorescence (XRF). The total Fe content is determined with

high reliability, with measurement procedures and quality assurance detailed in Liu et al.[6]. Fe serves as a tracer for absorbing dust and has a distinct association with absorption. White et al.[8] examined Fe's interference on BC determination using HIPS and derived the following relationship using 10 years of measurements from IMPROVE:

$$b_{abs} = A \times M_{BC,corrected} + B \times M_{Fe} \quad (3)$$

where b_{abs} is the absorption coefficient determined by HIPS, $M_{BC,correted}$ is the BC concentration corrected for dust interference on absorption, and M_{Fe} is the total Fe content measured by XRF. The coefficients A = 10.2 m^2/g BC and B = 6.6 m^2/g Fe represent the MAC estimates for BC and Fe, respectively."

Supplement Line 259-268: "

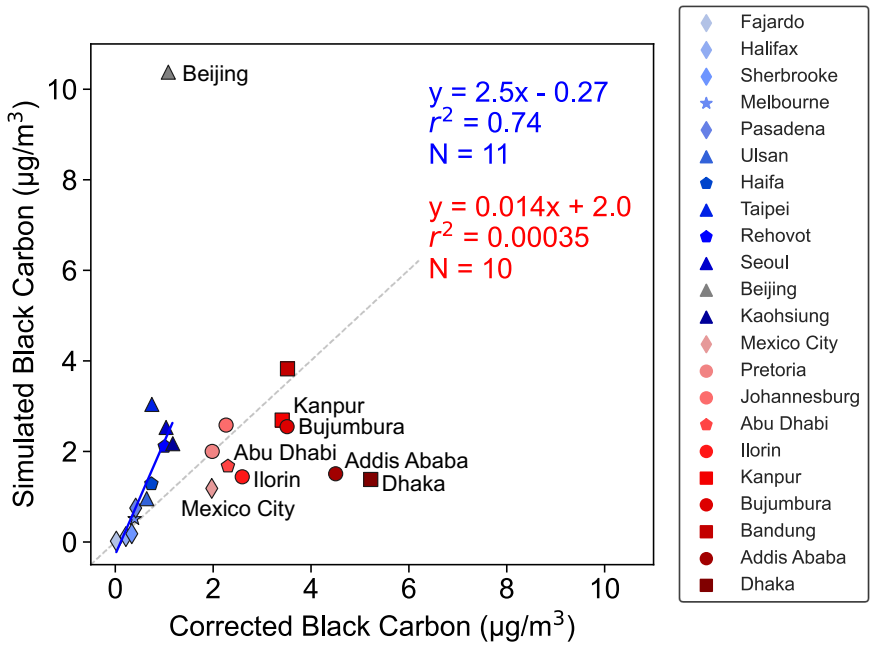

Figure S7. Annual mean BC concentrations across SPARTAN sites, with GCHP simulation from 2019 and SPARTAN measurements corrected for dust interference from 2019 to 2023. Annotations include the line of best fit (y), coefficient of variation (r^2), and number of comparison points (N). The lowest half of the measured concentrations are indicated in blue and the upper half in red. The Beijing site, marked in grey, is excluded from statistical calculations due to anomalies in its emissions estimates. Symbols indicate different regions (diamonds for North America, star for Australia, triangles for East Asia, pentagons for the Middle East, circles for Africa, and squares for South Asia)."

Minor comments

Figure 2

a) The line corresponding to the equation marked in red is not shown. Ther is no need to show 1:1 relation line. And explanations for the equation marked in blue should be given.

Reply: We have not included the line corresponding to the equation in red, as the very low r^2 value of 0.00019 renders the line meaningless. Considering the utility of the 1:1 line to orient the reader,

we retain the 1:1 line but lighten it to minimize any potential distraction. This adjustment has been applied consistently across all relevant figures. Additionally, we add explanations for the line of best fit in the revised Supplementary Information.

Supplement Line 78-80: "The r^2 value reflects the consistency in the relative spatial distribution between simulations and measurements, while the slope indicates whether the model systematically overestimates or underestimates the measurements."

b) It is written that the blue markers in Figure 2 are for the data mostly from North America and East Asia. On the other hand, one, it is written that in Europe r2 = 0.93 and slope = 1.2 (Lines 160-163). There are no explanations on the difference between Europe and North America (blue markers in Figure 2). This is inconsistent and confusing.

Reply: The results for Europe are based on complementary measurements from the European Monitoring and Evaluation Programme (EMEP), while the results shown in Figure 2 are derived exclusively from SPARTAN. Together, these evaluations demonstrate the skill of the model and the availability of relatively accurate emission data in these regions. To improve clarity, we revise the phrase "other available measurements for Europe" to "additional complementary measurements from the European Monitoring and Evaluation Programme (EMEP)" (Line 167) and include the following sentence in the revised manuscript.

Line 164-166: In addition to the evaluation based on SPARTAN data, we extend the analysis by incorporating measurements from other available sources for evaluation ("Methods" section).

Reviewer #4 (Remarks to the Author):

I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Review Feedback for Nature Communications manuscript NCOMMS-25-10179

This manuscript presents a dataset of globally distributed black carbon (BC) mass measurements obtained from the Surface Particulate Matter Network (SPARTAN) and complementary measurement networks to evaluate contemporary BC emission inventories. The authors employ the GEOS-Chem global chemical transport model in its high-performance configuration (GCHP) for high-resolution simulations to link BC emissions with ambient concentrations, facilitating comparison with the observational dataset. The results indicate that the model systematically underestimates BC concentrations at most Global South sites, with a normalized mean bias of -38%.

While previous studies have relied primarily on different models and satellite-based data, this manuscript underscores the necessity of long-term surface monitoring, particularly in the Global South. Prior studies that utilized satellite estimates often reported annual mean values, but such estimates should be interpreted with caution due to the increased error in regions lacking ground-based BC measurements. This study integrates ground-based measurements with modeling approaches/Simulations to demonstrate that BC emissions tend to be underestimated in the Global South compared to the Global North.

Although the manuscript is relevant to the scope of the journal, the novelty of the data presented is somewhat limited. The study does not introduce fundamentally new findings beyond those reported in previous characterizations of BC emissions, particularly in the Global South. However, the value of this work lies in the scarcity of comparable datasets from this region and its integration of ground-based observations. Given the significance of the study and the robustness of the analysis, I recommend the manuscript for publication. The scarcity of BC measurements in the Global South, particularly in Africa, makes this study highly relevant to the scientific community.

General Comments for Improvement

A key strength of this study is the acquisition of a comprehensive dataset of BC concentrations from globally distributed SPARTAN sites. However, the authors have utilized datasets that are inherently challenging to compare due to differences in instrumentation, sampling objectives, sampling heights (e.g., rooftop placement), and sampling periods.

- The authors consistently refer to "developing countries" throughout the manuscript. It would be more appropriate to use "low- and middle-income countries" (LMICs) to ensure inclusivity. Several arguments have been made

against the use of the term "developing world," and adopting LMICs would enhance precision and consistency with contemporary discourse.

- The authors should provide further discussion on the source contributions of BC emissions in the datasets used. For example, BC emissions in Africa arise from diverse sources, including wildfires, biomass combustion, and vehicular traffic, with the relative contributions varying across countries. These differences should be explicitly highlighted and discussed in the manuscript.

- The study includes 2,257 filter samples collected from 22 SPARTAN sites between 2019 and 2023. This period encompasses the COVID-19 lockdowns, during which human activities, particularly those contributing to BC emissions, were significantly reduced (by approximately 80% in some regions). The authors should clarify whether the data collected during this period were included in their comparisons and discuss the potential impact of pandemic-related emission reductions on their findings.

- The manuscript references BC measurements in Africa but includes data from only four sites across three African countries. These limited sampling locations may not provide a sufficiently representative characterization of BC emissions across the continent or Sub-Saharan Africa. The authors should acknowledge this as a limitation of the study and discuss its implications.

- In Figure 1, New Zealand appears to be missing from the map. The authors should clarify whether this omission was intentional or an oversight.