

Response to Specific Comments from Reviewer 1

Line 43 While I agree that much of the terminology in this field is operationally defined, the terms EC, soot, and others listed here overlap. This sentence needs correction and rephrasing.

Line 43-46 now reads as “BC is somewhat poorly defined, but is generally considered to be insoluble, refractory, and has an absorption exponent near one. Other materials such as elemental carbon (EC), and soot (Wei et al., 2013) are often very similar to BC, but each is operationally defined by how it is measured.”

Line 77 As the authors mentioned the mixing state and absorption enhancement in this paragraph, more details on BC heterogeneity, which mitigates the theoretical enhancement, should be discussed here.

Discussion about the absorption enhancement caused by changes in BC morphology and absorbing shell versus pure scattering shell are now added into this paragraph as suggested. Line 74-83 now reads as “Observations of absorption enhancement from ambient BC vary widely in field studies due to variations in coating thickness, coating material, source type, or methodological differences, but it is often much lower than laboratory values (Liu et al., 2015, 2017; Cappa et al., 2012, 2019; Healy et al., 2015; Krasowsky et al., 2016). Cappa et al. (2019) summarized absorption enhancements observed at the red end of the visible spectrum from 10 studies including ambient measurements, source sampling, and lab experiments. The absorption enhancement reported by those measurements ranged from 1.1 to 2.8. Lack et al. (2010) found that the absorption enhancement caused by the absorbing shell would be smaller than the absorption enhancement caused by the pure scattering shell. The non-spherical morphology of BC and the tendency of BC to compact when coated by organics also can both enhance and decrease absorption (Romshoo et al., 2021; Kelesidis et al., 2022).”

Romshoo, B., Müller, T., Pfeifer, S., Saturno, J., Nowak, A., Ciupek, K., Quincey, P. and Wiedensohler, A.: Optical properties of coated black carbon aggregates: Numerical simulations, radiative forcing estimates, and size-resolved parameterization scheme, *Atmos. Chem. Phys.*, 21(17), 12989–13010, doi:10.5194/acp-21-12989-2021, 2021.

Kelesidis, G. A., Neubauer, D., Fan, L. S., Lohmann, U. and Pratsinis, S. E.: Enhanced Light Absorption and Radiative Forcing by Black Carbon Agglomerates, *Environ. Sci. Technol.*, 56(12), 8610–8618, doi:10.1021/acs.est.2c00428, 2022.

Line 79 The expression is not professional enough and is too colloquial. This issue is prevalent throughout the manuscript.

Line 84 now reads as “The mass absorption cross section of BC (MAC_{BC}) is an alternative method to quantify the absorbing ability of BC containing particles versus absorption enhancement.” While we have changed this particular sentence, we disagree with the reviewer’s comment that the manuscript overall is too colloquial and not professional enough. All the co-authors have published extensively and nearly all native speakers of English, who have reviewed and edited this manuscript multiple times. We have reviewed the manuscript yet again and feel there are minimal grammar or phrasing issues that have not been resolved at this point. We strongly believe that scientific papers should, to the extent possible, avoid jargon and complex phrasing in an effort to explain things in a straightforward way for readers to understand.

Line 81 "At a certain wavelength" might be a better phrasing.

Line 87 now reads as “ MAC_{BC} is the particulate absorption divided by the mass of the pure BC at a certain wavelength” as suggested.

Line 86 to Line 93 The authors only list results from previous publications without providing their own summary. A synthesis or summary is needed here.

Line 92-100 now reads as “Bond and Bergstrom (2006) suggested a MAC_{BC} of $7.5 \pm 1.2 \text{ m}^2 \text{ g}^{-1}$ at 550 nm for fresh BC. The following campaigns demonstrate the variety of MAC_{BC} measured in the ambient during the past 15 years. Subramanian et al. (2010) reported a MAC_{BC} of $10.9 \pm 2.1 \text{ m}^2 \text{ g}^{-1}$ at 660 nm and $13.1 \text{ m}^2 \text{ g}^{-1}$ at 550 nm over Mexico City when using a single particle soot photometer (SP2) and the filter-based particle soot absorption photometer (PSAP) instrument during airborne measurements. Krasowsky et al. (2016) reported a MAC_{BC} enhancement of 1.03 ± 0.05 due to the coatings on BC. Zhang et al. (2017) found a MAC_{BC} with a mean of $10 \text{ m}^2 \text{ g}^{-1}$ and a standard deviation of $4 \text{ m}^2 \text{ g}^{-1}$ at 660 nm by using both SP2 and PSAP measurements. Cho et al. (2019) summarized MAC_{BC} estimated from more than 10 studies in East and South Asia in both ambient conditions and laboratory experiments, and the values ranged from 4.6 to $11.3 \text{ m}^2 \text{ g}^{-1}$.”

Line 126 There is no need to include this basic equation in the main manuscript.

This equation was requested by the second reviewer. We think this equation might also be helpful to people who don't have experience on airborne measurements.

Line 249 In Equation 2, E_{abs} includes the effects of both lensing and BrC. If using a wavelength of 660 nm, BrC might be negligible. The same applies to Equation 3. The wavelength used should be specified here, rather than just using lambda.

Thank you for pointing out this. We only compared E_{abs} and MAC_{BC} at 660 nm in this work. Line 257-268 now reads as “Absorption enhancement (E_{abs}) is the ratio of the absorption of all particles (including BC core and coating materials) to the absorption of BC alone (Lack and Cappa, 2010). E_{abs} at 660 nm ($E_{abs,660}$) was calculated in this study by Eq. 2:

$$E_{abs,660} = \frac{Abs_{Total,660}}{M_{BC} * MAC_{BC,core,660}} \quad (Eq. 2)$$

where $Abs_{Total,660}$ is the total absorption coefficient at a wavelength of 660 nm measured by the PAS, M_{BC} is the mass concentration of BC measured by the SP2, and $MAC_{BC,core,660}$ is the MAC of BC alone (without any other coating material) at 660 nm, which is set to be $6.3 \text{ m}^2 \text{ g}^{-1}$ (Bond and Bergstrom, 2006; Subramanian et al., 2010).

MAC_{BC} at 660 nm was calculated following Eq. 3:

$$MAC_{BC,660} = \frac{Abs_{Total,660}}{M_{BC}} \quad (Eq. 3)$$

Line 311 Did Mie factors vary significantly in each plume?

We only calculate plume integrated Mie factors to avoid noise caused by rapid changes within a plume. The variation of Mie factors within the same plume is beyond the scope of the current publication.

Line 362 One reason for discrepancies might be that Saleh's study focused on emissions, while samples collected during WE-CAN might have experienced some degree of aging.

Agreed. Line 371-373 now reads as “The discrepancy could also be partly because the data Saleh et al. used for their parameterization comes from controlled laboratory burns and not wildfires or because emissions observed during WE-CAN have all undergone some near-source aging before being observed by the aircraft.”

Line 376 The change in the absorption coefficient is due to changes in RI, MAC (intrinsic properties), and mass. Since the mass concentration of chromophores is unknown, most studies normalize the absorption coefficient to the OA concentration to obtain the MAC value. The basic assumption that all OA absorb equally need to specify in the context.

Line 387-392 now reads as “While only a fraction of organic aerosol mass absorbs light, both models and observations typically treat all organics as equally absorbing because of the inability to distinguish absorbing and non-absorbing molecules. The definition of bleaching or whitening was unclear in previous literature. Models tend to treat bleaching as the change of refractive index or decreasing of MAC (Brown et al., 2018; Wang et al., 2018; Carter et al., 2021), while observations or lab experiments mostly link bleaching to the decrease of normalized total absorption (Forrister et al., 2015; Palm et al., 2020; Zeng et al., 2022).” This is also specified in section 2.4 after equations. Line 267-279 now reads as “MAC of BrC and lensing is calculated at 405 and 660 nm (Eq. 4):

$$MAC_{BrC+lensing_\lambda} = \frac{Abs_{Total_\lambda} - M_{BC} * MAC_{BC_core_\lambda}}{M_{OA}} \quad (Eq. 4)$$

where M_{OA} is the organic mass measured by the AMS. Again, the $MAC_{BC_core_\lambda}$ is set to be 6.3 and 10.2 $m^2 g^{-1}$, respectively, at 660 nm and 405 nm yielding an absorption Ångström exponent (AAE, the negative slope of a logarithmic absorption coefficient against wavelength) of 0.99 for the BC core (Bond and Bergstrom, 2006; Subramanian et al., 2010; Liu, et al., 2015). It should be noted that both BrC and lensing contribute to the $MAC_{BrC+lensing_\lambda}$, and cannot be separated using this approach and $MAC_{BrC+lensing_\lambda}$ is the MAC of all organics without distinguishing absorbing and non-absorbing particles.

MAC of water-soluble BrC at λ nm ($MAC_{ws_BrC\lambda}$) is calculated using Eq. 5:

$$MAC_{ws_BrC\lambda} = \frac{Abs_{ws_BrC_\lambda}}{WSOC * (WSOM:WSOC)} \quad (Eq. 5)$$

where $Abs_{ws_BrC_660}$ is water-soluble light absorption and WSOC is water-soluble organic carbon mass, which are both measured by the PILS system. WSOM:WSOC ratio is set to be 1.6 (Sullivan et al., 2022). $MAC_{ws_BrC\lambda}$ is the MAC of all water-soluble organics without distinguishing absorbing and non-absorbing particles.”

Line 440 How do the authors define a near-source flight?

We are referring to the plumes where we directly followed the emissions from as near to the flame front as we were allowed to go. Line 452-454 now reads as “Despite the much longer transit time and distance, overall these emissions, which were measured 300 to 600 miles away, have a very similar $MAC_{BrC+lensing_405}$ to that of the near-source flights where we tracked emissions from as near to the fire source as allowed by air traffic control.”

Line 445 I suggest the authors examine BC with CO to align the data better.

We aligned all the measurement with CO, but the SP2 data sometimes would show a hysteresis due to the manually dilution, which is to prevent signal saturation. This is not an issue when using plume integrated method, because the whole peak of SP2 will be integrated. However, the plume is highly mixed during RF05 and RF08, and the plume integration method is not applicable anymore. Time averaging is applied for these two flights and can not fully removing the extreme values caused by the SP2 hysteresis. Line 458-460 now reads as “There are several extreme values that exist in the dataset, probably because of the SP2 hysteresis caused by variation in the dilution rate of the SP2 which cannot be totally eliminated from the 1-minute average.”

Line 458 Have the authors accounted for the Mie factor? I suggest using different symbols to represent absorption in liquid or in air.

$Abs_{BrC+lensing_405}$ is the absorption coefficient of water-soluble BrC at 405 nm, which is directly measured by PILS, and does not account for the Mie factor. Line 472-474 now reads as “The average water-soluble BrC absorption at 405 nm (Abs_{ws_BrC405} , 0.02 Mm-1 ppbv-1) which is directly measured by the PILS, is only 20% of the total absorption from BrC plus lensing ($Abs_{BrC+lensing_405}$, 0.11 Mm-1 ppbv-1), which is calculated from the PAS and SP2 (Eq.9).

Line 506 Absorption at 660 nm could also be due to the penetration of BC particles into the filter. A 0.2 µm filter is not sufficient to filter out all BC particles. How can the authors rule out the presence of BC?

According to Peltier et al. (2007), black carbon adheres to the PILS impactor and is not efficiently transferred to the liquid stream. The inline 0.2 µm liquid filter also helps remove more. They also pointed out that black carbon is also not oxidized by the TOC Analyzer, and any insoluble particles greater than 110 nm could not be oxidized by the TOC Analyzer based on tests with PSL. Line 521-523 now reads as “We know that absorption observed in the PILS at 660 nm is not BC because BC is insoluble and will be removed by the PILS impactor, the 0.2 µm filter in the instrument, and that BC over 110 nm in size will not be oxidized by the TOC analyzer (Peltier et al., 2007; Zeng et al., 2021; Sullivan et al., 2022).”

Response to Specific Comments from Reviewer 2

In section 2.1, the authors do not justify (with literature citations or otherwise) the conversion of optical properties to STP. I have never seen this done before, which does not mean it is an incorrect procedure, but the authors should add detail about what errors in (specifically) optical properties would arise if the data were not corrected, and cite previous literature showing the background and scientific basis of this procedure. Note that this is different than the pressure-dependent correction to the photoacoustic absorption signal.

The instruments on the aircraft sampled air through different inlets that had different residence times and thus the measured air arrived at the instruments at different temperatures and pressures. To make instruments comparable, the measurement must be converted to either ambient conditions or STP. STP conversion was chosen during WE-CAN for two reasons 1.) It introduces less error because only the measured temperature and pressure at the instrument location is used instead of also needing the measured temperature and pressure of the ambient air and 2.) it eliminates artifacts that would arise from air being measured at different altitudes having the same mixing ratios, but different mass concentrations. Mixing ratios are typically used for the gas-phase species (ppb etc.) and these are constant with variations in pressure if the amount of species remains constant. However, aerosol properties are typically reported as densities or concentrations ($\mu\text{g}/\text{m}^3$, etc.) because we don't know the molecular weight of the aerosol and cannot easily generate a mixing ratio. The concentrations will change with pressure even if the mixing ratios do not, therefore it is best practice to convert all concentrations to STP for airborne data to avoid a situation where mixing ratios remain constant with pressure while concentrations change even though the relative amounts of the species are constant. This is especially true for our study where we are interested in chemical changes causing changes in the ratio of aerosol to gas and not changes that would occur purely because of pressure changes. Optical properties (mass absorption coefficient, etc.) also need to be adjusted in the same way to be consistent, otherwise they would also vary with pressure. As noted in the paper, the entire WE-CAN dataset (https://data.eol.ucar.edu/master_lists/generated/we-can) has been converted to STP for these reasons and this data has been used in numerous other papers from WE-CAN including Palm et al., 2020 and Sullivan et al., 2022, which are already cited in the manuscript. The errors introduced by this conversion, which are small because we can accurately measure both temperature and pressure in the inlet lines, were included in the posted data.

Palm, B. B., Peng, Q., Fredrickson, C. D., Lee, B. H., Garofalo, L. A., Pothier, M. A., Kreidenweis, S. M., Farmer, D. K., Pokhrel, R. P., Shen, Y., Murphy, S. M., Permar, W., Hu, L., Campos, T. L., Hall, S. R., Ullmann, K., Zhang, X., Flocke, F., Fischer, E. V. and Thornton, J. A.: Quantification of organic aerosol and brown carbon evolution in fresh wildfire plumes, *Proc. Natl. Acad. Sci. U. S. A.*, 117(47), 29469–29477, doi:10.1073/pnas.2012218117, 2020.

Sullivan, A. P., Pokhrel, R. P., Shen, Y., Murphy, S. M., Toohey, D. W., Campos, T., Lindaas, J., Fischer, E. V. and Collett, J. L.: Examination of Brown Carbon Absorption from Wildfires in the Western U.S. During the WE-CAN Study, *Atmos. Chem. Phys. Discuss.*, (July), 1–29, doi:10.5194/acp-2022-459, 2022.