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Is Black Carbon an appropriate generalization in climate studies?

¿Es el Carbono Negro una generalización adecuada en estudios climáticos?

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ABSTRACT: Soot aerosols produced during incomplete combustion processes are agglomerates composed of quasi-spherical carbonaceous primary particles with markedly different arrangements (shape, size, and internal structure) and varying compositions. Although they have been considered to be one of the main contributors to climate change, it remains difficult to quantify their global warming potential (GWP) precisely. Exact knowledge of the optical properties of soot agglomerates (black carbon -BC) would be necessary to fairly establish a CO_2 equivalent GWP. BC warming effects depend on their size and shape, and despite their limited residence time in the atmosphere, their high radiative forcing would lead to GWP $\sim$ 2000. The quantification of BC concentrations in the atmosphere is often based on optical measurements, which usually lack accuracy. Our calculations on the direct radiative forcing of BC showed that the irregularity of the agglomerate, and their size markedly affect the cooling/heating capacity due to extreme variations of its absorbing and scattering characteristics. In addition, the estimation of the incremental radiative forcing of aerosols is challenging since it is highly dependent on local parameters such as cloudiness, surface albedo, aerosol concentration, etc., which are highly variable worldwide. Based on that, we do not recommend using a unique CO_2 equivalent GWP for BC.

RESUMEN: Los aerosoles de hollín producidos durante procesos de combustión incompleta son aglomerados compuestos por partículas primarias carbonáceas cuasi-esféricas con disposiciones (forma, tamaño, estructura interna) marcadamente diferentes y composición variable. Aunque son uno de los principales contribuyentes al cambio climático, sigue siendo difícil cuantificar con precisión su potencial de calentamiento global (GWP). Sería necesario un conocimiento preciso de las propiedades ópticas de los aglomerados de hollín (carbón negro - BC) para establecer de manera justa un GWP equivalente a CO_2. Los efectos de calentamiento del BC dependen de su tamaño y forma, y a pesar de su tiempo de residencia limitado en la atmósfera, su alta fuerza radiativa conduce a GWP $\sim$ 2000. Cuantificar las concentraciones de BC en la atmósfera a menudo se basa en mediciones ópticas, que generalmente carecen de precisión. Nuestros cálculos sobre el forzamiento radiativo directo del BC mostraron que la irregularidad del aglomerado, así como su tamaño, afectan marcadamente su capacidad de enfriamiento/calentamiento debido a las variaciones extremas de sus características de absorción y dispersión. La estimación del forzamiento radiativo incremental de los aerosoles es un desafío, ya que depende de parámetros locales como la nubosidad, el albedo de la superficie, la concentración de aerosoles, etc., los cuales son variables en todo el mundo. En base a eso, no recomendamos utilizar un GWP equivalente a CO_2 único para el BC.

1. Introduction

Soot is formed from incomplete combustion processes, in which transport is the main emitting sector. Soot particles

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are often coated with adsorbed liquid hydrocarbons, which may come from the fuel itself or, in the case of transport vehicles, even from the engine lubricating oil. Once diluted in the atmosphere in the form of aerosols, different equivalent nomenclatures are used to refer to this carbonaceous matter, such as soot particles (technologists), elemental carbon (chemists), or black carbon (environmentalists). When soot is coated with hydrocarbons (organic carbon for chemists), environmentalists designate it as brown carbon. No matter how it is denoted, soot aerosols are agglomerates (or aggregates) composed of quasi-spherical primary particles with very different arrangements (See Figure 1, left). Soot agglomerates are considered one of the main contributors to climate change. However, the quantification of this effect is difficult because the optical and radiative properties of soot agglomerates depend on their composition, size, and shape, internal structure (see Figure 1, right). Therefore, they also depend indirectly on the source and combustion conditions in which they are generated.

Small and irregular agglomerates have high absorption efficiencies with a consequent dominating heating effect, whereas larger and more compact particles have high scattering efficiencies, possibly with a dominating cooling effect. Moreover, they have different atmospheric residence times (from seconds to months) depending on their size and shape. When particles are deposited on earth, their effect depends on how reflective the surface is, being snow surfaces the most sensitive.

Knowledge about the spectral properties of soot agglomerates is essential to evaluate their climatic impact, both when they are suspended as aerosols and when deposited on the surface. Despite this, a vast majority of environmental studies ignore the specific characteristics of soot particles, or they simply consider them as spherical. Even more, a few of them consider that only soot agglomerates derived from the combustion of fossil fuels contribute to climate change, or that the contribution of fossil fuels and biofuels is intrinsically different [1], disregarding that soot particles can be emitted in different amounts depending on the combustion system and the type of fuel. This is just an indirect effect based on the combustion conditions, the chemical structure, and the oxygen content of the fuel [2].

2. The global warming potential of black carbon

Different parameters have been defined to quantify the impact on climate, such as radiative forcing (RF), global temperature potential (GTP) (proposed by [3]), and global warming potential (GWP) (suggested by IPCC in 1990 and defined in [4]). Quantification of the radiative properties of soot considering realistic particulate morphology, composition, and internal structure, would be useful for a hierarchical prioritization of environmental policies (traffic restrictions, prediction of hydrological resources), a guide for technological developments (filter design, engine control strategies), and a database for scientific developments (emission factors, dispersion models). Among the parameters mentioned, GWP is the most frequently used. It is defined as the cumulative radiative forcing (left ratio in Equation 1), including both direct and indirect effects, over a specified time horizon (Δt) resulting from the emission of a unit mass of contaminant *i* with respect to that of CO_2, as shown in Equation 1. Among the three-time horizons proposed by IPCC (20, 100, and 500 years), the usual time horizon employed is 100 years, although 20 years is also often used.

$$GWP_i = \frac{\int_{\Delta t} RF_i \, \mathrm{d}t}{\int_{\Delta t}^{m} RF_{CO_2} \, \mathrm{d}t} = \frac{\int_{\Delta t} a_i C_i \, \mathrm{d}t}{\int_{\Delta t}' a_{CO_2} C_{CO_2} \, \mathrm{d}t} \qquad (1)$$

Since the radiative forcing is proportional to the concentration of the contaminating component in the atmosphere, it can also be expressed (right ratio) as a function of the contribution of a unit mass of the component to the radiative forcing (a_i).

Very different values of GWP have been proposed for combustion-derived particles. Jacobson et al. proposed a GWP value for brown carbon of 95 to 191, with no discrimination between the underlying soot nucleus and the adsorbed hydrocarbons [5]. Later on, Jacobson *et al.* evaluated the light absorption capacity and the residence time of soot particle matter with respect to CO_2 (500000/1 and 1/240, respectively), which would lead to a GWP for BC of 2083, if other regional effects such as altered precipitation patterns are neglected [6]. Different values for 20 and 100 years have been proposed. For example, [7] proposed values for GWP(20) and GWP(100) for black carbon, of 2200 and 680, respectively. Contrary to the proposals by [5, 6], these values were obtained from integrating of instantaneous radiative forcing over time, following the conceptualization of equation 1. The same values were further adopted by [8] to assess the benefits of using liquified petroleum gas in transport in front of other fuels and by [9] to evaluate the environmental benefits of diesel particulate filters in diesel trucks.

Other authors, including [1] have proposed slightly lower values in the last two decades, GWP(20) = 1600 and GWP(100) = 460 (although these values were calculated by [9] from radiative forcing (RF) data), and [10], GWP(20) = 1600. Authors in [11] proposed an average value of GWP(100) = 480, after considering the differences in regional warming impact among different regions in the world. A similar study to that from [9], although focused

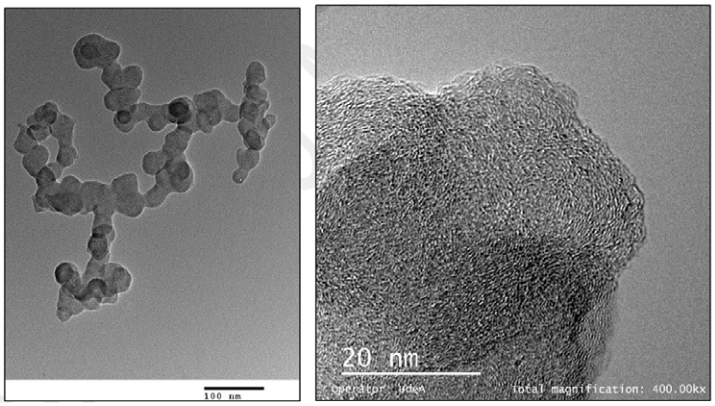

Figure 1 Example of soot agglomerate (left) and detail of its internal structure (right)

on the impact of filters in all types of vehicles, has been recently published using a GWP(20) = 2000, selected as an average of different values proposed in the literature [12]. In this publication, the authors justify the use of a unified value of GWP stating that: *"although engine soot is quite black when emitted, it is not pure black carbon (BC) as physically defined or elemental carbon (EC) since, the warming impact of black carbon, depending on the composition, surface color can change a bit but this takes time, and since we discuss a very limited residence time in the atmosphere this simplification of using BC-properties might be permitted"*. However, different from all these environmental studies, the effect of the size, shape, composition, and internal structure of soot agglomerates should not be neglected or simplified [13].

Only a few studies have proposed different warming effects depending on the size and shape. [14] revised previous calculations and assumed that *"realistic BC particle shapes"* increased the optical absorption effectiveness by a factor of 2, but still proposed generic values of GWP(20) = 2000 and GWP(100) = 500. More recently, [15] estimated that the contribution of cluster-dense aggregates formed in wildfires, with sizes larger than 1 micron and high fractal dimensions (higher than 2.5), was around 35 % higher than freshly emitted ultrafine soot aggregates (with fractal dimensions around 1.8), and 90 % more than the equivalent spheres.

Disregarding the indirect radiative forcing effects, such as those related to the effect of aerosols on cloud formation and properties or to the effect on the ground albedo when deposited, and the lifetime of aerosols, the direct radiative forcing of a layer of aerosols has often been estimated as follows (Equation 2):

$$DRF = -I_{irrad} D T_{atm}^2 (1-n)\left[\left(1-\omega_s^2\right)\beta\tau_{sca} - 2\omega_s\tau_{abs}\right] \quad (2)$$

where I_{irrad} is the globally-averaged incident solar flux at the top of the atmosphere, n is the fraction of clouds in the sky, T_{atm} refers to the atmospheric transmittance, and D is the fractional day length, which depends on the location and the season. The expression inside the brackets corresponds to the change in direct radiative forcing due to the presence of a layer of aerosols, which depends on the broadband albedo of the surface (ω_s), the upscatter fraction of the aerosols (β), and the scattering and absorption optical depths $\left(\tau_{\underline{sca}}, \tau_{abs}\right)$.

The upscatter fraction for a spherical aerosol can be estimated by integration of the scattering phase function of the aerosol over the cosine of the solar zenith angle along the sunlit hemisphere. This integration has usually been developed for spherical particles (using the Mie scattering phase function or simplified ones such as the Henyey-Greenstein function [15, 16] but can hardly be estimated accurately for irregular agglomerates. The optical depths can be quantified from the aerosol concentration and their absorbing and scattering characteristics, universally expressed through their mass-specific optical cross sections (MAC for absorption and MSC for scattering). If Equation 2 is expressed as a function of the absorbance $(A_a = \tau_{abs})$ and reflectance $(R_a = \beta\tau_{sca})$ of the aerosol layer and parametrized for varying values of A_a and R_a, the resulting direct radiative forcing is shown in Figure 2 (for a surface broadband albedo $\omega_s = 0.18$, typical of urban environments; left panel, and for a surface broadband albedo $\omega_s = 0.8$, typical for clean snow surfaces, the most reflective surfaces on earth, right panel).

If DRF is positive the aerosol layer contributes to climate warming, whereas if it is negative, it contributes to cooling. It can also be observed in Figure 3 (obtained with the Optipar model, using the Rayleigh-Debye-Gans approximation [17] that absorption and scattering of specific cross sections increase with increasing fractal dimension (a measure of the irregularity of the

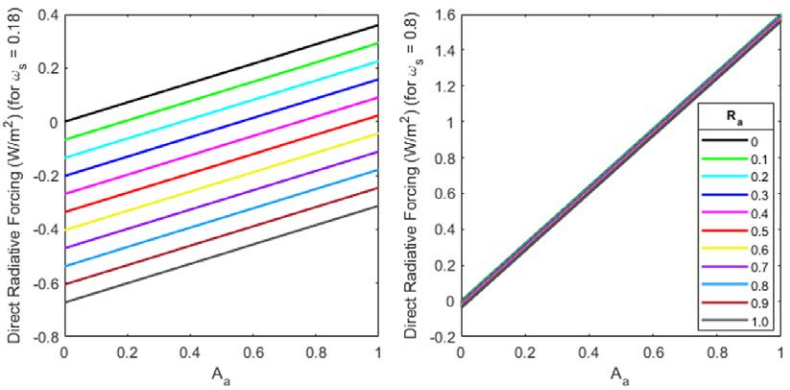

Figure 2 Direct radiative forcing of an aerosol layer above an urban surface (left) and above a clean snow surface (right)

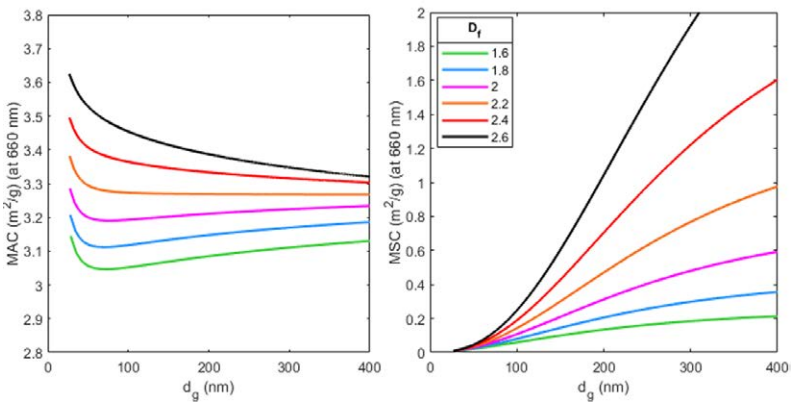

Figure 3 Effect of the agglomerate size (quantified with the diameter of gyration) on the mass-specific absorption (left) and scattering (right) cross sections for agglomerates with different fractal dimensions (at a wavelength of 660 nm as an example). Results were obtained with the software OptiPar [17].

agglomerate) and that the scattering cross section increases with the agglomerate size (quantified with its diameter of gyration). Since soot agglomerates are hardy larger than 300 nm and their fractal dimensions are in the range of 1.6-2.6, absorption is higher than scattering, and, therefore, low reflectance values can be expected for the soot aerosol layers. On the contrary, if the aerosol concentration is high, high absorbance values could be expected. Under these conditions, a noticeable heating effect can be achieved, especially if the layer is suspended over bright surfaces.

The warming effect of soot aerosols is very variable and difficult to quantify due to the high sensitivity of the radiative forcing to the size/shape of the agglomerates, to the internal composition and structure of the graphite-like material, to the black carbon concentration, and to the local conditions. Specifically, the quantification of black carbon concentrations in the atmosphere is often based on optical measurements, which usually lack accuracy. In fact, much of the misleading interpretation of the effect of black carbon in environmental studies possibly derives from commercial aethalometers, most of which attribute exclusively to black carbon the light absorption intensity at a wavelength of 880 nm, ignoring that any type of light-absorbing particle (LAP) absorbs light along the entire light spectrum. In fact, soot particles absorb even more light intensity in the visible range than in the infrared one, although it is true that they may show the main differences with respect to other LAP's (such as ash, sand, haze, or mineral dust) at around 880 nm.

3. Conclusions

Regarding our question of whether Black Carbon is an appropriate generalization in climate studies, we concluded that the estimation of the incremental radiative forcing of aerosols is challenging since it is highly dependent on local parameters such as the cloudiness, surface albedo, aerosol concentration, etc., which are highly variable around the world. In the case of soot agglomerates, the difficulty is even higher because their mass-specific cross sections highly depend on their size, shape, and internal structure. On the other hand, the upscatter fraction can be estimated by integration of

the scattering phase function of the aerosols, which are difficult to estimate in the case of agglomerates. The attribution of radiative forcing exclusively to black carbon aerosols is also very uncertain, because the quantification of black carbon concentration with optical measurements is uncertain.

Declaration of competing interest

We declare that we have no significant competing interests, including financial or non-financial, professional, or personal interests interfering with the full and objective presentation of the work described in this manuscript.

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Author contributions

Magín Lapuerta: Conceived and designed the discussion and analyses, original manuscript writing. Sofía González-Correa: Collected the data and performed data analyses and prepared the discussion. J. R. Agudelo: Conceived and designed the discussion, original manuscript writing.

Data available statement

Data included in this paper can be provided by the corresponding author J. R. Agudelo) upon request.

References

[1] M. Schulz, C. Textor, S. Kinne, Y. Balkanski, S. Bauer, T. Berntsen, and *et al.*, "Radiative forcing by aerosols from the aerocom present-day and pre-industrial simulations," *Atmospheric Chemistry and Physics*, vol. 6, no. 12, Dec. 2006. [Online]. Available: https://doi.org/10.5194/acp-6-5225-2006

[2] M. Lapuerta, F. Oliva, J. Agudelo, and A. Boehman, "Effect of fuel on the soot nanostructure and consequences on loading and regeneration of diesel particulate filters," *Combustion and Flame*, vol. 159, no. 2, Feb. 2012. [Online]. Available: https://doi.org/10.1016/j.combustflame.2011.09.003

[3] K. Shine, J. Fuglestvedt, K. Hailemariam, and N. Stuber, "Alternatives to the global warming potential for comparing climate impacts of emissions of greenhouse gases," *Climatic Change*, vol. 68, Feb. 2005. [Online]. Available: https://doi.org/10.1007/s10584-005-1146-9

[4] P. Forster, V. Ramaswamy, P. Artaxo, T. Berntsen, R. Betts, D. Fahey, and *et al.*, *Changes in Atmospheric Constituents and in Radiative Forcing*. Cambridge, United Kingdom and New York, NY, USA: Cambridge University Press, Oct. 2007.

[5] M. Jacobson, W. Colella, and D. Golden, "Cleaning the air and improving health with hydrogen fuel-cell vehicles," *Science*, vol. 308, no. 5730, Jun. 2005. [Online]. Available: https://doi.org/10.1126/science.1109157

[6] M. Jacobson, "Review of solutions to global warming, air pollution, and energy security," *Energy Environ. Sci.*, vol. 2, Oct. 2008. [Online]. Available: https://doi.org/10.1039/B809990C

[7] T. Bond and H. Sun, "Can reducing black carbon emissions counteract global warming?" *Environmental Science and Technology*, vol. 39, Jul. 2005. [Online]. Available: https://doi.org/10.1021/es0480421

[8] A. Research. (2017, Nov.) clearing the air: black carbon, climate policy and lp gas. [Online]. Available: https://silo.tips/download/clearing-the-air-black-carbon

[9] L. Hill. (2019, Sep.) The carbon dioxide-equivalent benefits of reducing black carbon emissions from u.s. class 8 trucks using diesel particulate filters: A preliminary analysis. [Online]. Available: https://tinyurl.com/2fmfyxfj

[10] M. Walsh, M. Nichols, A. Bremauntz, L. Rojas-Bracho, M. Williams, A. Friedrich, and *et al.* (2009, Jun.) A policy relevant summary of black carbon climate science and appropriate emission control strategies. The internantional council on clean transportation. [Online]. Available: https://tinyurl.com/59jyv8kx

[11] M. Reddy and O. Boucher, "Climate impact of black carbon emitted from energy consumption in the world's regions," *Geophysical Research Letters*, vol. 34, no. 11, Jun. 2007. [Online]. Available: https://doi.org/10.1029/2006GL028904

[12] A. Mayer, J. Mayer, M. Wyser, F. Legerer, J. Czerwinski, T. Lutz, and *et al.*, "Particulate filters for combustion engines to mitigate global warming. estimating the effects of a highly efficient but underutilized tool," *Emission Control Science and Technology*, vol. 10, Jan. 2024. [Online]. Available: https://doi.org/10.1007/s40825-023-00236-x

[13] M. Lapuerta, R. Ballesteros, and S. González-Correa, "Relationships between morphology and optical properties of vehicle-emitted soot," *Journal of Aerosol Science*, vol. 174, Nov. 2023. [Online]. Available: https://doi.org/10.1016/j.jaerosci.2023.106261

[14] J. Hansen, M. Sato, P. Kharecha, G. Russell, D. Lea, and M. Sidall, "Climate change and trace gases," *Transactions of the Royal Society A*, vol. 365, no. 1856, May. 2007. [Online]. Available: https://doi.org/10.1098/rsta.2007.2052

[15] R. Chakrabarty, N. Beres, H. Moosmüller, S. China, C. Mazzoleni,

M. Dubey, and *et al.*, "Soot superaggregates from flaming wildfires and their direct radiative forcing," *Scientific Reports*, vol. 4, no. 5508, Jul. 2014.

[16] H. Moosmüller and J. Ogren, "Parameterization of the aerosol upscatter fraction as function of the backscatter fraction and their relationships to the asymmetry parameter for radiative transfer calculations," *Atmosphere*, vol. 8, no. 8, Jul. 2017. [Online]. Available: https://doi.org/10.3390/atmos8080133

[17] M. Lapuerta, S. González-Correa, R. Ballesteros, R. Cereceda-Balic, and H. Moosmüller, "Albedo reduction for snow surfaces contaminated with soot aerosols: comparison of experimental results and models," *Aerosol Science and Technology*, vol. 56, no. 9, Jul. 2022.