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GEOSCIENCES

Relative contributions of fossil fuel and biomass burning sources to black carbon aerosol on the Southern Atlantic Ocean Coast and King George Island (Antarctic Peninsula)

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Abstract: Carbonaceous aerosols can affect climate, especially particles containing black carbon (BC). BC originated from the incomplete combustion of fossil fuel and biomass, which can heat the atmosphere and increase ice melting, but little is known about BC sources to Antarctica. We quantified the contribution of distant origin (biomass burning) and local emissions (fossil fuel) to atmospheric BC concentration in the King George Island (Antarctic Peninsula) and the Southern Ocean. We examine the BC concentrations using a multi-wavelength Aethalometer AE-33 and AE-42 aboard the Brazilian Oceanographic Research Ship Almirante Maximiano. The results indicate that the region is influenced by local sources and air masses coming from surrounding continents. Fossil fuel combustion was the major source of carbonaceous aerosols in the region, whereas the total average concentration was 41.8 ± 22.8 ng m^{-3}. The findings indicate a contribution of biomass burning coming from low and mid-latitudes of South America over the Antarctic Peninsula and the Southern Ocean around 62ºS latitude. We demonstrated that fossil fuel is the main contributor to atmospheric BC concentration for the Austral summer and autumn. Scientific stations, local tourism, and traffic are possible local BC sources. Our work invokes the urgency of questionable sustainability issues about Antarctica exploration.

Key words: Antarctica, Southern Ocean, aerosols, black carbon, climate changes, Aethalometer.

INTRODUCTION

The Southern Ocean has a large impact on atmospheric and oceanic circulations in the Southern Hemisphere and globally. The exchange fluxes of ocean and aerosols are essential to understand the behavior of particles in coastal areas (Chaubey et al. 2011). The Antarctica continent was the last region on the Earth to be reached and explored, where most of its territory is untouched by man. This polar environment provides the unique opportunity to study the natural and background aerosols in the atmosphere and the interaction of particles with snow and ice, as well as the chance to qualify/quantify the influence of local human activities. The addition of human actions to natural greenhouse effects has been increasing the global warming in the Earth's temperature, mainly by emissions of greenhouse gases and aerosol components that can absorb radiation from the sun (Jacobson 2012).

The carbonaceous aerosols are the main component of tropospheric aerosols during biomass burning events and polluted atmospheres due to fossil fuel combustion

(Weller et al. 2013). One of the main components of carbonaceous aerosol is black carbon (BC). BC particles can be described as a solid carbonaceous material with a graphitic microstructure that absorbs solar radiation in ultraviolet (UV) and visible wavelengths (Long et al. 2013, Novakov 1984, Shrestha et al. 2014, Weller et al. 2013), produced during the incomplete combustion of fossil fuels and biomass. It is the major contributor to light-absorbing aerosol (and highly refractory) in the atmosphere (Bisiaux et al. 2012, Lan et al. 2013, Olson et al. 2015). The climate effects of BC aerosol play an important role in the Earth's atmosphere and cryosphere due to its direct positive radiative forcing (Jacobson 2001, Weller et al. 2013).

The BC fraction related to biomass burning is known as brown carbon (BrC), the organic component of carbonaceous aerosols. The BrC is known for its brownish color, absorbs light principally at the low visible wavelengths and the near UV (Andreae & Gelencsér 2006, Feng et al. 2013, Hopkins et al. 2007, Olson et al. 2015). Sources of anthropogenic BC emissions are commonly liked to: a) power plant that burns coal, oil, and natural gas; b) on-road vehicles, off-road utility vehicles, ships, and aircraft that use petroleum and diesel; c) residential cookstoves and industrial activities; d) agricultural and forests biomass burn (Klimont et al. 2017).

Some authors have considered the BC emissions (total average) as the second most important human emission related to climate forcing (+1.1 W m^{-2}), indicating as the positive forcing (Chung & Seinfeld 2002, Jacobson 2001) and with a considerable potential to warm the atmosphere (Bond et al. 2013). In terms of the global average, aerosol particles have contributed significantly to average forcing, comparatively with the anthropogenic CO_2 forcing (around +1.5 W m^{-2}). In contrast, at regional scale, they show that aerosol forcings can be an order of magnitude greater than greenhouse gas forcings (Satheesh & Ramanathan, 2000). Jacobson (2012) indicates that BC, mainly at fossil-fuel soot, can heat the air more than 1 million times more per unit mass in the atmosphere than $CO_{2(g)}$.

Regarding the importance of BC aerosols to atmospheric warming and ice melting, some studies have been concerning about BC present at Antarctica related to climate change, reporting the BC observations in terms of deposition on the ice sheet, air masses transportation, concentration levels, and their impacts on the Antarctic environment (Babu 2017, Bisiaux et al. 2012, Chaubey et al. 2010, Evangelista et al. 2007, Graf et al. 2010, Hansen et al. 2001, Hara et al. 2008, 2010, Pereira et al. 2006, Srivastava et al. 2021, Weller et al. 2013, Wolff & Cachier 1998).

The intensification of human activities in Antarctica, as scientific exploratory and tourism, has increased anthropogenic species' emissions, especially in austral summer (Babu, 2017, Graf et al. 2010, Pereira et al. 2006, Tin et al. 2009, Tomasi et al. 2007). The local impact of pollution has been growing, and the influences of BC has earned a notorious highlight due to the human interventions, mainly for common activities as incineration from scientific stations, emissions of fossil fuel burning by electricity energy power plant and by powered vehicles, combustion derived emissions from shipborne and aircrafts surrounding at Antarctica (Lynch et al. 2010, Pereira et al. 2006, Polyakov et al. 2020b, Weller et al. 2013).

The residence time of BC aerosols in the atmosphere is around 1-4 weeks (Lan et al. 2013), reaching a long distance (Ramanathan et al. 2007). Some studies have treated the physical and optical properties of BC aerosols as a priority (Hadley & Kirchstetter 2012, Koch & Hansen 2005), evoking the presence of the Antarctica aerosols on the spatial and temporal scales coming from different regions (Polian et

al. 1986, Weller et al. 2013). Furthermore, biomass burning continues to increase in South America due to deforestation and is one possible contributor to BC in Antarctica. Pereira et al. (2006) showed that central South America and Africa are significant sources of particles related to the coast and west of Antarctica.

Until now, the contribution of distant and local BC emission sources to the Antarctic Peninsula is not fully understood. In this work, we quantified the contribution of biomass burning (distant origin) and fossil fuel (local origin) to atmospheric BC concentration in the King George Island, Antarctic Peninsula, and in a stretch of the Southern Ocean.

MATERIALS AND METHODS

Online aerosols sampling: the Aethalometer measurements

The multi-wavelength Aethalometer® (Magee Scientific, Berkeley, CA USA) performs a real-time measurement sampling of BC aerosol through optical attenuation (absorbance) of light from LED lamps emitting at 7 wavelengths: 370, 470, 520, 590, 660, 880 and 950 nm. The lamps are responsible for the differentiation of the material collected according to their origin. Once aerosols, particles from various sources have distinctive spectra and, this way, transmit different intensities according to the wavelength of the light.

The aerosol sample is collected on the filter tape with a constant circular area (spot) and a background filter reference portion being used to control the stability of the optical source. The tape automatically advances when it reaches a load limit, depending on the concentration and flow rate. The advancement of the tape is by a predefined interval.

The optical performance of the Magee Scientific Aethalometer® is validated by the *Neutral Density Optical Filter Kit*, which uses NIST-traceable optical standards. Software routines measure the optical intensities at all wavelengths and compare the analysis instantly with the original reference values, verifying the analysis and validate the data. The Aethalometer is an absolute measurement based on sample flow, spot size, and optical attenuation (Hansen 2007).

We used two types of Aethalometer BC monitor sampler in this work: the model Aethalometer AE-33 (AE-33) and the Aethalometer AE-42 (AE-42). The main difference between each other is the filter substrate. While AE-33 uses a tetrafluoroethylene (TFE)-coated glass fiber filter tape, the AE-42 uses a quartz fiber filter tape. The TFE-coated glass fiber filter tape is less sensitive to air sample relative humidity. The AE-33 is an upgrade from other versions and uses a "dual spot," enabling Aethalometer to be used under dynamically varying ambient conditions. It provides valid BC measurement data out-of-the-box, with no need for post-processing.

The aerosol sampling for both devices was continuous and online, with a detection range <0.01 to >100 μg m^{-3}. The AE-33 performed high time resolution (fundamental rate of 1 Hz) analytical response (sampling, analysis, and all calculations), among the sensitivity to time-base and sample flow rate settings approximately 0.03 μg m^{3} and the detection limit (1 hour): <0.005 μg m^{-3}, with a resolution of 1 ng m^{-3}. The resolution and sensitivity of AE-42 is 0.1 μg m^{-3}. The collocated accuracy is 10% (Hansen 2007).

Due to the low concentration of BC in the Polar Area, the automatic advance of the tape was performed every twenty-four hours, and the flow rate was set at the maximum (5 L min^{-1}).

In situ BC aerosol measurements with AE-33

The aerosol samples were collected in situ BC measurements with AE-33 at Bellingshausen

Station (Russian Antarctica station, 62° 12' S. 58° 58'W), located at King George Island, on Fildes Peninsula (15.4 m asl, 120 km off the coast of Antarctica) (Figure 1), during the OPERANTAR XXXIII/2015.

The Bellingshausen station is located close-by the bases of Julio Escudero (Chile), Great Wall (China), General Artigas Station (Uruguay), King Sejong Station (Republic of Korea), and a bit further away from the Comandante Ferraz (Brazil), Henryk Arctowski (Poland) and Machu Picchu Base (Peru). The local additionally contains the flight's arrival point at Base Area Presidente Eduardo Frei Montalva (Fuerza Aerea de Chile), supporting all cargo arrivals and people from the surrounding stations. The locomotion between each station is by car and truck.

The Antarctica Peninsula, and its nearby islands, are considered to have the warmest living conditions in Antarctica, and it is undersea exposure all year round. The bay is suitable for navigation and cargo delivery for Antarctica bases, and it is dominated by pervasive ice caps, with more than 90% of the island being glaciated.

The AE-33 was installed in a shelter with electrical support and safe conditions to climate interferences, at the height of ~3 m above the ground. The equipment mode was set to be automatic sampling every 1 min with a speed pump set for a 5 L min^{-1} flow and a non-size-selective aerosol inlet. The meteorological records indicated that the air temperature showed an average of 1.4 ˚C, relative humidity mean of 91%, and wind speed of 6.4 m s^{-1}.

The sampling was done from February 7th and 21st, 2015. Because of some electrical problems, we had a few issues amid the sampling procedure, extraordinarily on February 13th, 2015; consequently, this day was excluded from the data results.

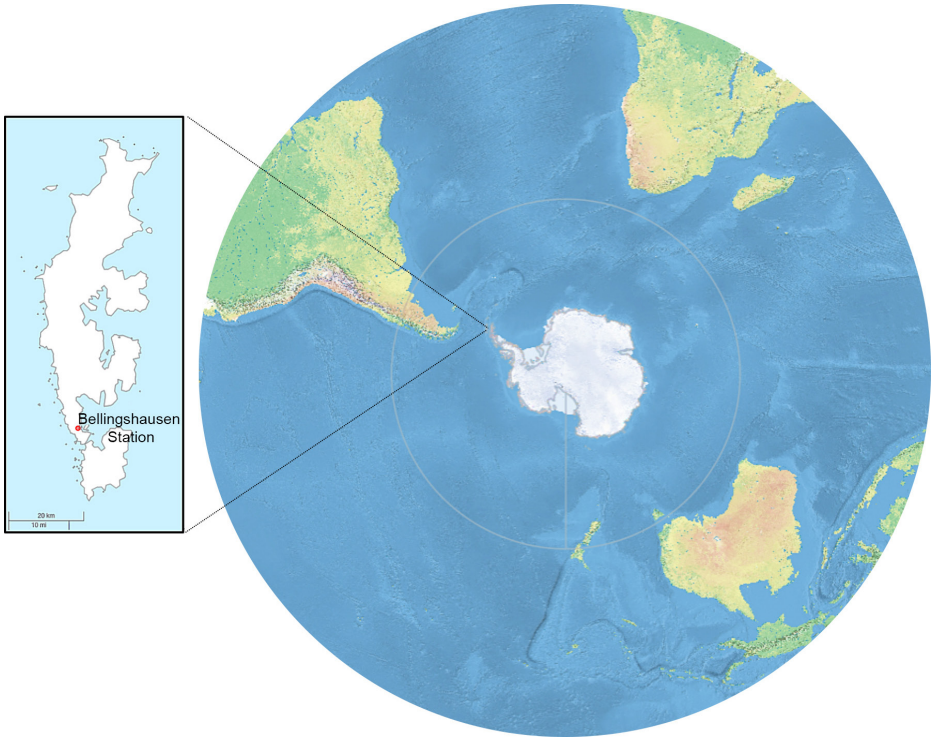

Figure 1. Sampling point located at Bellingshausen Station, King George Island, coastal Antarctica (SOOSmap, SCAR Antarctic Digital Database).

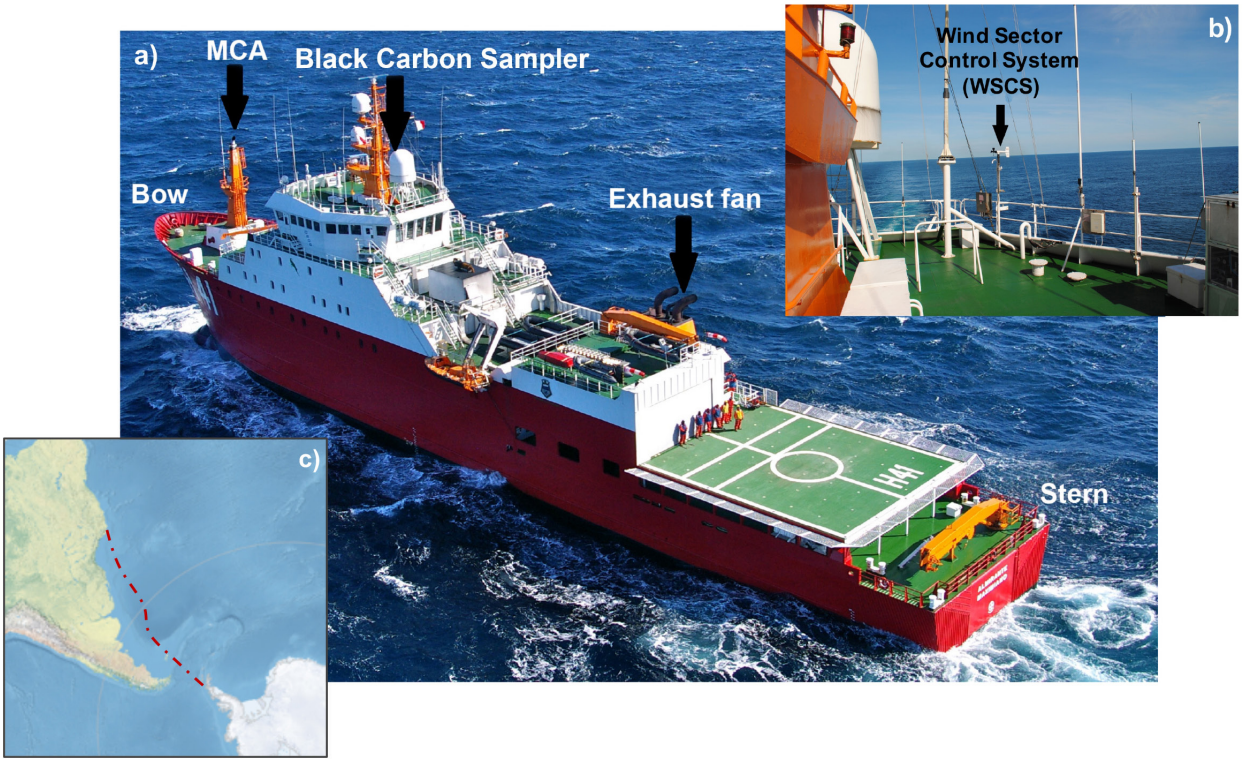

Figure 2. a) The Brazilian Oceanographic Research Ship Almirante Maximiano and its detailed parts, indicating the location of the BC sampler AE-42; **b)** sampling system WSCS installed on the ship to minimize the influence of the ship's exhaust; and, **c)** Ship's route between the Admiralty Bay (Antarctic Peninsula) and the Guanabara Bay in Rio de Janeiro/Brazil, at March 11th to April 17th, 2013.

BC measurements by a stretch from South America to the Antarctic Continent with AE-42

The portable AE-42 was used in a stretch of the Southern Atlantic Ocean, with the support of the Oceanographic Research Ship Almirante Maximiano of the Brazilian Navy, which was carried out in 2013 from March 11th to April 17th, during the OPERANTAR XXXI/2013. The BC concentration was calculated punctually throughout the ship's route between the Admiralty Bay, Antarctic Peninsula, and the Guanabara Bay in Rio de Janeiro/Brazil.

The equipment was installed on the brick of the ship (Figure 2a), with the air inlet disposed approximately 15 meters above the mean sea level. The equipment mode was set to automatic sampling every 2 min, with a speed pump set the flow for a 5 L min^{-1} and with a non-size-selective aerosol inlet. A sampling system named Wind Sector Control System (WSCS) (Evangelista et al. 2007), which allows aerosol collection only when the wind direction is contrary to sampling, was used (Figure 2b). The direct influence of the ship's exhaust chimney on the collection of aerosol samples was avoided.

The wind direction is favorable, and the bow is sterned, so the ship's exhaust releases smoke in the opposite direction of the sampler. For sampling, every time the ship's auxiliary combustion engine (MCA) was turned on, the sampler was turned off since the MCA exhaust fan is unfavorable to sampling. The sampling carried out continuously during the ship's journey was interrupted twice during the docking period in Buenos Aires (April 5th to 10th, 2013) and Rio Grande (April 11th to 14th, 2013).

To processing the AE-42 data, the primary analysis was carried out related to the elimination of events of contamination from the ship's emissions. To eliminate this effect, we considered the database resulting from the use of the WSCS. All remaining observed BC peaks of very short duration were also eliminated if higher than 100 ng m^{-3}, since they cannot be explained by any BC dispersion model of continental-scale (Bisiaux et al. 2012, Bond et al. 2013). These spurious events were also checked with respect to the BC spectral data to ensure the fossil fuel origin of the peak.

Data processing and BC discrimination

The Aethalometer estimates the BC mass concentrations (ng m^{-3}) from the change of light transmission through a filter by the optical attenuation (*ATN*) technique:

$$ATN = -100 \times ln\left(\frac{I}{I_0}\right) \quad (1)$$

Where I_0 = reference signal of light intensity of the incoming light, and I = point signal of light intensity.

The increase in light attenuation is proportional to the increase in the presence of BC in the filter; however, this relationship between ATN and BC concentration is nonlinear (Wang et al. 2011, Weingartner et al. 2003). The aerosol attenuation coefficient (b_{atn}) of sampled particles can be calculated using the change in light attenuation as a function of time, the volumetric flow rate, and filter spot (Sandradewi et al. 2008a). The absorption coefficient (b_{abs}) can be calculated as:

$$b_{abs} = \frac{b_{atn}}{C} \quad (2)$$

C is the multiple scattering parameter given by Weingartner et al. (2003), whereas b_{atn} is the correction of b_{abs} to solve the "shadowing" effect of the particle, which occurs as the filter gets more highly loaded. Thus, the BC concentration:

$$BC = \frac{b_{abs}}{\alpha_{air}} \quad (3)$$

Where σ_{air} is the mass absorption cross-section and is given by the equipment fabricant for each wavelength of the aethalometer.-

The elemental carbon is specifically the aerosol component optically quantified by 880 nm (BC_{880nm}) and the organic carbon, with a fraction that can preferentially absorb in the UV region, is quantified by 370 nm (BrC_{370nm}) (Olson et al. 2015). Due to the measurement procedure, BC absorbs uniformly across the spectrum resulting in a tracer for fuel particulate matter and biomass since the optical absorption increases strongly at shorter wavelengths. Once the optical absorption coefficient being a sum of biomass burning and fossil fuel burning fractions, the combination of a level of BC absorption spectrum and a fittest BrC absorption curve can be isolated numerically from the 7 measurement points (Sandradewi et al. 2008a).

The AE-33 monitor, with built-in analysis via a two-component model, is feasible to distinguish BC from fossil fuel and biomass combustion, giving automatically the percentage of biomass burning (*BB%*), calculated by (Sandradewi et al. 2008a) model.

The exponents which describe the spectral wavelength-dependent extinction (or optical depth) of light by aerosols are called Ångström exponents (α) (Ångström, 1929), and the power law of wavelength dependence describes it:

$$\lambda^{-\alpha} \propto b_{abs} \quad (4)$$

Where λ is the wavelength, built by the exponential curve (Sandradewi et al. 2008b), the exponent α has been shown to increase when the particles have a significant contribution from biomass combustion.

The equations to obtain the Ångström exponents, referring to the contribution of fossil fuels (a_{cf}) and to the burning of biomass (a_{bb}), are:

$$\frac{b_{abs}(470nm)_{cf}}{b_{abs}(950nm)_{cf}} = \left(\frac{470}{950}\right)^{-a_{cf}} \quad (5)$$

$$\frac{b_{abs}(470nm)_{bb}}{b_{abs}(950nm)_{bb}} = \left(\frac{470}{950}\right)^{-a_{bb}} \quad (6)$$

$$b_{abs}(l) = b_{abs}(l)_{cf} + b_{abs}(l)_{bb} \quad (7)$$

Where the values of λ were used as referenced by (Sandradewi et al. 2008b). The difference in absorption coefficient wavelength dependency is assumed as absorption due their spectral dependencies, once to fossil fuel is λ^{-1} and biomass burning is λ^{-2} (Kirchstetter et al. 2004).

HYSPLIT backward trajectories

The acquisition of backward trajectories images from HYSPLIT (Hybrid Single-Part Lagrangian Integrated/ NOAA ARL HYSPLIT) was made to understand and gain some observations about the origins of the air masses. The HYSPLIT free available model (http://www.arl.noaa.gov) is a complete system for computing complex dispersion trajectories and deposition simulations (Draxler & Rolph 2010). HYSPLIT is designed to support a wide range of simulations related to long-range transport, dispersion, and pollutant deposition. The retro-trajectories inform the path traveled by the air masses before reaching the sampling point in a specific geographic coordinate.

Climatological data from the GDAS (Global Data Assimilation System) with 1° of the spatial resolution were used. The Global Data Assimilation System (GDAS) is the system used by the National Center for Environmental Prediction (NCEP) Global Forecast System (GFS) model to place observations into a gridded model space for the purpose of starting weather forecasts with observed data. The GDAS adds the following types of observations to a gridded, 3-D model space: surface observations, balloon data, wind profiler data, aircraft reports, buoy observations, radar observations, and satellite observations.

The trajectories were analyzed from March 16th to 27th, 2013, starting from 10 meters above ground level. The model was set up to start a new trajectory every 4 hours, and the maximum duration for each trajectory was 3 days. The daily trajectories maps, corresponding to February 6th to 11th, 13^{th,} and 17th, 2015, were used the same configuration, except for a new trajectory every 1 hour.

Fire spots

Fire spot data used were from National Institute for Space Research (INPE) free access database (https://queimadas.dgi.inpe.br/queimadas/bdqueimadas), operating by Aqua satellite (MODIS sensor). INPE currently uses this satellite routine for the fires' time series of South America. The algorithm, named Collection6, is the same applied by the National Aeronautics and Space Administration (NASA) (Giglio et al. 2016). The detection algorithm uses native 4, 11, and 12 μm brightness temperatures derived from the corresponding 1 km MODIS channels. The fire spots were obtained for March 16th to April 17th, 2013, and for February 6th,11th, 13th, and 17th, 2015.

RESULTS AND DISCUSSION

Figure 3 shows the daily average from total BC concentration data's sampling at Bellingshausen Station, which indicates a variability between the higher concentration 366.10 ± 49.10 ng

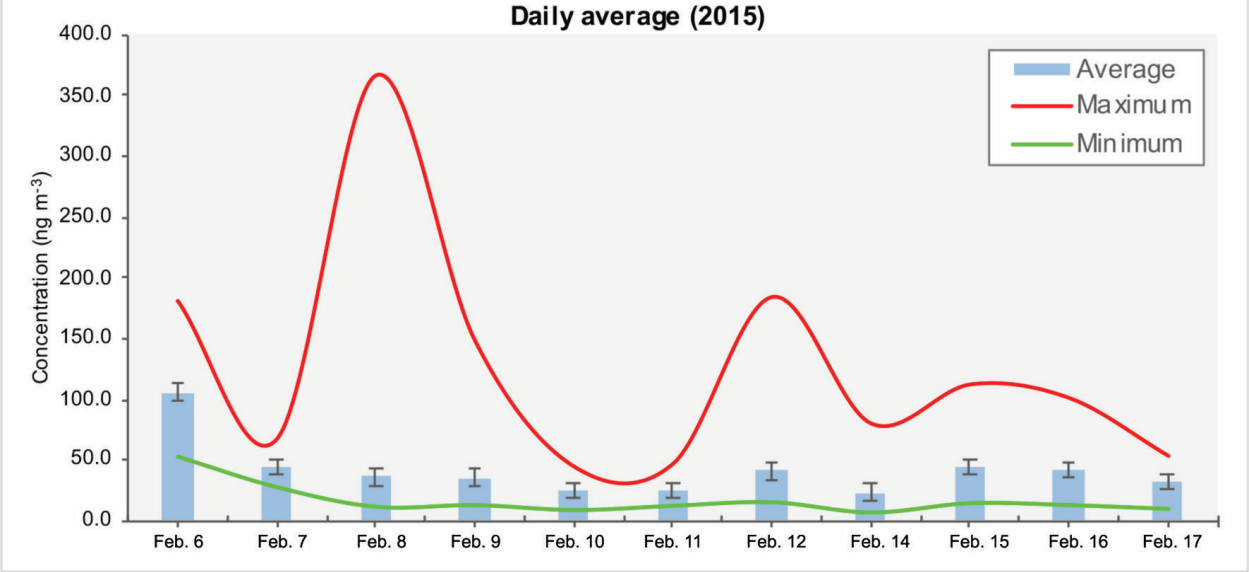

Figure 3. Average of BC daily series concentration, grouping by 30-min measurement data (bars), the maximum and minimum values, at Bellingshausen Station in 2015.

m^{-3}(Feb. 8th) and the minimum 8.40 ± 17.50 ng m^{-3} (Feb. 14th), demonstrating a strong influence of local sources.

Weller et al. (2013) found lower values, in a range of 2.1 – 1.6 ng m^{-3} to BC concentrations at Neumayer station, associated with local impact and large meridional transport. Additionally, some estimates indicate a lower presence of BC concentrations at the South Pole, ~0.65 ng m^{-3} by Bodhaine (1995) for annual mean, growing at coastal sites recording annual mean around 1-2 ng m^{-3} at Halley (Wolff & Cachier 1998), increasing at the Antarctica Peninsula, appeared ~8.3 ng m^{-3} on Brazilian base (Pereira et al. 2006) for annual measurements, and significantly higher at Hansen et al. (2001) samples, on McMurdo Station, appear as a lower range concentration centered at ~20 ng m^{-3} and high mode at ~300 ng m^{-3}. The summer data at Indian stations by Chaubey et al. (2010), Maitri and Larsemann Hills, resulted at ~75 ± 33 ng m^{-3} and 13 ± 4 ng m^{-3}, respectively, attributed to activities over Antarctica. Table I summarizes relative and previous BC concentrations reported in the Antarctic region.

Our data show a high mean compared to previous studies, around 41.80 ± 22.80 ng m^{-3}, indicating the significant human impact at the sampling area and highlighting a BC concentration gradient from the South Pole to the Southern Ocean, as mention by Weller et al. (2013). These BC values can be attributed to the equipment that supports scientific facilities, like energy generators, and also by a presence of ships and flights arriving in the coastal area, considering growing Antarctica tourism and scientific activities during austral summer over the years (Graf et al. 2010, Hall & Saarinen 2010, Pereira et al. 2006). The contributions of local BC concentration are significant, in this case, because there is an increase quickly, often following an irregular pattern, reaching concentration values of up to two orders of magnitude higher than the total average, indicating BC emission of local origin. In contrast, the BC concentrations due to air masses coming from long distances increase significantly over time and decline almost in the same pattern (Pereira et al. 2006).

In our findings, the relationship between biomass burning fraction (*BC_bb*) and fossil fuel

Table I. The previous BC concentrations reported in the Antarctic region.

BC concentrations (ng m^{-3})	Location	Instrumentation	Year of measurements	Reference
0.65 (annual mean)	Amundsen-Scott Station (South Pole)	Aethalometer	1987 - 1990	Bodhaine (1995)
0.3 - 2 (monthly mean)	Halley Station	Aethalometer	1992 - 1195	Wolff & Cachier (1998)
8.3 (annual mean)	Comandante Ferraz Base	Aethalometer AE-09	1993, 1997 and 1998	Pereira et al. (2006)
20 (low range of concentrations) to 300 (high-concentration mode)	McMurdo Station	Aethalometer	1995 - 1996	Hansen et al. (2001)
2.1 – 1.6 (annual mean)	Neumayer station	Aethalometer AE-10	1999 to 2009	Weller et al. (2013)
		Multi-Angle Absorption Photometer (MAAP)	2006 to 2011	
75 (average value)	Maitri	Aethalometer AE-41	2009	Chaubey et al. (2010)
13 (average value)	Larsemann Hills			
41.80 (mean value)	King George Island	Aethalometer AE-33	2015	This issue

fraction (*BC_Fossil Fuel*) contribution on total BC emission, calculated by Sandradewi et al. (2008b) model, suggests a considerable fossil fuel contribution(~80% of total) performance at the sampling period (Figure 4). The biomass related BC appeared on February 6th, 8th, and 9th, 2015 in considerable percent, compared to other days of the sampling, corresponding to around 20% of the total BC.

Recent studies have further defined the main sources of local chemical contamination in Antarctica as derived from abandoned waste disposal sites, chemicals disposed of through the sewage system, emissions of combustion processes, and the release of other persistent contaminants (Tin et al. 2009). Within these compounds and sources of contamination are cited fuel spills, heavy metals, polychlorinated biphenyl (PCB's) contamination, and polycyclic aromatic hydrocarbons (PAH's). Pereira et al. (2006) reported that the most important local source of BC at Comandante Ferraz Base (Almirantado bay, King George Island) comes from the diesel engines in the electrical power plant at the Brazilian station, also linked to the burning process of organic waste by the local station at the region.

The α often characterizes the spectral dependence of light absorption, which means that aerosols emitted from different sources absorb light with different characteristics of spectral dependence. The relevant values of α of BC (from fossil fuel) is around 1, and BrC (from biomass burning) varies from 1.5 to 7 (Hoffer et al. 2006, Utry et al. 2014). Therefore, it is typical for atmospheric particles to appear coated/mixture with different constituents, converting α values dependent on wavelength due to chemical properties changes of the carbonaceous aerosol (Utry et al. 2014). However, the α is an important tool for understanding the behavior of aerosol absorption and helping to interpret the contributions of BC sources better.

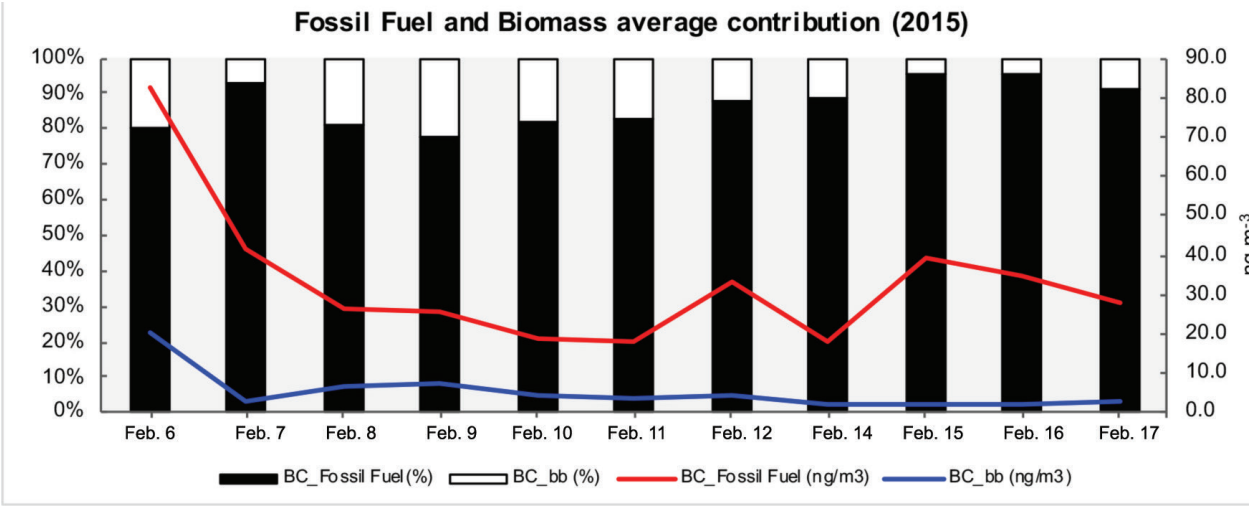

Figure 4. The fossil fuel (BC_Fossiel Fuel) and biomass (BC_bb) fractions contribution to BC in terms of percentage (left side) and concentration (right side).

The literature indicates a range of values of α from different measurements and sources, sampled at various sites. Bergstrom et al. (2007) found that α for aerosols emitted from traffic road or diesel trucks were about 0.8-1.1, while some authors indicated sources linked to biomass aerosols with a range of 0.9-3.5 (Chen & Bond 2010). The general results show similar values of ~1.0-1.1 linked to diesel soot or traffic studies and biomass/wood burning are referred around 2 (Kirchstetter et al. 2004, Sandradewi et al. 2008b, Schnaiter et al. 2003). Here, the results indicate the range α = 0.9-2.0 and the total average around ~1.5 for period sampling (Figure 5). The α suggests that the region is strongly biased towards fossil fuel contribution. Still, there is a biomass burning contribution that is influencing the air BC content of the region as well.

Figure 6 shows the trajectories and the fire spots obtained for specific periods from along with the surrounding continents. It is possible to observe that the air masses pass through South America and arrive at the sample point; that is, there was a favorable transport from BC to Antarctica, especially between February 6th and 11th, 2015. In the following days, the trajectories shift slightly towards the south, and the contribution of BC biomass burning decreases. This coincides with the BC contribution linked to the biomass burning source, indicated by the fire spots.

The BC concentration calculated for the AE-42 showed an average concentration of 10.90 ng m^{-3} in the Antarctic latitudes. Figure 7a illustrates the total BC profile of concentration through the latitudes (blue line) for sampling in 2013. To analyze the temporal evolution of BC concentration values over the years, a comparison was made with series from 2002 to 2004 by Evangelista et al. (2007), as shown in Figure 7a.

The profile of the data obtained in 2013 follows the same trend of the series of BC measured in 2002 (Oct. 29th to Nov. 16th), 2003 (Oct. 29th to Nov. 16th), and 2004 (Feb. 14th to Mar. 6th) from Evangelista et al. (2007) observations (Figure 7a). According to these authors, in general, the emissions of continental BC present a minimum value in the summer due to the elevation of the boundary layer and the predominant rainfall regime in December, January, and February. The maximum concentrations of BC present a bimodal pattern on the southeast coast of the

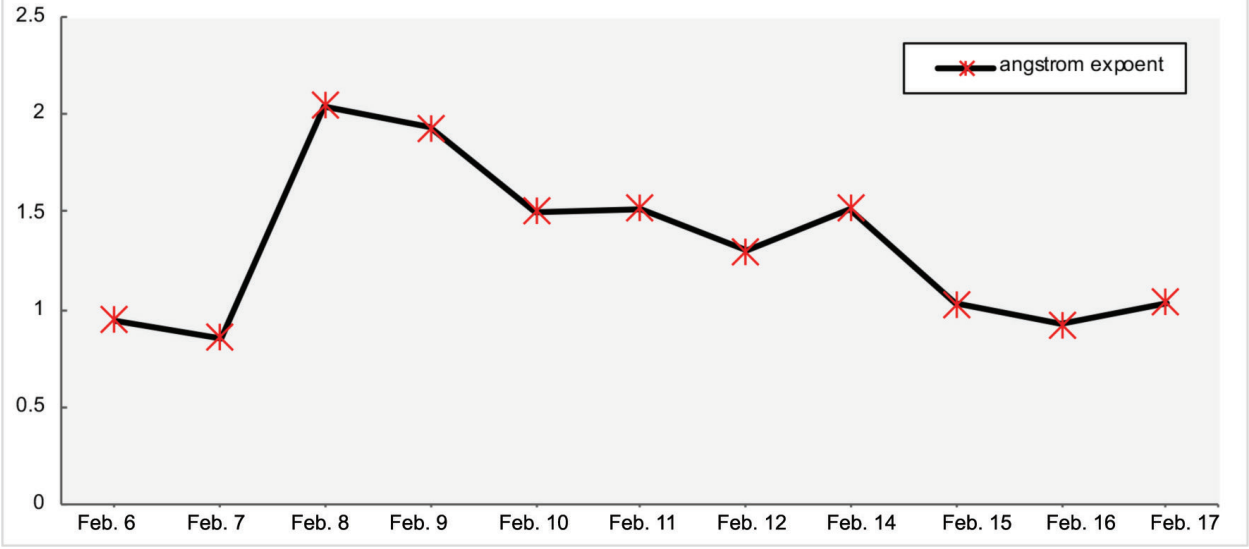

Figure 5. The average daily (2015) in terms of Ångström exponent (α)

Atlantic due to the two different sources. The first predominant pattern of maximum occurs from July to August. The BC emitted by urban and industrial sources tends to be concentrated at superficial levels due to reducing the boundary layer in the stable troposphere (Castanho & Artaxo 2001). This process represents the most significant value of the increase in BC in the urban areas of Brazil, Uruguay, and Argentina and on the east coast of the Atlantic, in the range of latitude 22ºS to 35°S. The second pattern of maximum concentrations of BC occurs from August to November. It is characterized by dry periods in the central regions of South America, when the most biomass burning events arise.

As aerosols are transported according to wind currents, an important transfer event of BC arising from biomass burning is the South Atlantic Convergence Zone (ZCAS), which is related to surface circulation in South America (Evangelista et al. 2007). Figure 7b,c indicates a profile of fire spots by the latitudes, demonstrating the same pattern as the outlines in Figure 7a, emphasizing these events as contributors to BC mass in Antarctica.

The AE-42 results of the BrC_{370nm} and BC_{880nm} were evaluated to designate the contribution of BC from burning biomass and burning fossil fuel, respectively. In areas of significant urban conglomerates, mainly in remote regions with low anthropogenic influences, the contribution of BC arising from the burning of fossil fuel is more significant than the contribution of burning biomass. Figure 7 shows that the concentrations of BrC_{370nm} remained lower than those of BC_{880nm} for most of the ship course; however, at latitude 62°S, some concentrations of BrC_{370nm} were much higher than the concentrations of BC_{880nm} (the area that is highlighted with a circle in Figure 8). The difference in the concentration of BrC_{370nm} and BC_{880nm} in some points of latitude 62°S, as well the range of latitude 22°S to 30°S, is indicative of the presence of BrC of biomass burning origin.

On March 18th, it was registered more than 10% of concentration data for BrC_{370nm} much higher than that of BC_{880nm}, not only in average values but also in its measured point-by-point concentration (Figure 8). In addition to the local contribution, these types of events clearly show

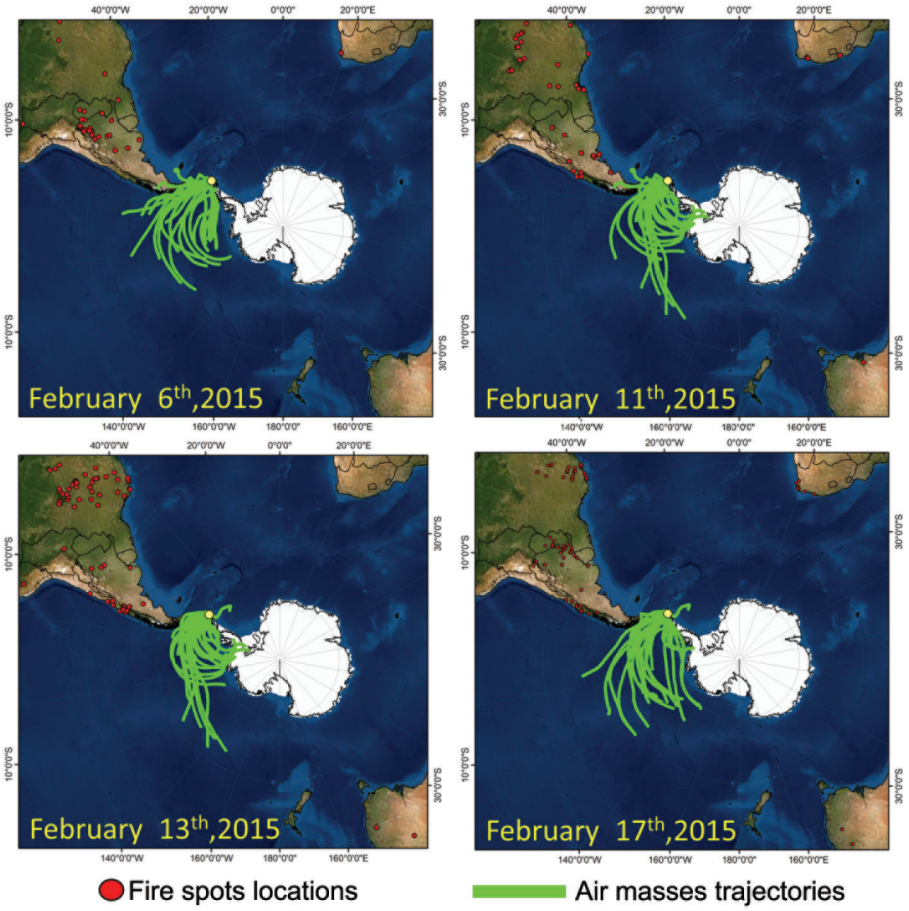

Figure 6. HYPSLIT trajectories for February 06th, 11th, 13th and 17th,2015, show the fire spots influences obtained for specific periods from INPE's free access database (https://queimadas.dgi.inpe.br/queimadas/bdqueimadas).

the occurrence of biogenic material arriving in Antarctica from distant origins.

Recently, Srivastava et al. (2021) showed observations on the concentration variations of fossil fuel and biomass burning in BC in the Indian Southern Ocean during the austral summer, revealed higher values comparing to results here, whereas the BC mass concentration variation from 150 ng m^{-3} to 450 ng m^{-3} between the latitudes (23ºS to 60ºS). The authors attributed fossil fuel as a major source of BC and indicated that the air masses transporting aerosols come from the southern part of African and eastern Madagascar regions by the cyclonic wind. The higher BC concentration (250-350 ng m^{-3}) observed in the latitude range of 57-60ºS were linked to the convergence of north-westerly and south-easterly winds. Wolff & Cachier (1998) proved the annual cycle of BC concentrations in Antarctica and partially attributed these concentrations to biomass burning in the Southern Hemisphere through a Lagrangian transport model. Pereira et al. (2006) related the presence of specific BC particles measured in the Antarctic continent with biomass burning in South America using a wind field model for the sampling period. Fiebig et al. (2009) confirmed aerosol transport from biomass burning in tropical latitudes (central region of Brazil) to Antarctica. This relationship was established by investigating a transport case that occurred during the 2007 Antarctic winter using a Lagrangian model of transport, and the observations coincided with the peak of the burning season in Brazil.

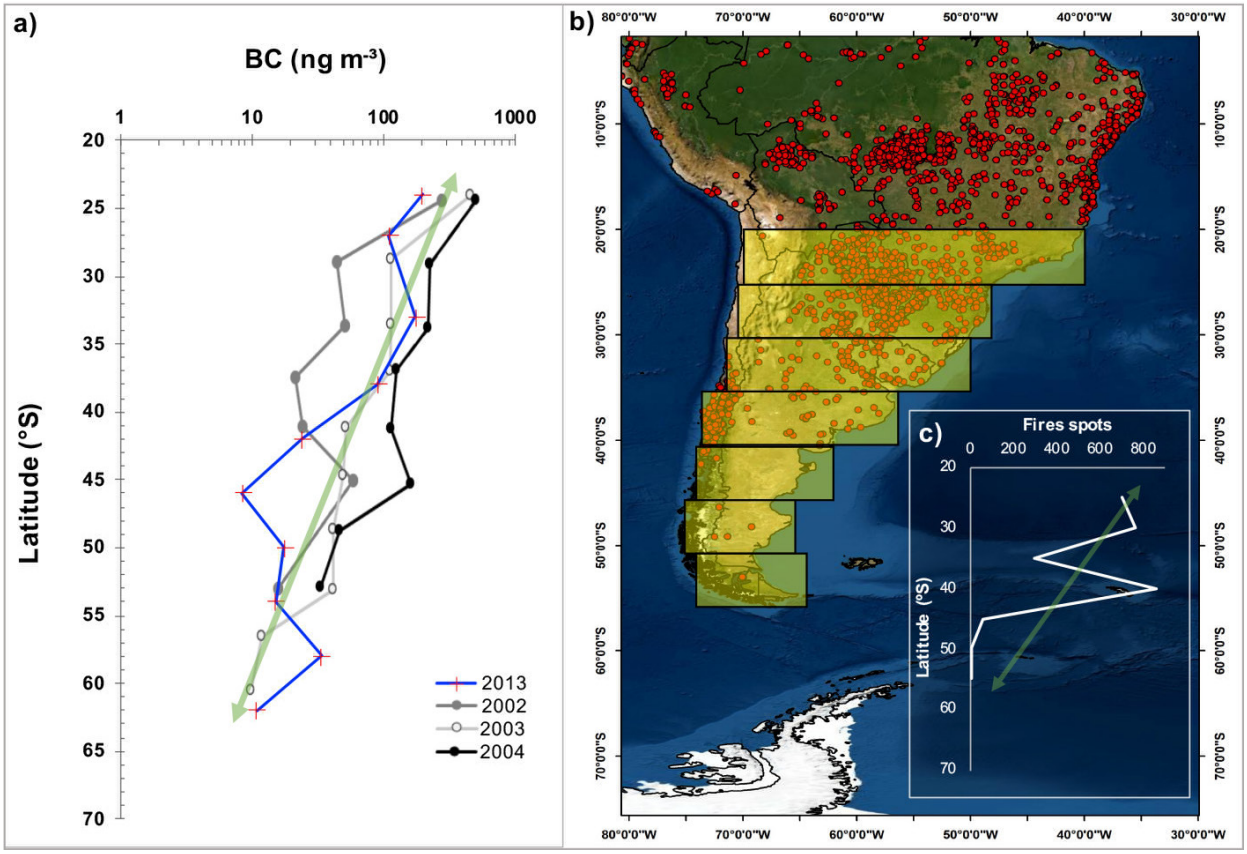

Figure 7. a)Total BC concentration measured from March 16th to April 17th, 2013 between latitudes 22°S and 62°S, performed by AE-42 aboard the Oceanographic Research Ship Almirante Maximiano (blue line), comparing a historical series of total BC from 2002 to 2004 by Evangelista et al. (2007); b) spots fires from the South American continent, at March 13th to April 17th, 2015. The bands in yellow mark the fires by latitude, wherein c) the profile of spots fires along the latitudes is illustrated.

To evaluate the contributions of the air masses over the ship's route the trajectories were generated during sampling (Mar. 16th to 27th, 2013) (Figure 9). The observations suggest that the aerosols transported to the Antarctic region come from South America, western Southern, Pacific Ocean, and the Antarctic continent. Figure 9 indicates the fires spots produced by the surrounding continents (South America, Australia, and Africa). It's possible to note some trajectories came from the western sector of the Pacific Ocean, which indicates the chance of transporting particulate material from Australia, where the burning season occurs at the beginning of the year.

The air masses arise closer from low latitudes in the western sector of the Southern Ocean and southern South America, as the region of Punta Arenas, Ushuaia, and the Chilean Canals in Chile, representing a significant transport of particulate material to the Antarctica region. There is evidence of air coming from the sub-Antarctic area and the middle area of Antarctica, in which there has been an increase in wind currents from the west and cyclone activities. These finds support the transport of dust into the atmosphere around Antarctica (Cataldo et al. 2013).

The air masses shown in Figure 9 come from the western sector of Chile and are influenced by areas to the east of South America, indicating

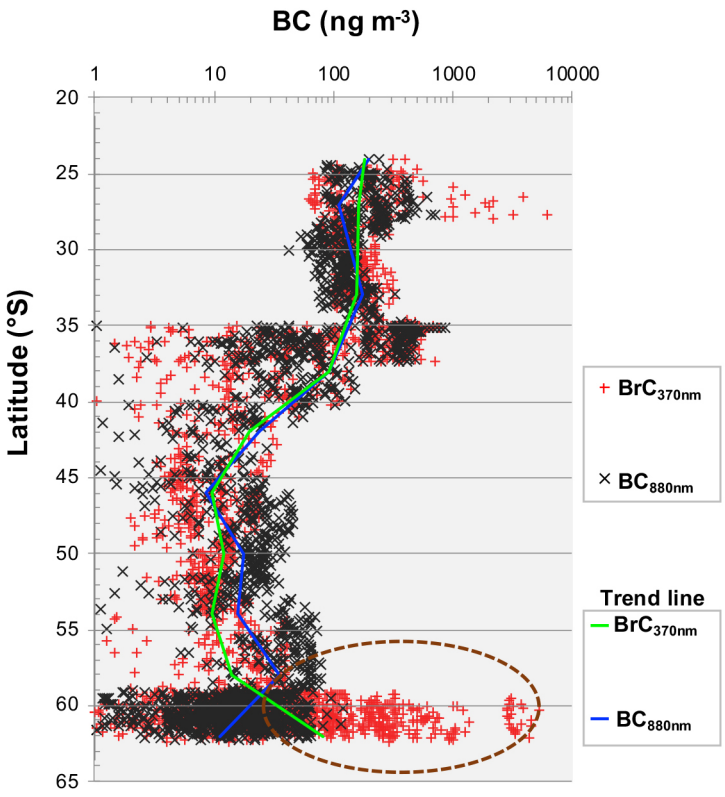

Figure 8. The BrC_{370nm} and BC_{880nm} measurements punctually for each latitude, performed with the AE-42 in ng m^{-3}. The highlighted area shows the difference between BrC_{370nm} and the average of BC_{880nm}, around 62ºS.

the transport of particulate matter from Patagonia, as previously indicated by Gassó et al. (2010). Due to the limitations of mathematical models and the contamination itself generated by several external sources during sampling, it is complicated to know the exact location of the origin of the aerosol measured in Antarctica. Therefore, it is pertinent to discuss the presence of biogenic BC in the continent from a distant origin.

Indeed, the long-term distant transport is an important mechanism and must be considered in discussed and argumentation about effects of aerosols at Antarctica, but there is a lack of knowledge about the environmental implications over BC particles presence. The particles that contain a significant amount of BC exert a warming influence on the atmosphere-surface system by absorption of light, and as a consequence decreasing cooling effects on the atmosphere (Polyakov et al. 2020a). BC particles released in the troposphere are alleged to be responsible for the earth's global warming processes (Novakov 1984).

Dust particles affect the radiation to varying degrees, depending on the minerals composition that makes it up and coated with BC, enhancing the optical effects (Jacobson 2012). Consequently, the effect of light absorption aerosols changes the vertical temperature profile of the atmosphere (Conant et al. 2002, Gao et al. 2008, Jacobson 1998), thus influencing convection and mixing in the planetary boundary layer (Barbaro et al. 2013).

To summarize, when suspended in the atmosphere, BC particles warmed the air and, when deposited on snow and ice, speeding up the melting of glaciers, directly impacting the climate of the regions where it is deposited (Cereceda-Balic et al. 2020, Kang et al. 2020). Ultimately, aerosols could influence temperatures mainly

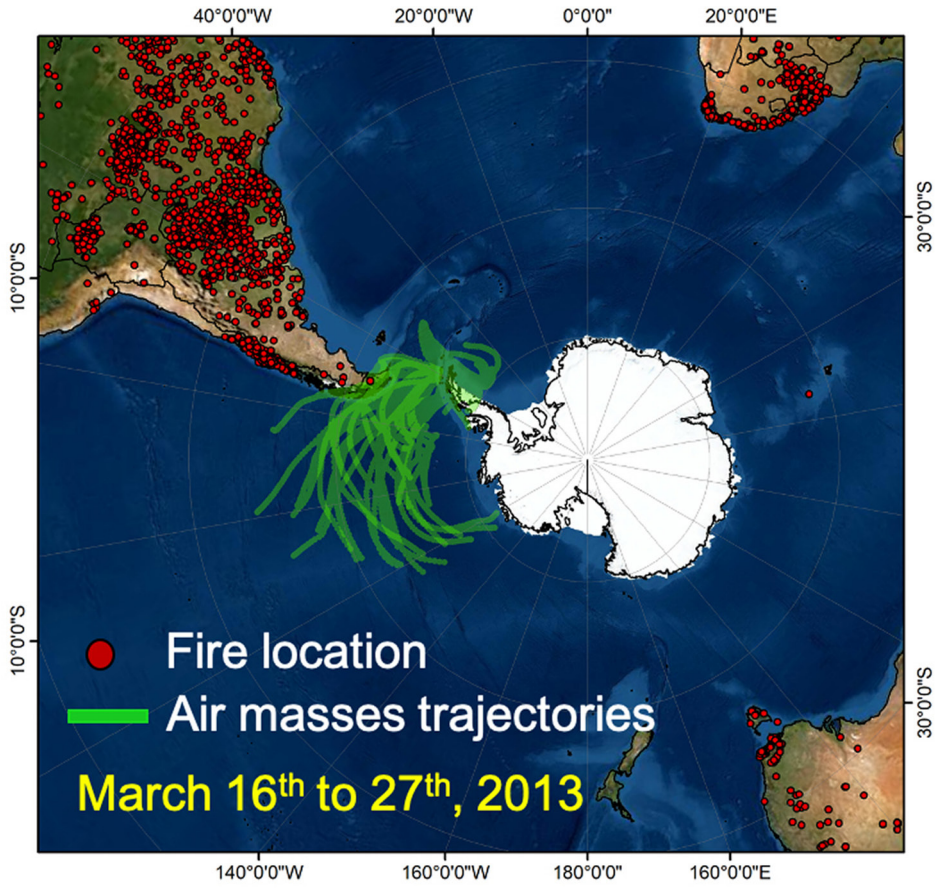

Figure 9. The trajectories of air masses during the Oceanographic Research Ship 's route (March 16th to 27th, 2013), showing the fire spots influences obtained for specific periods from INPE's free access database (https://queimadas.dgi.inpe.br/queimadas/bdqueimadas).

by energy transfer between the ground and the atmosphere (Jacobson 1998).

The scientific community has been debating intensely about the recent environmental observations in Antarctica, especially related to changes in temperature. There is considerable concern about the largest areas of Antarctica, mainly at the coast, that has lost mass through acceleration coastal glaciers melting and ice calving (Helsen et al. 2008). At the Artic, some reports indicate that BC may significantly contribute to the rapid warming involved by sea ice loss (Koch & Hansen 2005). Paolo et al. (2015) show that the ice shelves have significantly lost their thickness by rapid warming over Antarctica. Moreover, Quayle (2002) indicates that the Antarctica peninsula has experienced some of the most rapid air temperature increases on Earth (2ºC over the past 40 to 50 years). Some records indicate a significant rise in the atmosphere's temperature since the mid-twentieth century, unprecedented in the Antarctica continent (Turner et al. 2016).

Subsequently, the effects of BC on the atmosphere occur significantly at local and regional scales, once BC's lifetime being the controlling parameter of its radiative forcing (Bauer et al. 2013). Additionally, to observations and studies about BC transported in the atmosphere to Antarctica and deposited to the polar ice sheet, the local impacts must be taken into account in interpreting the carbonaceous particle, especially BC data, for global studies in the Antarctica region.

CONCLUSIONS

In the context of climate change, improving our understanding of polar pollution is crucial. The identification of BC by Aethalometer continuous measurements has proven to be a good tool for source characterization in a remote environment. In this study, we observed:

- The values obtained for BC concentration on King George Island in 2015 were 41.80 ± 22.80 ng m^{-3}(mean) and 36.70 ng m^{-3} (median), which were high compared to most other studies.
- The source segregation for the samples showed that, on average, 80% of the BC data come from fossil fuel combustion, and 20% is related to biomass burning.
- The average value found across latitudes for the measurement onboard the Oceanographic Polar Ship in 2013 was 10 ng m^{-3}, proving to be similar to the results of previous studies.
- At latitude 62ºS, biomass burning was an interesting influence, which indicated a contribution of more than 10% of the values obtained at this latitude.

It is plausible to assume that the most significant contribution of BC to the study, in general, is from fossil fuel combustion since in the summer for the Southern Hemisphere, there are slight burning spots from the surrounding continents. A thorough understanding of fire events and an accurate prediction of air masses and continual measurements for the determination of BC in the Antarctica atmosphere are deemed essential, especially in the period of the dry season in the regions of South America, which appears the most biomass burning events arise (around August to November).

The high levels of BC concentration in sampled region can be explained by human interventions, linked to the expansion of anthropogenic actives in King George Island, corresponding to the increase in circulation of airplanes, ships, and activities as tourism, logistic operations and maintenance in all station.

The snow and ice environments are very sensitive to variations on local temperature due to the human implications, responding quickly to these changes, and climate feedbacks in the polar regions warrant urgent attention, highlighting the essential observations of the Southern Ocean and Antarctica continent fluxes exchanges. Therefore, it is reasonable to assume an interconnection between the number of carbonaceous aerosols and the temperature variation of the local atmosphere, given Antarctica Peninsula and adjacent islands, considering these are suffered from local anthropogenic influences and by distant sources.

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