

Sources and Transport of Urban and Biomass Burning Aerosol Black Carbon at the South–West Atlantic Coast

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Abstract The total extent of the atmospheric impacts associated to the aerosol black carbon (BC) emissions from South America is not completely described. This work presents results of BC monitored during three scientific expeditions (2002, 2003 and 2004) on board of a Brazilian oceanographic vessel *Ary Rongel* that covered the South–West Atlantic coast between 22–62°S. This latitudinal band encloses major urban regions of South America and the outflow region of the SACZ (South Atlantic Convergent Zone), which is an important mechanism of advective transport of heat, moisture, minor gases and aerosols from the South America continental land to the Southern Atlantic Ocean. Our results showed that aerosol BC enhanced concentrations from urban/industrial origin can be transported to the South–West Atlantic Ocean due to the migration of sub-polar fronts that frequently reach tropical/subtropical regions. Despite the decrease of aerosol BC concentrations southwards (from $\sim 1,200 \text{ ng m}^{-3}$ at latitude 22°S to $\sim 10 \text{ ng m}^{-3}$ at latitude 62°S), several observed peak events were attributed to regional urban activities. Most of such events could be explained by the use of air mass back trajectories analysis. In addition, a global model simulation is presented (Goddard Institute for Space Studies – GISS GCM BC simulation) to explore the origins of aerosol BC in the South–West Atlantic. The model allowed isolating the biomass emissions from South America and Africa and industrial (non-biomass) pollution from other regions of the globe. This model suggests that the apportionment of about half of the aerosol BC at the South–West Atlantic may derive from South American biomass burning.

Key words black carbon · South–West Atlantic · Antarctic Peninsula · back trajectories · GCM

1 Introduction

The Intergovernmental Panel for Climate Change (IPCC 2001) estimates that aerosol black carbon (BC) contributes with $+0.8 \pm 0.4 \text{ W m}^{-2}$ to the global atmospheric temperature increases, which is comparable to the methane contribution. Due to the shorter lifetime of aerosol BC in the atmosphere, while methane is globally distributed, it is more associated to climate effects at the regional scale than at the global scale (Wang 2004). This is consistent with the findings of Menon et al. (2002), based on a GISS climate computer model and aerosol data from 46 ground stations in China. They reported that intensified flooding and droughts over the northern and southern regions of China during the summer season could be linked to significant amounts of aerosol BC inputs into the low troposphere from high polluted regions of Asia. According to that study, the aerosol BC heats the air, changes the regional atmospheric stability and vertical motions, resulting in alterations of the hydrologic cycle with significant regional climate effects. Their model also indicates that, besides the regional impacts, teleconnection processes can be expected resulting in changes of rainfall and surface temperature patterns in other parts of the world.

For the South American continent, especially Brazil, both source-terms of aerosol BC (fossil fuel and biomass burning) are expected to have significantly raised during the last decades, as a result of growing urbanization and increased deforestation of Amazon, “Cerrado” and the Coastal Atlantic forest. In general, the inter-annual behavior of the continental atmospheric BC emissions present a minimum during the summer season (December, January and February) due to the boundary layer elevation and a predominant rainy regime, mainly at the (sub)tropical domain. The maximum concentrations in the southeastern coastal areas show a bi-modal pattern due to two different sources. The first one predominates during July and August, the austral winter season, when aerosol BC emitted from urban/industrial sites tends to concentrate

at surface levels due to the boundary layer lowering in a stable troposphere (Castanho and Artaxo 2001). This represents the major process of aerosol BC atmospheric increase in the coastal West Atlantic atmosphere that encloses the largest urban regions of Brazil, Uruguay and Argentina, in the latitudinal band of 22–35°S. From August on, the aerosol BC atmospheric concentrations start a steady reduction at urban domains (Castanho and Artaxo 2001). The second source-term is triggered during the austral late winter and in the spring season (August–November) being characterized by drier weather at Central South America, when most of the biomass burning events occur (Kaufman et al. 1998). Some of the large scale biomass burning events may export aerosol BC to the West-Atlantic coast and the effectiveness of that process depends on the intensity and positioning of the SACZ (South Atlantic Convergent Zone), Freitas et al. 1996. The SACZ is an anti-cyclonic air current flow regime that drives air from the central, northern and southeastern regions of Brazil, to the south, at the West-Atlantic coast. Its occurrence is related to the South Atlantic Subtropical High (SASH) circulation. The center of the SASH tends to move seasonally to the east over the Atlantic Ocean in the summer and to the west toward the continent in the winter, when the SASH ridge increases from the center to the border, with easterly winds on the eastern, central, and northern regions (Nobre et al. 1998). According to (Nobre et al. 1998) by the end of August, convection starts to migrate in the Amazon basin toward its western position, at the border of Brazil and Peru, and keeps its southward migration to the end of September. By that time, moderate convection is also observed over southeastern Brazil. At the end of September the two systems merge and a well defined band of convection oriented NW–SE is established.

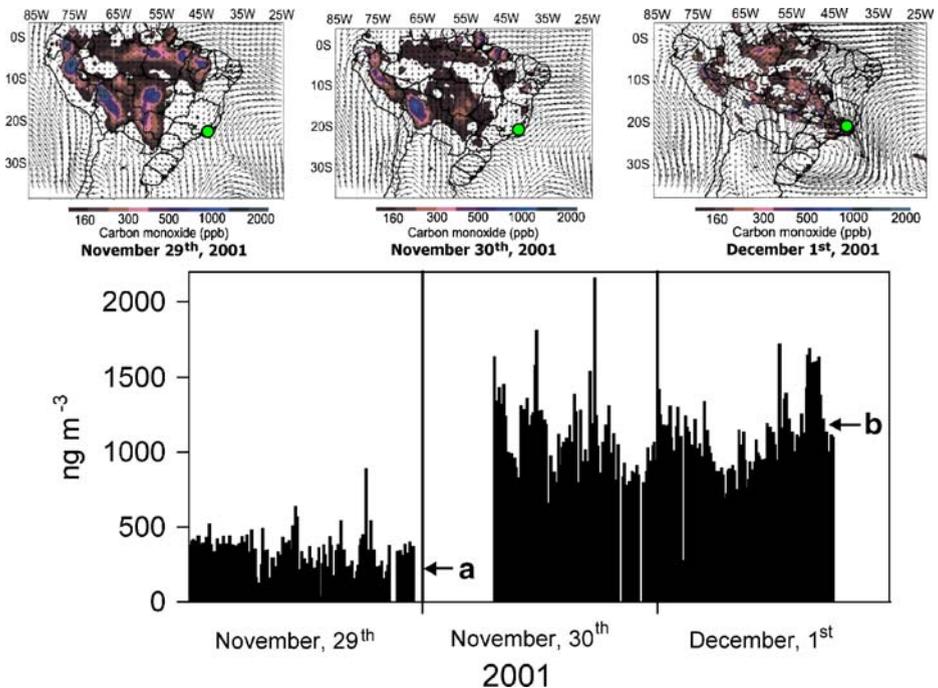


Fig. 1 (upper side) Biomass burning plume dynamics inferred from carbon monoxide by a dispersion model (MASTER-IAG/University of São Paulo); (bottom side) Aerosol BC time series during the corresponding days (A: 315 ng m⁻³ and B: 1050 ng m⁻³ average levels). Green circle indicates the location of the monitoring ground station at Ilha Grande Island/Brazil

Figure 1 depicts an event related to the above mechanism through a daily-modeled biomass burning plume displacement inferred from CO concentrations for the period November 29th–December 1st, 2001. In this example, aerosol BC was being continuously monitored at Ilha Grande Island, located at the Atlantic coast ($22^{\circ} 11'S$; $44^{\circ} 12'W$). Aerosol BC time series exhibited an increase in the average from 315 ng m^{-3} to $1,050 \text{ ng m}^{-3}$, with a peak of $2,200 \text{ ng m}^{-3}$, during the transit of the plume over the sampling site. In that episode, the predominance of the marine influence (during November 29th) was changed to the continental influence (during November 29th–December 1st). Both aerosol BC and CO are enhanced during biomass burning events and their concentrations are usually tightly correlated. Within a season interval Chen et al. (2001) found correlated levels of $r \sim 0.7\text{--}0.9$ for these two tracers.

In the present study we investigate the aerosol BC dispersion from most important South America urban sites to the South–West Atlantic coast during three oceanographic cruises, as part of the Brazilian Antarctic Program, during the summers of 2002, 2003 and 2004. We have combined measured aerosol BC to air mass back trajectories analysis for the better understanding of the dispersion mechanisms of its continental sources. In addition, we have investigated the importance of South American biomass burning aerosol BC (to the South–West Atlantic coast and Antarctic Peninsula) relative to other aerosol BC continental sources by employing a global model simulation, the Goddard Institute for Space Studies (GISS) GCM BC simulation.

2 Study Area

Major uncertainties in the aerosol BC emissions at large urban regions of the globe are the spatial non-homogeneity of emissions and the amplitude of changes in atmospheric concentrations from diurnal to inter-annual time bases. Regarding South America, where the majority of urban areas are located at the South–East Atlantic coast, careful measurements on board of ships off shore may provide data on the average apportionment of aerosol BC from the coastal urban areas, despite their non-homogeneous sources. To minimize the

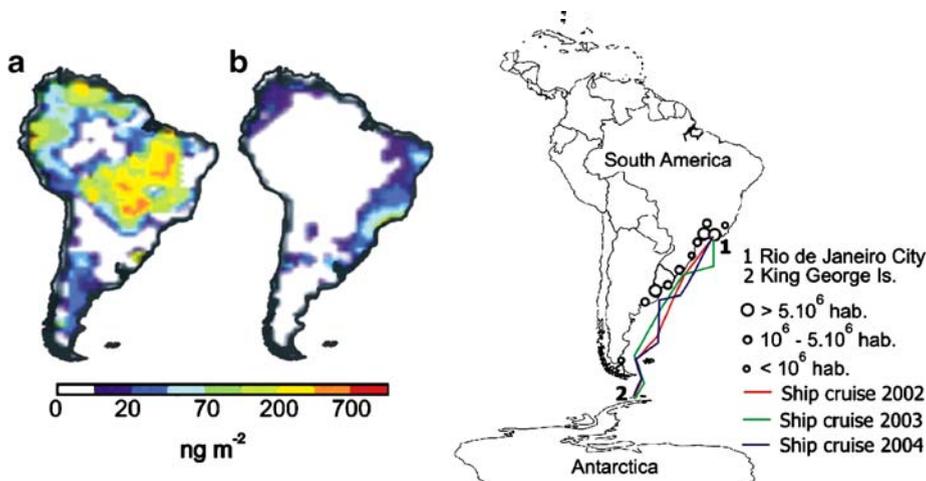


Fig. 2 (left side) NASA estimated BC emissions from South America, **a** of biomass burning and **b** of urban activities; (right side) Ship cruise along the Southern Atlantic Ocean and main metropolitan centers

influence of particular meteorological conditions during a ship cruise mission, we have conducted aerosol BC measurements during three field campaigns. The cruises were from Rio de Janeiro (22° 54'S; 43° 10'W) to the Brazilian Antarctic Station Comandante Ferraz (62° 05'S; 058° 24'W) at King George Island/Antarctic Peninsula. Their duration was 18 days for 2002 (October 29th–November 16th), 18 days for 2003 (October 29th–November 16th), and 22 days for 2004 (February 14th–March 6th). Figure 2 (right part) depicts the ship cruise trajectories between Rio de Janeiro city and King George Island, indicating the positioning of the most important urban sites at the coastal region; its left part shows NASA estimated aerosol BC emission for South America, where (a) is related to the biomass burning component and (b) to urban/industrial sources. Taking into account that our database was obtained for the period November–February, during the 3 years of observation, we have considered that a relatively predominance of the urban component would exist, compared to the biomass burning component that predominates in the months of August–November.

3 Material and Methods

Aerosol BC was monitored by an aethalometer (Magee Scientific, model Ae-10) (Magee Scientifics, Hansen and Schnell 1991) installed aboard the Brazilian Navy oceanographic research vessel Ary Rongel. Specific attenuation cross-section for the aerosol black carbon deposit is $19 \text{ m}^2/\text{g}$, considering the quartz micro-fiber filter used (as recommended by

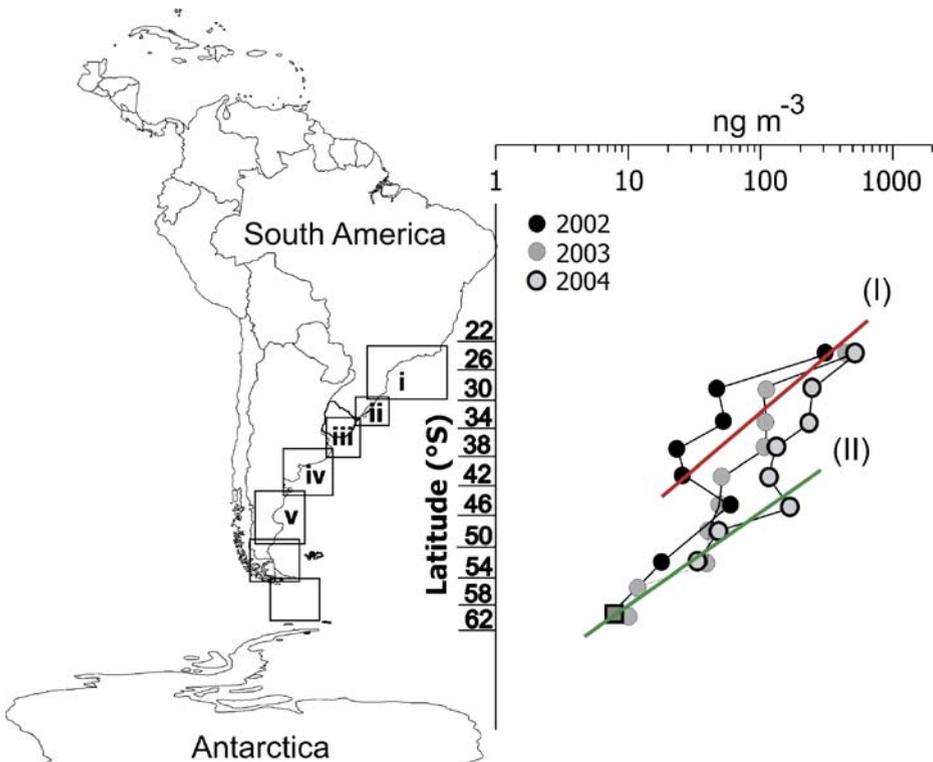


Fig. 3 South–West Atlantic aerosol BC average profiles and fitted trends. (■) Average value (8.7 ng m^{-3}) obtained at King George Island/Antarctic Peninsula (Pereira et al., 2006)

Magee). An inlet was employed to prevent marine sea salt spray admission in the filter holders, due to storms and the ship movements. It was designed to provide a cut off size of $10\ \mu\text{m}$. It was positioned at the top of the ship ($\sim 10\ \text{m}$ above sea level). Acquisition of data was maintained at a constant airflow rate of 15 slpm and the equipment was programmed for continuous 10 min-acquisition periods. Back trajectories were obtained from the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) Model, in the most pronounced peaks of aerosol BC observed at the time series (model source: <http://www.arl.noaa.gov>). The aerosol BC monitoring system was connected to an electric-mechanical device coupled to a wind anemometer in the ship to control the sampling and prevent contamination by the ship stack's emissions. Sampling was interrupted any time the plume entered a sector of 45° having the intake at its origin, and maintaining about one additional minute when the contaminated sector was cleared.

4 Results and Discussions

The average profiles obtained from the on-board aerosol BC monitoring for years 2002, 2003 and 2004 depicted an almost constant decreasing trend from the tropical to sub-polar

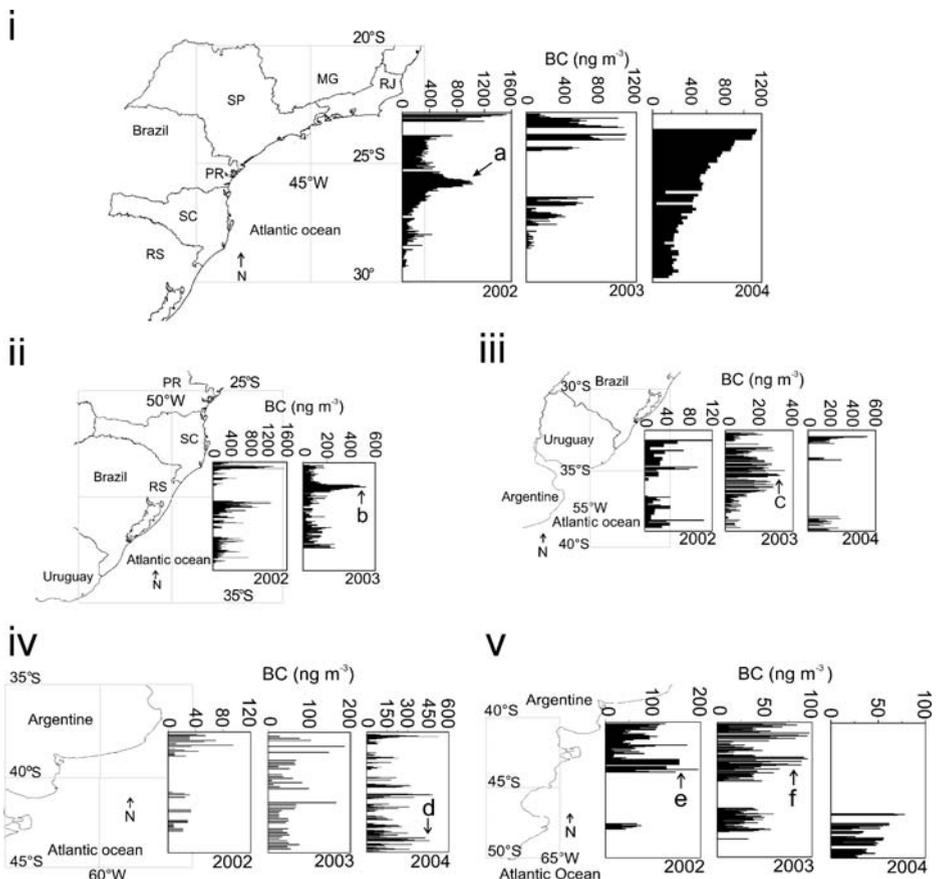


Fig. 4 Aerosol BC concentrations at different latitudinal bands. Arrows identify selected peaks (a, b, ..., f)

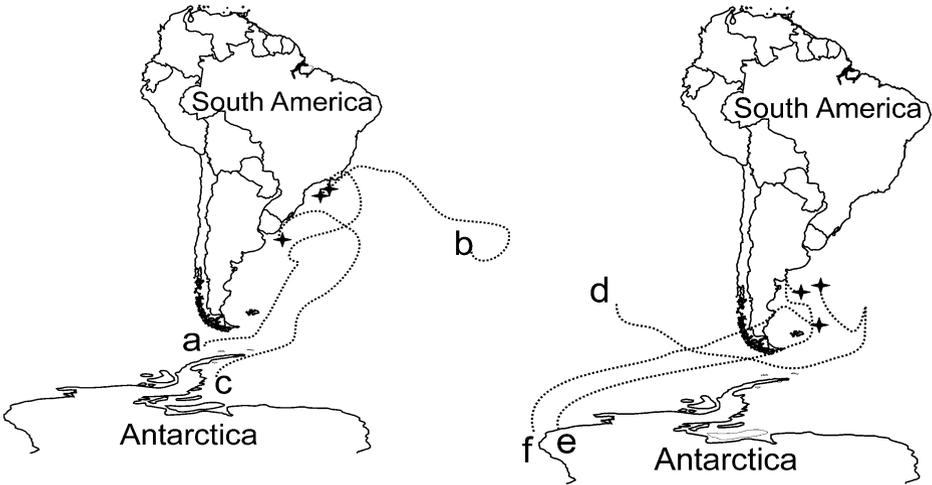


Fig. 5 Air mass back-trajectories corresponding to aerosol BC peaks at Fig. 4

latitudes, Fig. 3. The basic feature of the decrease of atmospheric concentrations with respect to latitude could be described by logarithm fittings as presented by Eqs. 1, 2 and 3:

$$\log C_{bc,2002}(ng\ m^{-3}) = 14.06 - 2.76 \log x, \quad r^2 = 0.56 \quad (1)$$

$$\log C_{bc,2003}(ng\ m^{-3}) = 15.93 - 3.19 \log x, \quad r^2 = 0.90 \quad (2)$$

$$\log C_{bc,2004}(ng\ m^{-3}) = 15.59 - 2.95 \log x, \quad r^2 = 0.83 \quad (3)$$

where “x” refers to the latitude. Kruskal–Wallis non-parametric analysis indicated that a significant statistical difference exists among the results of the three cruises ($p < 0.05$). Aerosol BC concentrations tended to be higher for successive cruises. The most pronounced differences occurred at the latitude band of 30–50°S. Maximum values were found around

Table 1 Main events (peaks) of aerosol BC observed during the three ship cruises along the South Atlantic ocean

Date	Identification of events	Latitude (S)	Longitude (W)	Time (h) ^a (GMT)
Oct 30th, 2002	a	23° 49'	044° 42'	03:40
Nov 13th, 2002	e	44° 16'	056° 13'	23:50
Nov 16th, 2002	g	54° 20'	061° 40'	08:00
Oct 31st, 2003	b	30° 56'	049° 51'	21:00
Nov 9th, 2003	c	35° 30'	053° 40'	22:30
Nov 12th, 2003	f	46° 47'	058° 25'	13:10
Nov 14th, 2003	h	54° 00'	060°40'	08:30
Nov 15th, 2003	I	60° 30'	059° 30'	08:20
Feb 22nd, 2004	d	43° 09'	058° 12'	22:00

^a Refers to the time of peak mode.

latitude 25°S what suggests a large input of aerosol BC from the “urban complex” composed by the cities of Rio de Janeiro and São Paulo, which at the resolution of Fig. 2, represent an unique source-term. A second peak (deviation from the trend) was observed just below latitude 35°S and may represent the influence of Argentine urban activities. Concentration values between 40 and 62°S decreased steadily to a minimum level as those observed in the Antarctic Peninsula since the 1990s (Penner et al. 1993; Evangelista 1998; Wolff and Cachier 1998; Pereira et al. 2006). The two fit curves at Fig. 3 represent the effects due to BC sources in Brazil (I) and with the accretion of Argentine and Uruguay sources (II). Despite this non-continuous behavior, results of the three campaigns are related to the aerosol BC long-term average observed at the Brazilian Antarctic Station Comandante Ferraz at King George Island (8.7 ng m⁻³), Pereira et al. 2006.

4.1 Case studies

Figures 4 and 6 depicts the position of the peaks presented at Table 1, with their respective references at the coast line. Legends in the figure represent the states of Brazil located at the Atlantic coast (RJ: Rio de Janeiro, SP: São Paulo, PR: Paraná, SC: Santa Catarina, RS: Rio Grande do Sul). Figures 5 and 6 shows back trajectory for the selected peaks, obtained from the HYSPLIT transport and dispersion model (<http://www.arl.noaa.gov/ready.html>). They were calculated to match the ship coordinates at the dates and approximate time of interest for the BC peaks at sea level.

The Fig. 4 presents an exponential-like decrease of concentrations by the time the ship left Rio de Janeiro city, which is characteristic of the typical behavior of a distance-to-source pollutant dispersion pattern. Event “a” represents a pulse concentration within this decrease trend. The corresponding air mass back trajectory at Fig. 5 shows the occurrence of a sub-polar air mass originated at the West Antarctic Peninsula before the event. The sub-

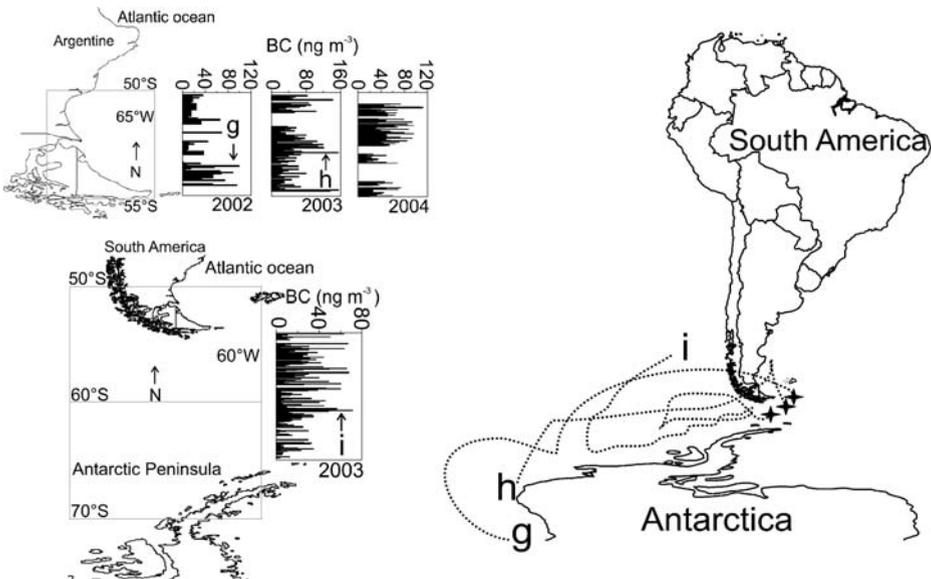


Fig. 6 (left side) Aerosol BC concentrations at the Southern Atlantic Ocean. Arrows identify selected peaks (g, h, i); (right side) Air mass back-trajectories corresponding to the peaks on the left side

polar front moved northeast over the Atlantic Ocean, up to the tropical South America coast, reaching Brazil. At the vicinity of Uruguay it started to develop an anti-cyclonic motion striking the coastal region of Southeast Brazil just at its urban area. The peak of aerosol BC was observed in the latitudes corresponding to the frontier between São Paulo and Paraná State. Event “a” can be identified as a typical example of advective transport of aerosols from the Brazilian urban sites to the West–South Atlantic. Differently from the previous one, the event “b” is influenced by an air mass derived from the middle South Atlantic Ocean, driven to the coast by the SASH circulation. The air mass developed a zonal motion in the east-to-west direction reaching the continental region of Brazil. It flowed initially through the coastline, probably induced by the topography of the Serra do Mar mountain ridge, and also through the large urban areas of Brazil, before reaching the ship coordinates. Aerosol BC at event “b” has the same source-terms of event “a”. Both events “a” and “b” were characterized by their well-defined shape and their short duration, which correspond to rapid advective transport due to strong winds associated to the above meteorological systems. Event “c” also presented similar characteristics of event “a”, this is, influenced by the advection of sub-polar frontal systems. The air mass, originated at the Antarctic Peninsula, moved northwards through the Atlantic Ocean and around the latitude 50°S it developed an anti-cyclonic motion affecting the South of Brazil and Uruguay. After then, it reached the ship coordinates. These two regions can be the source-terms of the aerosol BC peak. Although event “c” is identified in the time series at Fig. 4 (significantly high compared to the background line base), it does not have a comparable marked resolution as those observed during events “a” and “b”. Conditions of atmospheric stability during the event “c” and/or the associated less significant urban source-terms at the continental region may be the causes of the broad-like peak shape observed. Event “d” was characterized by its very short duration. Corresponding back trajectories analyses pointed out to a source-term localized at the Ocean Pacific most southwestern region, the Drake Passage, what is not reasonable. Therefore such type of event may represent the ship’s contamination. Events “e”, “f”, “g” and “h” are all characterized by only small increases in BC concentrations compared to the background base line and also by the migration of sub-Antarctic polar air through non significant urban sources at South America. Figure 2(left)a indicates that from latitude 35°S to Terra del Fuego in Chile, aerosol BC emissions are controlled more to biomass burning process than to urban/industrial activities. Although of lesser intensity (mostly associated to agriculture practices and house heating) biomass burning can be, potentially, the source-terms of aerosol BC corresponding to the above events. Event “i” (Fig. 6) is associated to an air mass which origin is related to the South America. Its anti-cyclonic motion at the Drake Passage, parallel to the Antarctic Peninsula, would probably transport the aerosol BC to the ship coordinates.

5 South America Biomass Burning Dispersion: Model Simulations

In order to investigate the importance of South American biomass burning aerosol BC relative to other BC sources, we have used a global model simulation, the Goddard Institute for Space Studies (GISS) GCM BC simulation (Koch and Hansen, 2005; Koch et al. 2006; *in press*). Koch and Hansen (2005) used this model to isolate various source regions to examine the origins of aerosol BC in the Arctic. Koch et al. (2006; *in press*) investigated the origins of aerosols in other regions, including the southern hemisphere. To do this, they performed multiple simulations isolating (industrial) sources from Asia, North America, and Europe and biomass sources from Africa and South America, relative to a full-

simulation including all emissions. Here we use these simulations to look at the dispersal of South American biomass burning aerosol BC.

The model includes mass transport of all aerosol species, although we only consider aerosol BC here. For a full model description we refer to Koch and Hansen (2005) and Koch et al. (2006; *in press*). Industrial (here includes residential sources) BC is assumed to be emitted as insoluble and becomes soluble as it ages (with e-fold age of approximately 1 day). Biomass aerosol BC is assumed to be emitted along with soluble components and is assigned a solubility of 50%.

Industrial aerosol BC emissions are from Bond et al. (2004). Biomass burning emissions are based on the Global Fire Emissions Database (GFED) model estimates for the years 1997–2001 (van der Werf et al. 2003; 2004). According to these inventories, 8.2 Tg BC is emitted annually, 3.7 Tg BC from biomass burning and the rest from industrial sources. South America emits 1.2 g (about one third of global) biomass BC and about 0.2 Tg BC from other sources. Thus, according to these estimates, about 84% of South American BC is from biomass burning.

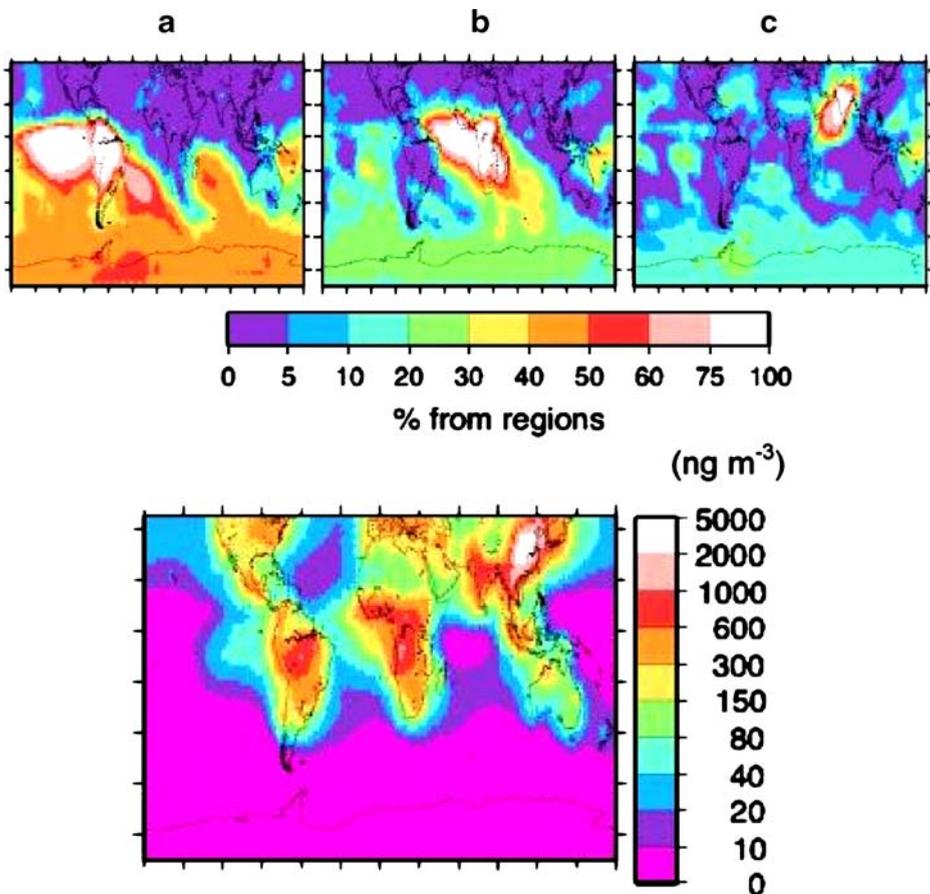
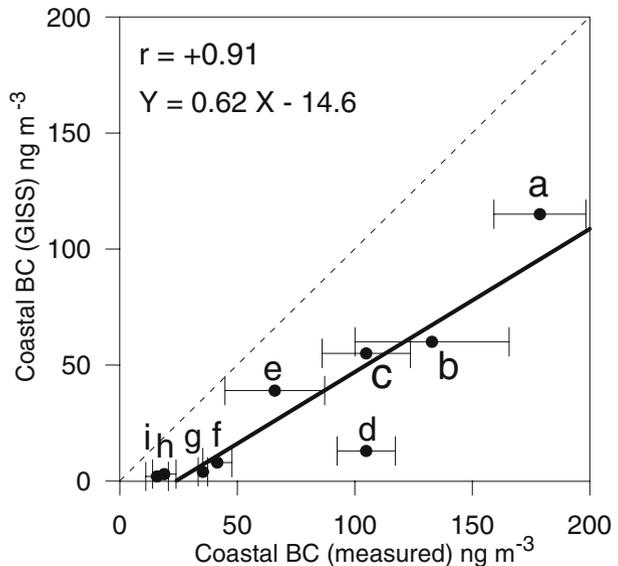


Fig. 7 (upper side) Percent contributions to aerosol BC surface concentrations coming from South American biomass burning (a), African biomass burning (b) and South Asian industry and residential sources (c); (lower side) GISS model results for aerosol BC surface concentration (ng m^{-3})

Fig. 8 Predicted aerosol BC values from GISS model and measured values along the coastal South America. Letters indicate latitudinal bands (see text)



The upper part of Fig. 7 shows the percent contribution to aerosol BC surface concentrations coming from 3 sources: (a) South American biomass burning, (b) African biomass burning and (c) Indian industrial and residential sources. In the South Atlantic about half of the aerosol BC comes from South American biomass burning, with this source dominating in the central east region. Further north, African biomass burning dominates. Further south various sources contribute, although the South American source is the largest. Thus we see that 10–30% of the aerosol BC in the southern oceans each comes from African biomass and Indian sources. Pollution in these regions is lofted high into the atmosphere, advected poleward in the general circulation and descends in the polar vortex (Koch et al. *in press*).

In Fig. 7 (bottom part) we show the annual mean aerosol BC surface concentration for the model simulation (including all sources). In the southern hemisphere most of the aerosol BC comes from biomass burning in South America and Africa, and this aerosol BC tends to be advected westward from these continents. The model experiments that isolate the biomass pollution from South America and Africa and industrial (non-biomass) pollution from other regions allow us to discern the sources of South Atlantic aerosol BC.

A comparison between the coastal Atlantic aerosol BC estimated by the GISS model and the averaged aerosol BC measured during the three ship cruises exhibited a significant discrepancy. Measured values are higher compared to modeled values. Main differences are observed within two latitudinal bands: between latitudes 26 and 42°S, a factor of 1.2 to 2.1 is present; and between latitudes 42 and 62°S a factor ranging from 5.2 to 8.8 exists. Figure 8 shows the correlation between model predicted values and measured aerosol BC at the Atlantic coast. In the figure, letters (a, b, ..., i) indicate latitudinal bands: a (26–30°S; 45–49°W); b (30–34°S; 49–52°W); c (34–38°S; 52–53°W); d (38–42°S; 53–57°W); e (42–46°S; 57–58°W); f (46–50°S; 58–60°W); g (50–54°S; 60–61°W); h (54–58°S; 61–59°W) and i (58–62°S; 59–58°W).

Biomass burning products apportionment in the Atlantic coast is dependent on the continental wind regime, which must be favorable to the atmospheric transport from the

mainland to the coast line; on the intensity and frequency of fire spots; and on the climate regime concerning the accumulated precipitation level. During favorable atmospheric transport conditions, as the example depicted in Fig. 1, the migration of biomass burning plumes towards the Atlantic coast was responsible for increases of aerosol BC to a factor of 3.3 higher than the regional background from industrial sources. Here the GISS model considers a database of emissions enclosing the period 1997–2001, which corresponded to a period of relative low fire spot incidence. In contrast to the period of atmospheric monitoring from 2002 to 2004, when fire spot frequency was ~ 50% higher (1997–2001: mean annual fire spots was 113,782 and 2002–2004: mean annual fire spots was 225,987; <http://www.dpi.inpe.br/proarco/bdqueimadas>). Although the database presented here corresponded to the initial descending period of the biomass burning season (Table 1), its effects over the South American continent may still remain. If we consider that the dominant outflow positioning of the SACZ at the Atlantic coast occurs between latitudes 15 and 45°S, the discrepancy between model and measured values at the latitudinal band of 26 and 42°S can be better explained. In addition, we have also to consider the contributions of industrial sources, which were pointed out by the HYSPLIT Model (air mass backtrajectories “a”, “b” and “c” at Fig. 5). Higher discrepancies observed for the latitudinal band 42–62°S probably can not be merely deduced from the uncertainties on inventories of biomass burning and industrial emissions. Lioussé et al. (1993) reported that specific attenuation cross-section for the aerosol black carbon (σ_{BC}) may change from 20 m²/g for Savana biomass burning to 5 m²/g for aerosol BC at remote regions of the globe. The optical attenuation technique employed to the measurement of BC using the aethalometer makes use of an algorithm in which the concentrations are inversely proportional to σ_{BC} . Therefore if the differences between model and measurements (from Patagonia to Antarctic Peninsula) were due to instrumentation, we would expect a higher discrepancy between them. Besides reduction in emissions inventories uncertainties, for the mid and high latitudes at the Southern Atlantic, improvements in model characteristics and measuring techniques are still required.

6 Conclusions

We have measured and analyzed aerosol black carbon (BC) dispersion over the South–West Atlantic coast up to the northern Antarctic Peninsula. The dispersion at the South–West Atlantic was attributed to mixed coastal industrial sources and biomass burning sources revealed by the results obtained during three scientific cruises in 2002, 2003 and 2004. In addition, the geographically aerosol BC biomass burning dispersion between South America and Antarctic Peninsula was inferred from a global model simulation, the Goddard Institute for Space Studies (GISS) GCM BC simulation.

The oceanic monitoring at the latitudinal band 22–62°S, permitted to investigate the aerosol BC plume dispersion from identified urban emission sources at South America and to observe the effect of main synoptic scale systems associated to them. In addition to the well known effect of the SACZ that transports aerosol BC (and other biomass burning byproducts) from central Brazil to the South–West Atlantic coast, we have observed that sub-polar front trajectories along the Atlantic is responsible for the dispersion of aerosol BC from the urban sources towards the South–West Atlantic atmosphere. Aerosol BC average concentrations obtained from the long-term monitoring at the north of the Antarctic Peninsula could be explained by the average trend obtained at the South Atlantic. Despite the decreases of aerosol BC concentrations southwards (from ~1,200 ng m⁻³ at latitude 22°S

to $\sim 10 \text{ ng m}^{-3}$ at latitude 62°S), peak events could be associated to air mass back trajectories analysis which described the air mass enrichment by aerosol BC as it migrates through the source regions.

The use of a global model simulation, the Goddard Institute for Space Studies (GISS) GCM BC simulation, allowed isolating the biomass pollution from South America and Africa and industrial (non-biomass) pollution from other regions of the globe. It permitted to conclude that the apportionment of about half of the aerosol BC at the South–West Atlantic may derive from South American biomass burning.

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