Contribution of biomass fires to black carbon supply in a tropical river basin assessed using a Lagrangian atmospheric transport model and MODIS burned area product

Matthew William Jones [1]
Timothy Andrew Quine [1]
Carlos Eduardo de Rezende [2]
Thorsten Dittmar [3]
Luiz Eduardo Oliveira Cruz de Aragão [1]

[1] University of Exeter
University of Exeter, Stocker Road, Exeter, Devon, UK, EX4 4PY
{mwj202, t.quine, l.aragao}@exeter.ac.uk

[2] Universidade Estadual do Norte Fluminense, Centro de Biociências e Biotecnologia, laboratório de Ciências Ambientais, Av. Alberto Lamego 2000, Campos dos Goytacazes, Rio de Janeiro, Brazil, 28.013 – 602
crezende@uenf.br

[3] Max Planck Research Group for Marine Geochemistry, University of Oldenburg
Institute for Chemistry and Biology of the Marine Environment, Carl-von-Ossietzky-Str. 9-11, 26129 Oldenburg, Germany
tdittmar@mpi-bremen.de

Abstract. Black carbon (BC) is known to be a potential sink of carbon for the global carbon cycle, particularly if long-term ocean stores are reached. Fluvial transport to the oceans can occur through the dissolution of BC in river water. Evidence from the Paraiba do Sul river basin, Brazil suggests that river DBC concentration is related to charcoal formed during the deforestation of the Brazilian Atlantic Forest. However, we highlight several key potential sources of BC to the basin that are yet to be considered. We hypothesize that external biomass fires are a source of BC to the basin on the basis that BC released from them can be transported over large distances before being deposited. This hypothesis is tested by quantifying the number of biomass fires intercepted by trajectories en route to the basin using the HYSPLIT model and a MODIS burned area dataset. We then create a Black Carbon Fallout Index (BCFI) which is rationalized by our assumption that atmospheric BC delivery to the basin is proportional to the number of interceptions of air masses en route to the basin. Our results suggest that the BC fallout from air masses reaching the basin in the dry season can explain 50% of the variance in DBC measured in the PSR channel during a subsequent collection campaign ($p<.001$). Spatial and temporal variations in the supply of BC to the basin throughout the dry season may in part be linked to the fires associated with the cultivation of sugarcane in southeast Brazil.

Keywords: Dissolved Black Carbon, Biomass fires, MODIS, HYSPLIT

Introduction

Black carbon (BC) is an extremely recalcitrant form of carbon that cannot easily recombine with oxygen to form CO_2 (Masiello 2004). BC is the product of incomplete combustion and a primary component of soot, charcoal and graphite (Andreae and Crutzen 1997) and represents a significant sink for the global carbon cycle. This form of carbon cannot easily be sequestered into other carbon pools and is likely to have a long residence time of 10^2 to 10^4 years (Crutzen and Andreae 1990, Forbes et al 2006).

Current estimates show that biomass burning produces around 50-270 Tg yr^{-1} of BC globally (Kuhlbusch 1999, Forbes et al 2006). Studies suggest that most of this carbon produced during biomass burning remains in situ (Schmidt and Noack 2000). However, this

process also generates a small global BC aerosol flux of 3 to 6 Tg yr^{-1} to the atmosphere (Penner et al 1993, Moss et al 2010, Bond et al 2004). Moreover, fossil fuel combustion may raise this global BC aerosol flux to 8 Tg yr^{-1} (Bond et al 2004). Once suspended in the atmosphere, BC becomes increasingly soluble, allowing it to be deposited by precipitation in the process of wet deposition (Weingartner et al 1997). This is the predominant form of deposition for BC (Forbes et al 2006).

The atmospheric residence time of BC can vary between 40 hours and one month (Ogren and Charlson 1983), allowing for the transportation of suspended BC over long distances. This explains the sedimentation of BC in deep-ocean cores (Masiello and Druffel 1998, Verado 1997, Suman et al 1997). On the other hand, the flux of particulate BC to coastal ocean sites is thought to be higher than that to deep-ocean sites (Suman et al 1997) and aeolian transport can explain only 30-40% of sedimentary deposition in coastal oceans (Kuhlbusch 1998). Thus, fluvial transport of particulate BC is known to be an important flux to marine environments. In recent decades it has been recognized that dissolved black carbon (DBC) may be a secondary transport mechanism through which BC reaches the oceans through rivers (Hockaday et al 2006, Mannino and Harvey 2004). Although it is highly recalcitrant, there is evidence that BC degrades in soils until it reaches a soluble state in which it can be transported by fluvial systems to the ocean (Mannino and Harvey 2004). These fluxes of BC are significant because the residence time of BC in the marine environment is thought to be an order of magnitude higher than in soils (Forbes et al 2006).

Dittmar et al (2012) have investigated the DBC concentration in the Paraiba do Sul River (PSR) channel, southeast Brazil. Their results suggested that the river transports 0.027 Tg yr^{-1} to the oceans. These authors hypothesize that the most significant source of BC transported by the PSR is charcoal derived from the deforestation of the Atlantic Forest which has taken place within the basin over centennial timescales. However, this analysis did not encompass a formal and spatially explicit evaluation of other internal and external sources of BC to the PSR basin, notably the release of BC from biomass and fossil fuel burning from agricultural and industrial sources in the vicinity of the basin. We argue that it is entirely plausible that biomass burning, particularly in the vast area of sugarcane plantations that cover 10% of the area of nearby Sao Paulo state (Lara et al 2005), could supply BC to the basin. It is known that BC can travel large distances as an aerosol (Ogren and Charlson 1983) and that the burning of sugarcane can considerably increase the concentration of atmospheric aerosols downwind (Lara et al 2005). In addition, the possibility that fossil fuel burning from internal and external population centers was not thoroughly investigated during the study.

To compliment the analysis of the PSR basin carried out by Dittmar et al (2012), our aim is to disentangle the sources of BC for the Paraiba do Sul river basin. Here we present an initial assessment of the extent to which biomass burning can explain the spatial variation in BC concentration in the river. Specifically, we analyse the relationship between the number of burned area pixels intercepted by air masses en route to each part of the basin and compare this distribution to the spatial trend in DBC in the Paraiba do Sul River. The analysis combines a Lagrangian atmospheric transport model that enables the quantification of back-trajectories of air masses reaching the river basin with MODIS burned area data for the period 1st May 2009 and 31st October 2009.

Methodology

The Paraiba do Sul River drains catchments on the eastern seaboard of Brazil in the states of Rio de Janeiro, Minas Gerais and Sao Paulo (Figure 1a). The total drainage area of its basin is 55,500 km^2 and the length of the main channel is 790 km when measured from the

convergence point of the Paratinga and Paraibuna river tributaries in the headwater zone. 98 tributaries and their catchments were identified using 90m spatial resolution Shuttle Radar Topography Mission (SRTM) data as a Digital Elevation Model (DEM) to derive the basin hydrology for the area using a hydrographic mesh with a scale of 1:2,500,000 from the Brazilian National Water agency (ANA). Hydrology tools provided by ArcGIS 10 software were used to delineate the PSR, its tributaries and the basin and catchments that they collectively drain (Figure 1b).

We have taken advantage of field data available from the study by Dittmar et al (2012), who collected DBC concentration data in April 2010 from 21 positions along 620 km stretch of the Paraiba do Sul River (Figure 1a). In addition, monthly MODIS MCD45A1 burned area product data with a resolution of 500m was obtained for the period from 1st May 2009 to 31st October 2009 and used as a record of biomass fires over the South American continent during the dry season.

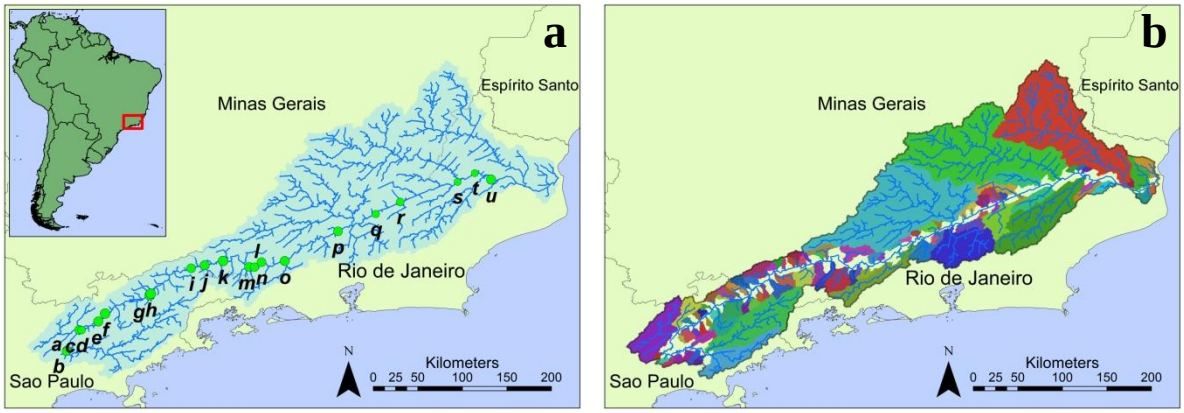

Figure 1: Maps showing **(a)** The drainage basin (shaded blue area) which covers four states and drainage channels (blue lines). The points *a* to *u* represent BC sampling positions on the main channel collected by Dittmar et al (2012) in April 2010. These data cover a distance of just under 620 km. **(b)** A map showing the delineated basin (black outline), channels (blue lines) and tributary catchments (multiple colours, purely for illustrative purposes).

We estimated the number of burned area pixels intercepted by air mass trajectories reaching the centre-points of 126 0.2° x 0.2° grid cells that were used to divide the basin. The trajectories were modeled using the Air Resources Laboratory's (ARL) Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model. The model was run using Global Data Analysis System (GDAS) data provided by the ARL. The centre-points of the grid cells were used as endpoints for back-trajectories that were modeled to ground level. Trajectories were obtained at 6-hour intervals between 1st May 2009 and 31st October 2009 and their duration was set to the period of 1 month. More than 90,000 back-trajectories were modeled to the 126 points in the basin throughout the period. The 1 month period for each trajectory run was chosen because BC aerosol can be transported for up to 1 month (Ogren and Charlson 1983), whilst the period of collection was chosen because it covers the majority of the dry season in which sugarcane and other agricultural biomass is harvested (Lara 2005). A 2 km buffer was applied to the trajectories and as a result the endpoint for each trajectory was assumed to represent a 2 km x 2 km grid cell.

Using the MODIS and trajectory data we calculated a Black Carbon Fallout index (BCFI) for each endpoint. The number of burned area pixels intercepted per month was calculated by accounting for any cell in which a burnt pixel detected by MODIS algorithm and a trajectory calculated by the HYSPLIT model occurred on the same day. The BCFI for each endpoint was defined as the number of interceptions per month reaching the endpoint, divided by the area of the endpoint (4 km^2). Its units are interceptions per square kilometer per month. The

spatial resolution of the BCFI dataset was resampled to 0.05° x 0.05° using an Inverse Distance Weighting (IDW) interpolation technique. This technique was chosen because it explicitly accounts for the first law of geography; that everything is related to everything else but that near things are more related than distant things (Miller 2004). To create each 0.05° x 0.05° grid cell, nine neighboring endpoints were considered and the weighting of each measurement decreased with the square of its distance from the cell. Thus a BCFI was computed for each 0.05° x 0.05° grid cell. The average BCFI value for each tributary catchment (Figure 1b) was subsequently calculated for each month (Figure 2). This mean BCFI ($\overline{BCFI}$) value was multiplied by the area of each catchment to estimate the total number of interceptions by trajectories entering each catchment in each month ($BCFI_C$). The sum of all $BCFI_C$ values from all catchments equates to a proxy for the total BC supply to the entire PSR basin ($BCFI_B$).

We believe that this BCFI technique provides an adequate proxy of BC supply to the basin because the emission of BC to air mass trajectories from biomass fires is likely to be proportional to the number of biomass fires encountered. Our assumption is that all BC emitted from biomass fires and intercepted by air masses reaches the basin. This assumption is unlikely to hold in the environment because fallout is likely to occur en route to the basin. Therefore our calculated BCFI can be treated as a proxy for maximum potential transfer of BC from biomass fires to the basin.

We completed a downstream analysis of the potential impact of biomass-derived BC on the concentration of BC at each tributary outlet by calculating the total BC fallout from all catchments up to the outlet ($\Sigma BCFI_C$) and normalizing by the total area of all catchments up to the outlet (ΣA_C). This normalization was conducted so as to account for varying discharge levels between catchments, which we assume to be proportional to the area of the catchment. The ratio $\Sigma BCFI_C/\Sigma A_C$ represents the concentration of biomass-derived BC fallout in the PSR basin that is supplied by the river's tributaries at each tributary outlet. We assume that all BC transferred to the basin by air masses is removed by tributaries draining these catchments and that this process occurs with equal efficiency in all catchments. As a result, estimates are treated as the maximum potential transfer of BC from biomass fires deposited in the basin to the PSR.

Results and Discussion

We first analysed the spatial and temporal distribution of black carbon on a catchment scale using estimates of the average catchment BC fallout ($\overline{BCFI}$) and the total basin BC fallout ($BCFI_B$) for each month (Figures 2 and 3). In May and June the highest $\overline{BCFI}$ occurred in catchments in the middle reaches of the channel whilst catchments in the upper reaches exhibited a higher $\overline{BCFI}$ than those in the lower reaches. The total basin BC fallout ($BCFI_B$) fell from 450,000 interceptions in May to 225,000 interceptions in June (Figure 3) and the differences in the spatial distribution suggest that most of this fall was sourced from the lower and middle reaches of the PSR, despite $\overline{BCFI}$ remaining most concentrated in this area throughout the period (Figure 2). $BCFI_B$ peaked in July at 820,000 interceptions and the spatial pattern changed markedly, with $\overline{BCFI}$ increasing dramatically in some of the lower reaches but most notably in the lower reaches. In August the $BCFI_B$ reduced once more to 335,000 interceptions and the fall in $\overline{BCFI}$ was most noticeable in catchments found in the lower reaches of the basin. The $\overline{BCFI}$ in catchments in the upper reaches remained relatively high. However, in September a dramatic shift in the spatial pattern occurred in which the $\overline{BCFI}$ increased in catchments in the lower reaches, but lowered in upper and middle reaches. The overall impact of these changes was a small rise in $BCFI_B$ to 390,000 interceptions. In

October the level of fire interceptions was extremely low in comparison to earlier periods of the dry season. The $BCFI_B$ was just 10,000 interceptions and $BCFI_C$ was zero in many catchments throughout the month. Interestingly, the temporal pattern of total BC fallout in the basin ($BCFI_B$) appears to align well with the annual pattern of sugarcane burning which occurs in the dry season between May and November (Lara et al 2005). This suggests that fires associated with sugarcane may have a particularly strong influence on BC supply in the PSR basin. In addition, the headwater and central zones of the basin exhibits consistently high BC fallout concentrations between May and August (Figure 2). The relative proximity of these areas of the basin to the major area of sugarcane plantation in Sao Paulo adds weight to the hypothesis that sugarcane plantations are a large control of BC fallout in the area.

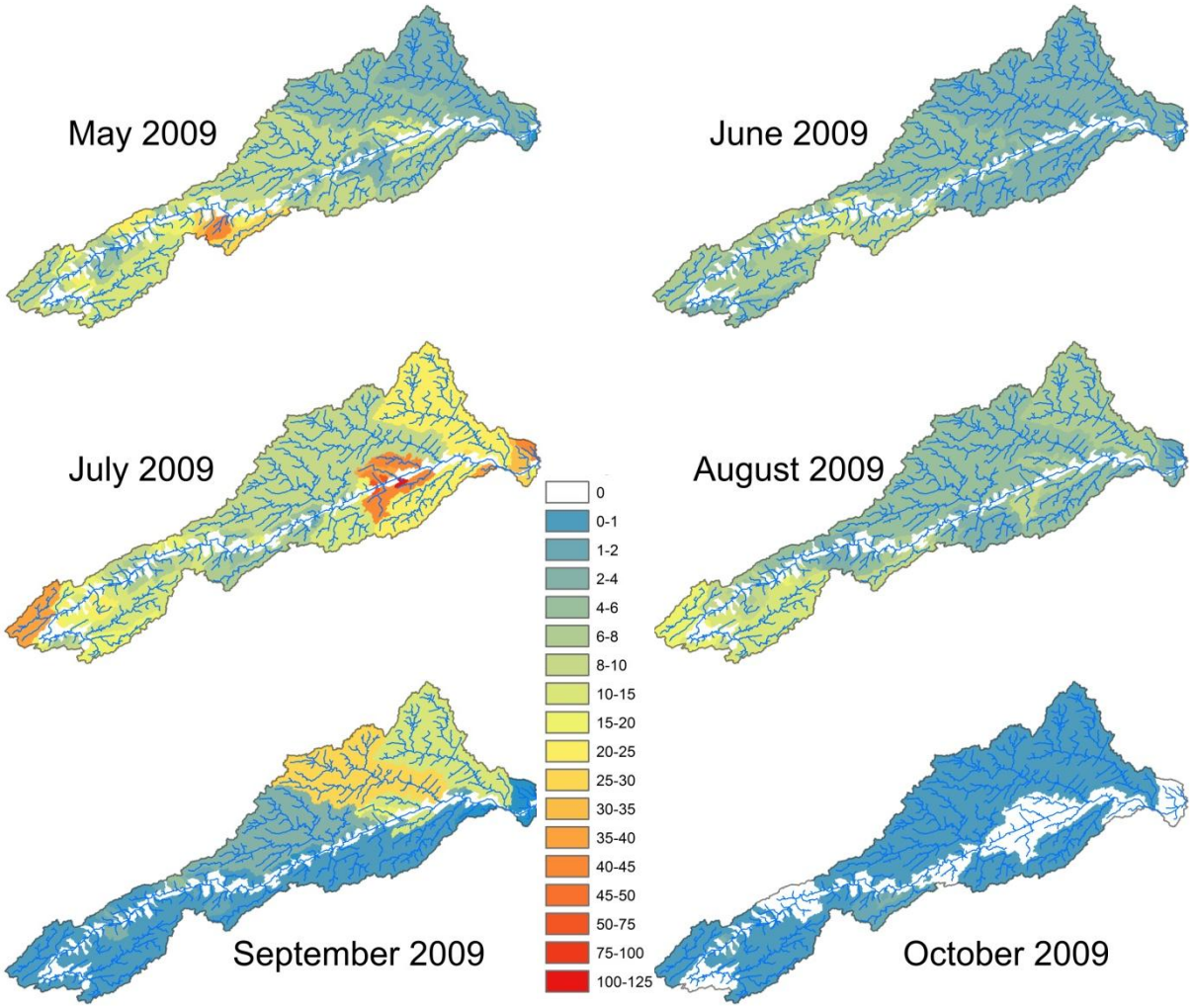

Figure 2: Average Black Carbon Fallout Index ($\overline{BCFI}$, interceptions km^{-2} $month^{-1}$) for catchments in the PSR basin.

The focus of the study was to examine whether interceptions of biomass fires by trajectories entering the basin can explain the spatial pattern of DBC found within the PSR channel. We investigate the effects of BC fallout to basins over the entire study period (May-October 2009) on the concentration of biomass-derived BC supplied to the PSR channel. The normalized catchment supply of BC from biomass burning ($\Sigma BCFI_C/\Sigma A_C$) shows a strong alignment with the pattern of downstream variations in BC concentration measured by Dittmar et al (2012) in April 2010 (Figure 4). This pattern appears to replicate large increases in BC concentration at around 100-120 km downstream and a large decrease at around 500-510 km downstream. The average $\Sigma BCFI_C/\Sigma A_C$ value between sampling points from Dittmar

et al (2012) exhibit a strong and significant relationship with the BC concentration at the downstream sampling point ($R^2 = 0.504$, $\rho < 0.001$). Thus, 50% of the downstream variance in BC concentration can be explained by downstream variations in the supply of biomass-derived BC from catchments. These results suggest that the fallout of BC derived from recent biomass fires had an influence on DBC concentrations measured in the PSR River in April 2010. However, without a thorough analysis of the other potential sources of BC for the basin it is not possible to make firm conclusions. In addition, future studies may attempt to estimate the output of black carbon from the biomass fires intercepted by trajectories and the proportion of this that is likely to be transferred to the basin. This would allow for a direct comparison to the estimations of BC output from the PSR made by Dittmar et al (2012). The efficiency of the removal of BC from each catchment and the relationship between catchment discharge and area also requires further investigation.

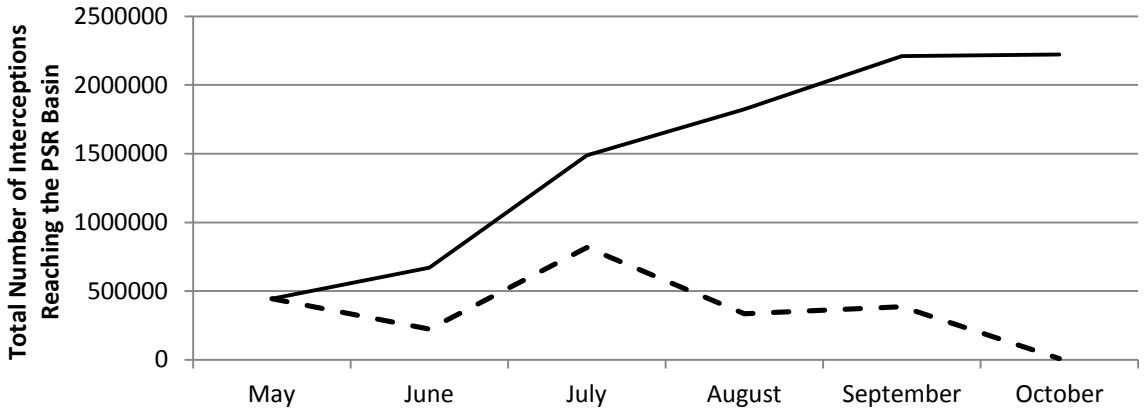

Figure 3: Total basin BC fallout ($BCFI_B$, interceptions $month^{-1}$) in each month of the study period..

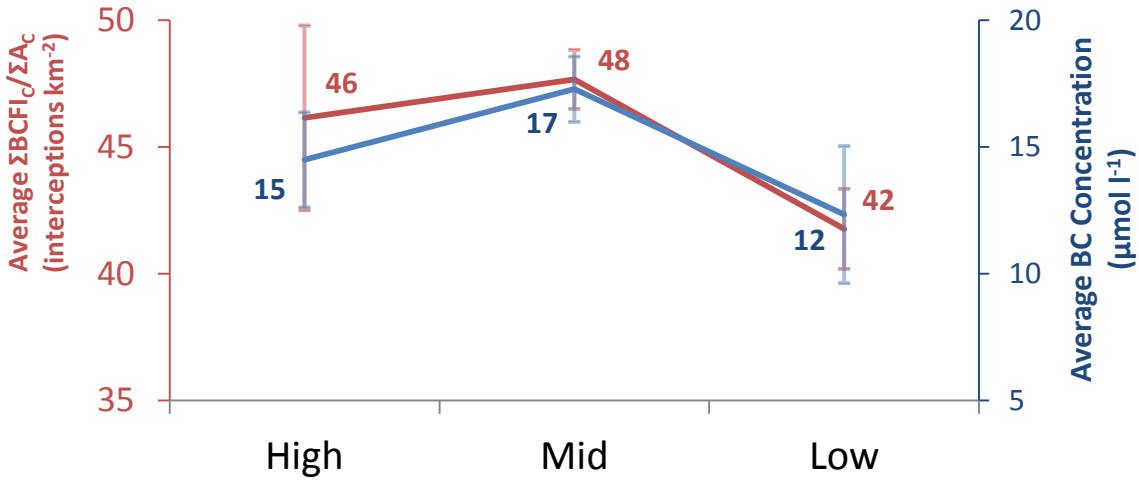

Figure 4: Average $\Sigma BCFI_C/\Sigma A_C$ for the entire study period (May-October 2009) at tributary outlets plotted for the upper (0-150 km), middle (150-450 km) and lower (>450 km) reaches of the river (red line). Also plotted in the same distance classes are average BC measurements collected in April 2010 (blue line) by Dittmar et al (2012). Error bars show one standard deviation from the mean.

Our results present evidence that recent biomass burning may be an important source of BC to the PSR basin. This study provides a new perspective on the potential sources of BC to the basin that may add to the large BC source derived from charcoal remnants of the Brazilian Atlantic forest that has previously been identified by Dittmar et al (2012). However, it is important to consider the spatial influence of this charcoal on the DBC concentrations measured in the river using deforestation data. The influence of internal and external fossil fuel sources of BC fallout should also be considered as several large cities are located in the

vicinity of the basin and it is known that fossil fuels contribute a substantial amount towards estimates of BC aerosol emissions (Bond et al 2004).

Conclusions

Our analyses of the spatial and temporal pattern of black carbon fallout in the Paraiba do Sul basin revealed that the average BC fallout index for individual catchments is consistently high in the headwater and central basins and that there was a 2009 dry season peak in fallout in July. The increasing BCFI up to July and subsequent fall is well aligned with the burn period of sugarcane, whilst the spatial concentration tends towards western areas of the basin that are closer to the sugarcane plantations of Sao Paulo. This suggests that biomass fires associated with sugarcane harvest may significantly affect BC supply to the basin. However, no formal analysis has been conducted to test this. Future studies of a similar nature may benefit from the delineation of sugarcane fires from the complete MODIS dataset.

The concentration of biomass-derived BC entering the PSR from tributary catchments appears to show a strong and significant relationship with the concentration of DBC in the river. 50% of the variance in channel DBC can be explained by downstream variations in the concentration of BC supplied to the river. This finding suggests that biomass-derived BC may be an important additional source of BC to the PSR basin. Thus, fallout of BC derived from recent biomass burning may be a more important source them previously estimated once aeolian transportation is accounted for. The assessment of the relationship between our BCFI and DBC concentration datasets from other periods would improve our confidence in these results because DBC concentration is sensitive to discharge. In addition, a more extensive field campaign that encompasses soil and aeolian sampling over a larger area of the basin would allow for analysis of the quality of our BCFI as a proxy for black carbon fallout.

This work has been a useful initial exercise in our attempts to disentangle the sources of BC to the PSR basin. Ongoing research by this group will attempt to quantify the inputs of other sources to the basin, specifically the inputs from deforestation and fossil fuel burning. In forming this complete picture of BC supply from all sources to the basin it will become possible to fully assess the relative importance of each source.

These analyses were completed using a proxy for BC fallout into the basin and the assumption that all BC intercepted by trajectories reaches the basin. An improved method may include the quantifications of the BC produced by the fires that are intercepted by trajectories that reach the basin and the fraction of this that falls out en route. Importantly, this would enable an empirical estimate of BC supply to the basin from biomass fires. This would increase the potential for quantifying the relative importance of biomass, fossil fuel and charcoal sources. In addition, the properties of the catchments that govern the transport of BC from the catchments to the main channel require further investigation. Nonetheless, we argue that the strength of the relationship observed between the proxy estimation of biomass-derived BC delivery to the channel and the DBC measurements in the channel provides compelling evidence for a significant input of BC from recent biomass burning to the PSR.

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