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Modelling the contribution of regional aerosols to fluvial fluxes of dissolved black carbon

Matthew Jones (1,2), Tim Quine (1), Carlos Rezende (3), Thorsten Dittmar (4), Ben Johnson (5), Marcus Manecki (4), Jomar Marques (3,4), Luiz Aragão (6,1)

(1) Earth System Science, College of Life and Environmental Science, University of Exeter, United Kingdom, (2) Geography, College of Science, Swansea University, United Kingdom, (3) Laboratório de Ciências Ambientais, Centro de Biociências e Biotecnologia, Universidade Estadual do Norte de Fluminense, Brazil, (4) ICBM-MPI Bridging Group for Marine Geochemistry, Institute for Chemistry and Biology of the Marine Environment, University of Oldenburg, Germany, (5) Earth System Mitigation Studies, Met Office, UK, (6) Divisão de Sensoriamento Remoto, Instituto Nacional de Pesquisas Espaciais, Brazil

Each year around 27 Tg of dissolved BC (DBC) is exported to the global oceans by rivers (Jaffé et al. 2013). Usually, this flux is thought to result from the degradation and mobilisation of BC from soil charcoal stocks in river catchments whilst the input of BC aerosol (soot) to river systems is assumed to be negligible. Global emissions inventories show that 7.6 (2-29) Tg of BC is emitted to Earth's atmosphere each year, 4.8 (1-15) Tg of which is from energy-related combustion sources and 2.8 (1-14) Tg is from open biomass burning (Bond et al. 2013). Given the magnitude of these emission estimates, it is conceivable that BC aerosol deposited to the land surface contributes significantly towards riverine fluxes of DBC. This contribution likely explains why substantial DBC fluxes are observed in freshwater systems draining glaciated and polar catchments that are free of charcoal sources (Ding et al. 2015; Khan et al. 2016).

Through a physical model of BC supply to the Paraíba do Sul River in Southeast Brazil, we show that BC aerosol is an important source of DBC even in catchments with large stocks of charcoal produced by pervasive historical deforestation. Initially, stocks of charcoal BC in the river catchment were represented using spatial datasets for deforestation combined with charcoal BC production factors. Stocks of aerosol deposits in the catchment were also simulated by combining aerosol emission grids with the HYSPLIT air mass trajectory model and a simple deposition function. Secondly, DBC concentrations were modelled for the Paraíba do Sul River according to 50,000 scenarios with varying rates of BC transfer from each stock to the river. Finally, we analysed the ability of each scenario to reproduce the spatial variability in DBC concentration observed in water samples collected over four sampling campaigns between 2010 and 2014. We found that the best-performing scenarios included a 5–18% contribution from aerosol stocks. This range is in good agreement with the previous observation that DBC loads in rivers draining glaciated Alaskan catchments are approximately 10% of those in neighbouring boreal catchments (Ding et al. 2015). At the global scale, the range would equate to an annual riverine export of 1.4-4.9 Tg of aerosol-derived BC.