Supplementary Material:

Long-term Brown Carbon and Smoke Tracer Observations in Bogotá, Colombia: Association to Medium-Range Transport of Biomass Burning Plumes

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S1. Diurnal Cycle of Black Carbon and Mixing Layer Height

Urban ambient Black Carbon (BC) concentrations were determined from hourly data from 6 stations in Bogotá's Air Quality Monitoring Network. The BC is collected with Aethalometers model AE33. During the same period, Black Carbon data was collected at the Monserrate Sanctuary site as described in detail in the Methods section of the main manuscript. The daily mean profile for both datasets is shown in Figure S1. This analysis shows an overall similar diurnal profiles, with peak concentrations observed in the morning, likely associated to rush-hour traffic emissions. The mean ratio between ambient and background concentrations is 0.15. The similarity between both profiles, however, and the presence of a morning peak at the Monserrate site, suggests observations at the site are strongly impacted by urban emissions.
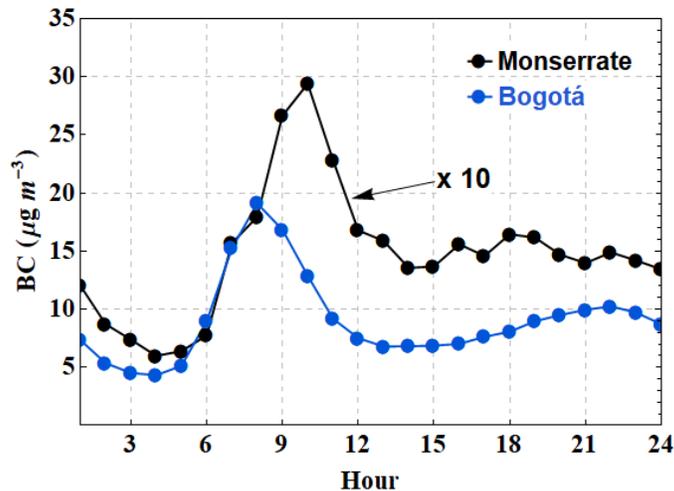
Figure S1. Hourly mean Black Carbon concentration measured at the Air Quality Monitoring Station Network of Bogotá (Blue) and at the Monserrate Sanctuary site (Black).

The two-hour lag between these two signals is likely produced by the altitude difference between the two measurement sites and the mixed layer morning expansion. To confirm this hypothesis, we analyzed mixing layer height (H_{mix}) from Bogotá's airport atmospheric sounding data. There is typically one radiosonde launch per day. Hourly H_{mix} was estimated by adding the difference between the hourly surface temperature recorded at the ground to the surface temperature reported in the radiosonde data. There is typically one radiosonde launch daily at 12Z (7:00 am local time). The Mixing Height is then computed as the height at which the temperature of a dry-adiabatically lifted air parcel with temperature equal to the hourly reported surface temperature

intersects the 12Z temperature profile. This is a modification of the method of Holzworth (1967), which was conceived with a focus on air quality applications. This was performed for all available data during the three-year monitoring period, and then, hourly mean values were computed.

Figure S2 shows the hourly mean estimate of H_{mix} according to the method described. The mixing height exceeds the Monserrate site altitude between 8.30 am and 9:00 am, roughly two hours after the beginning of the convective layer expansion.
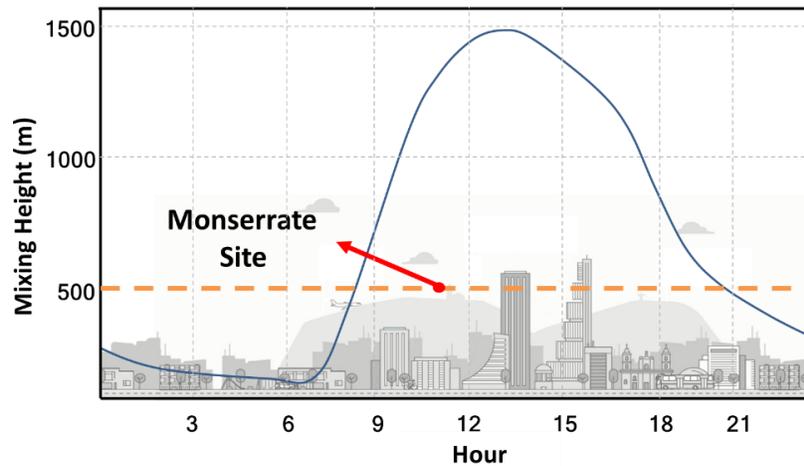
Figure S2. *Mean hourly mixing height according to the 12Z radiosonde temperature profile and hourly surface temperature reported in Bogotá. The Monserrate site height is depicted as the orange dashed line.*

S2. Annual Cycle of Daily Fire Counts

As described in detail in the manuscript (Section 2.4), we applied several methods to count the number of fires and produce time series of daily fire counts. We considered buffer-radii of 200 km, 400 km, 600 km, 1000 km, and 1500 km from Bogotá, and selected only those fires within that region. Additionally, annular regions, those formed between circular buffers of different radius were analyzed to identify specific emission areas in relation to their distance to Bogotá. Figure S3 shows mean monthly variation for number of daily fire counts for each buffer area identified for entire monitoring period. This analysis concludes counting types that includes farthest active fires (e.g. Figure S3-a, d, g) have the maximum peak in March, same month whit BrC highest concentrations.
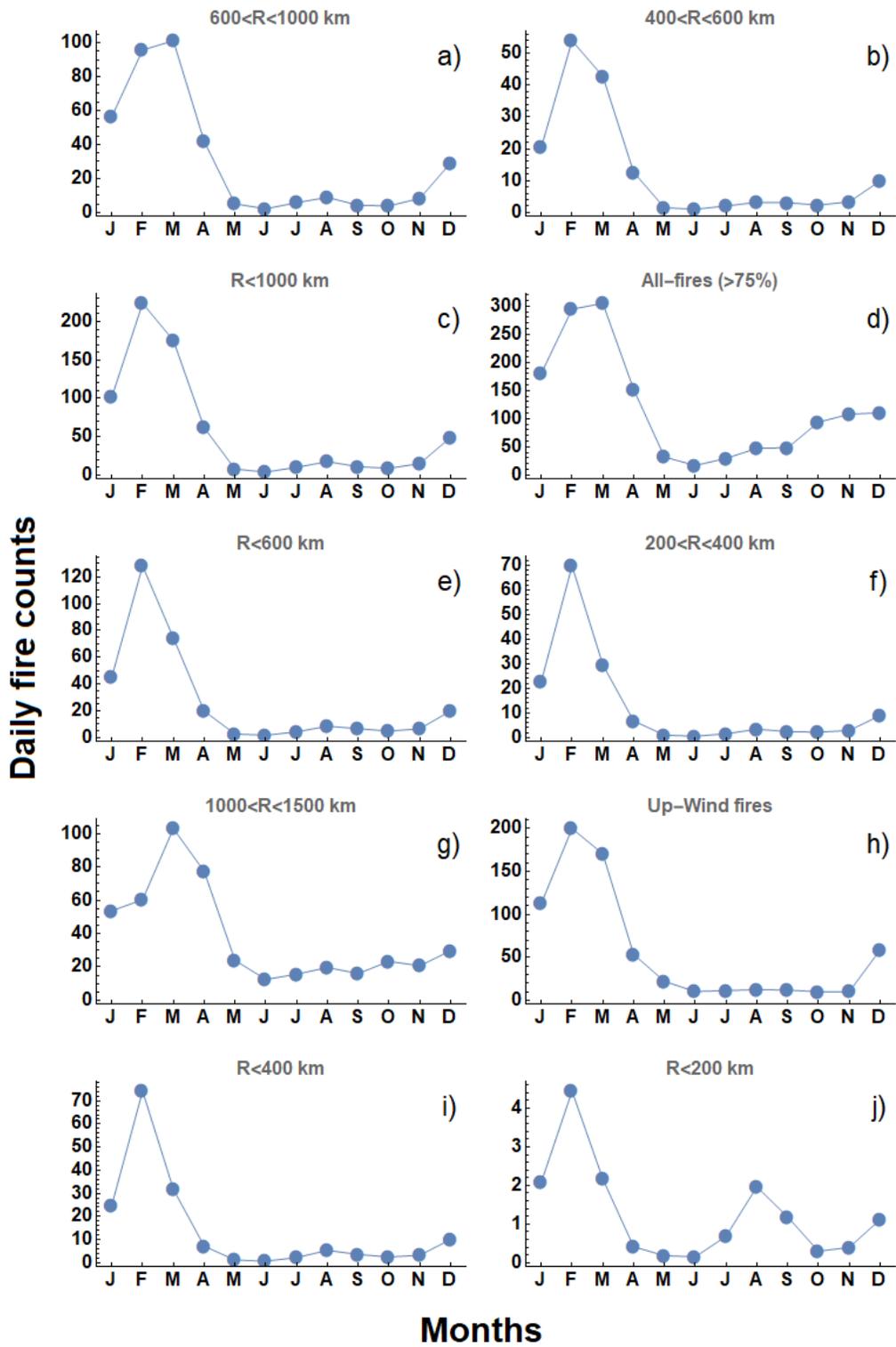
Figure S3 *Annual profile for all fire counting considered in the article.*

S3. Brown Carbon, Black Carbon, and Smoke tracer observations

We carried out three field campaigns, two during the high biomass burning activity season (Campaigns C1 and C3) and one during a low fire activity season (Campaign C2). During those campaigns, $PM_{2.5}$ filter samples were collected and analyzed for smoke tracers levoglucosan, water-soluble organic carbon (WSOC), and water-soluble potassium (WSK). Additional analysis to determine elemental and organic carbon were carried out. Elemental carbon concentrations were compared against on-line Black Carbon observations. The tracers were detected in concentrations above the limit of detection in every campaign, and good agreement was observed between chemical and optical measurements for both smoke tracers and fossil fuel tracers.

Figure S4 shows the time series of continuous Brown Carbon and Black Carbon observations at the Monserrate Site, as well as the daily mean concentration of the filter-based smoke tracers that were analyzed.
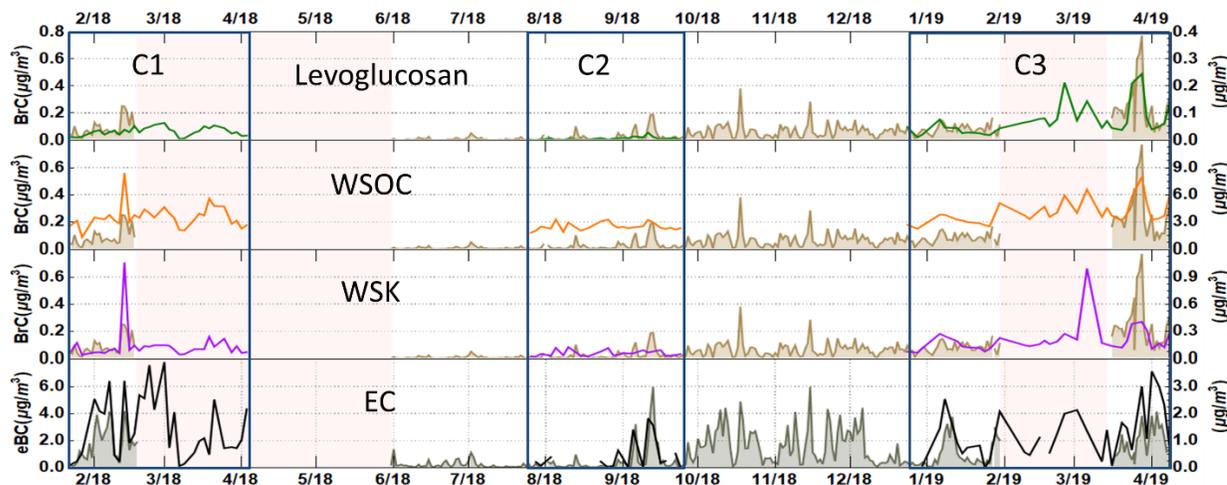
Figure S4. Daily time series of Brown Carbon (Brown, in the first three panels), Black Carbon (Gray, in the lowermost panel) for the entire observation period. Levoglucosan (green, upper panel), WSOC (Orange, second panel), WSK (Purple, third panel), and EC (Black, lower panel), are included for the three filter sampling campaigns carried out. Letters C1, C2, and C3 mark the periods of filter samples collection.

S4. Biomass burning emissions footprint

In order to identify the spatial distribution biomass burning aerosol sources that could affect Bogotá's air quality, we paired the in-situ BrC concentrations observed at the Monserrate Site, HYSPLIT back-trajectories, and MODIS detected fires to apply two different methodologies. Concentration Weighted Trajectory (CWT) is a widely used method to determine spatial distribution of sources in air quality applications. In this procedure (Equation 1), each grid cell in the domain is assigned a concentration obtained by weighting measured receptor-site concentrations by the residence time of associated back-trajectories that crossed that grid cell (Siebert, 1994), i.e.,

$$\bar{C}_{ij} = \frac{\sum_{k=1}^{N} C_k \, \tau_{ijk}}{\sum_{k=1}^{N} \tau_{ijk}} \qquad\qquad (1)$$

In Equation 1, $\bar{C}_{ij}$ is the average concentration assigned to grid-cell (i,j), C_k is the concentration recorded at Monserrate site in day k, τ_{ijk} is the residence time in the cell (i,j) of a back-trajectory arriving at the receptor site in day k, and the summation is over all sampling days N.

In our analysis, we separated the observation period in seasons, namely DJFM (high-BB season), AMJJ (low-BB season), and ASON (transition period). The result of applying Equation 1 to these seasons for the three years of BrC data is shown in Figure S5a, S5c, and S5e. The spatial distribution is strongly influenced by the seasonal wind patterns. During DJFM, longer trajectories are observed because the southward displacement of the ITCZ causes the region to be in the trade-wind regime. Therefore, DJFM has both, high-BB activity and winds blowing from those source regions towards the receptor. The second period, AMJJ, has the lowest fire activity and a clear wind direction predominance from south and west, i.e., away from the potential source regions. The third period (ASON) has no dominant wind direction, with short trajectories and therefore high density of trajectories in the vicinity of Bogotá. The highest BrC concentrations are assigned close to the city, this, likely due to the high density of trajectories crossing those grid cells. This methodology attributes concentrations depending trajectories density and beyond continental areas, this attributions does not identify sources that could emit BrC to the atmosphere.

After identifying the above mentioned limitations when the CWT methodology is applied, we developed a variation of this method that allows us to relate the measured concentrations to the

origin of the emissions. We performed two modifications to Equation 1. First, we calculated the Fire Radiative Power (FRP) for MODIS hot-spots that were within a 150 km buffer of the back-trajectory location. This is, we defined the variable FRP_{ijk} as the sum of the FRP for all individual fires in grid cell (i, j) that were within a 150 km radius of the location of a back-trajectory arriving at the receptor site on day k. Additionally, we applied a dilution term, in which an exponential decay in term as air masses travel towards the receptor. Therefore, in this method, $\bar{C}_{ij}$ is calculated as

$$\bar{C}_{ij} = \frac{\sum_{k=1}^{N} C_d * FRP_{ijk} * e^{\frac{-t}{\tau}}}{\sum_{k=1}^{N} FRP_{ijk} * e^{\frac{-t}{\tau}}} \qquad (2)$$

The term $e^{\frac{-t}{\tau}}$ in Equation 2 accounts for dilution. The dilution time-scale was assumed to be 5 days, i.e., $\tau = 5\ days$. Unlike the CWT methodology, the new Concentration Trajectory Weighted by FRP methodology identifies specific areas where the MODIS sensor usually identifies large-scale active fires over which air masses eventually reaching the measurement site pass by. Therefore, those areas could be potential sources with impact on the BrC concentrations measured in Bogotá. The results of applying this method for the three years of measurements are shown in FigureS3b, S5d, and S5f. The resulting spatial footprint of Biomass Burning sources clearly suggest that open biomass burning in the eastern savannas of the Orinoco river basin are the most up-wind significant sources of smoke for our measurement site.
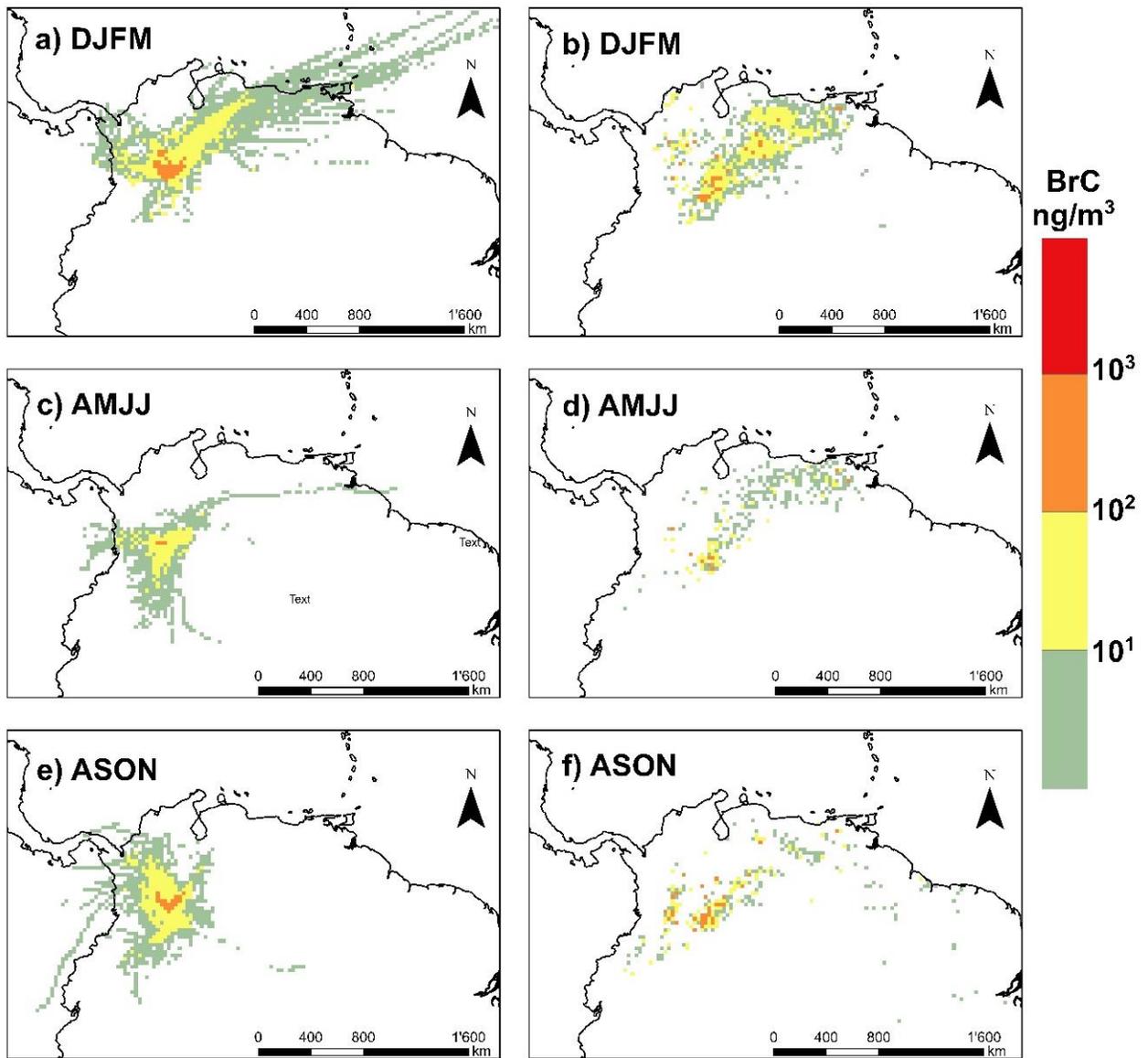
Figure S5 *Mean Brown Carbon concentrations, $\bar{C}_{ij}$, after applying the spatial allocation algorithms described by Equation 1 (panels a., c., and d.) and Equation 2 (panels b., d., f.).*