

Black Carbon Emissions from Associated Natural Gas Flaring

Supporting Information

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Associated gas composition and characteristics

Table S1: Composition of associated gas in mol %. The percent carbon is the mass of carbon divided by the mass of the fuel including H₂O, CO₂, and N₂. The percent carbon of the hydrocarbons (HC) excludes these gases. ¹Hypothetical gas compositions used in Caulton et. al. (2014) [4]. ²Approximated gas composition in Haynesville (HV), Antrim (Ant), New Albany (NA), Fayetteville (FV), Marcellus (Mar), and Barnett (Bar) formations [3]. ³Average gas composition in the Bakken formation, ND as tabulated by EERC [16].

	High ¹	Med ¹	Low ¹	HV	Ant	NA	FV	Mar	Bar	Bakken
methane	1.000	0.800	0.600	0.950	0.620	0.899	0.973	0.852	0.868	0.577
ethane	0.000	0.100	0.200	0.001	0.042	0.011	0.010	0.113	0.067	0.199
propane	0.000	0.050	0.120	0.000	0.011	0.011	0.000	0.029	0.020	0.113
n-butane	0.000	0.030	0.050							0.028
isobutane										0.010
n-pentane	0.000	0.020	0.030							0.006
isopentane										0.004
hexane										0.002
heptane										0.001
octane										0.000
nonane										0.000
nitrogen				0.001	0.290		0.007	0.003	0.029	0.052
carbon dioxide				0.048	0.038	0.079	0.010	0.004	0.017	0.006
water										0.000
% Carbon				0.712	0.506	0.691	0.736	0.751	0.719	0.727
% Carbon HC	0.749	0.761	0.773	0.749	0.753	0.750	0.749	0.756	0.754	0.772
Density (g/m3)	0.668	0.886	1.089	0.636	0.487	0.636	0.663	0.766	0.702	0.985

BC emission factor relationship with flare volume

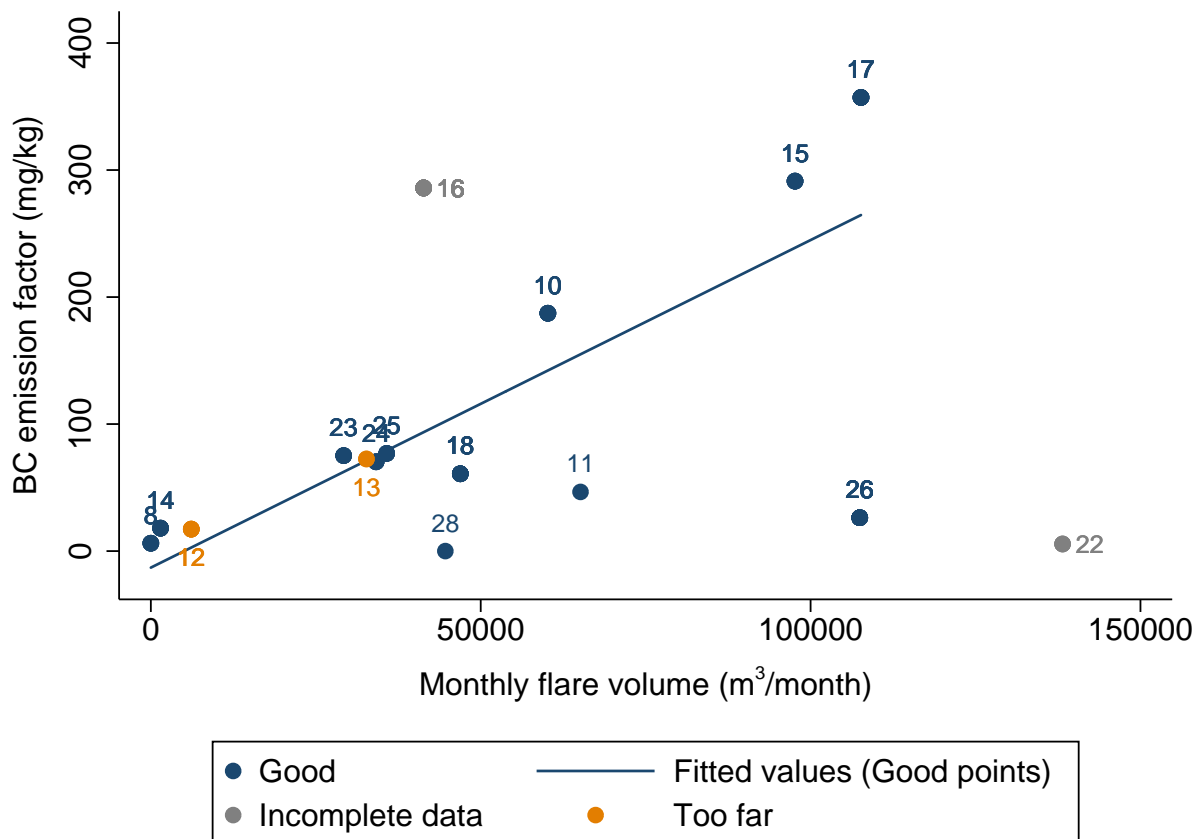


Figure S1: BC emission factor versus m³ of gas flared per month, as reported in North Dakota Industrial Commission (NDIC) database [9]. The points represent the average BC emission factor for each flare. “Good” points are flares that could be clearly matched with well-by-well flaring data had complete flare volume information. “Incomplete data” points are those where information was not available for flaring volume from some wells connected to the flare. “Too far” data points were those where the GPS location was not within 200 m of the well heads listed in the NDIC database. The fit line uses only the “good” points.

BC mass coating estimation

The SP2 indicates that most particles are thinly coated. Figure S2 shows a distribution of the incandescent lag times for individual particles for a range of mass-equivalent particle diameters and can be used to indicate BC mixing state (“thickly-” or “thinly-coated” BC cores, as in Subramanian et al. 2010 [15].) Particles that have a lagtime near zero are considered to be thinly coated. The peak and most of the distribution is near zero in this example flight and is similar for the other flights. Few thickly-coated particles were observed. Thickly-coated particles have high lagtimes (2 μ s and above), and approximately indicate particles with larger than 40 nm coating). Figure S2 represents both BC in flares and BC in background.

There could be some BC cores that have a BC-on-edge structure [13], though they are in the minority, and there is some uncertainty of such a classification based on SP2 operating conditions [14]. However, most of the flare BC emissions are thinly-coated, based on this analysis.

To estimate possible absorption enhancement due to particle coating, we estimated the amount of material in the plume that might condense on the BC. The fraction of condensable hydrocarbons relative to methane in the fuel gas was estimated using the fuel composition in the Bakken (Table S1) from EERC at measured

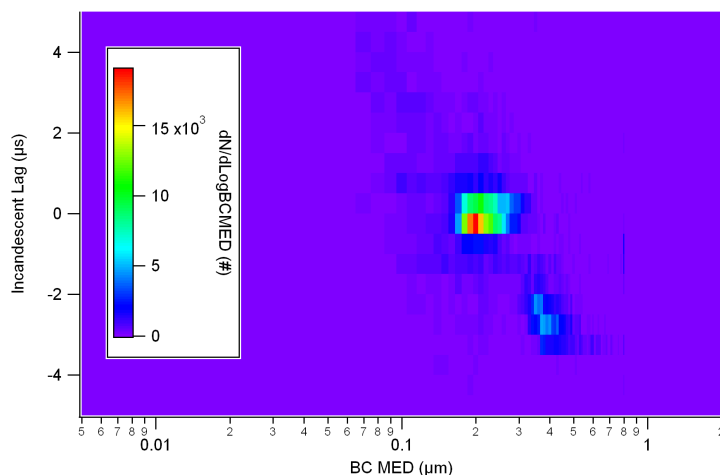


Figure S2: Lagtime analysis for BC coating thickness.

temperature in the plumes (6.7 ± 2.4 °C) at 0.07 grams condensable material per gram methane. The emission factor for CH₄ (6 g/kg) measured here was multiplied by this ratio to determine an approximate emission factor for condensable hydrocarbons of approximately 0.4 g/kg associated gas. The assumption is that the destruction removal efficiency does not vary between hydrocarbons.

The emission factor of condensable hydrocarbons is about three times greater by mass than that of BC. If this material condensed with an even distribution among BC particles of 200 nm diameter, the mass of each particle would increase by nine fg and each particle would be coated with a shell of 50 nm. The absorption enhancement of such a particle, assuming a core-shell morphology, was determined using Mie theory as in Bond et al. (2006) using Matlab code developed by Mätzler [2, 7]. The predicted absorption enhancement was a factor of 1.4.

SP2 calibration

BC mass was measured with the Single Particle Soot Photometer (SP2, Droplet Measurement Technologies) [12, 8]). In line with community recommendations[1], the SP2 was calibrated using Fullerene soot, Lot Number L20W054, using the fullerene soot mobility density equation from Gysel et al. (2011) [5]. In subsequent laboratory experiments, thermodenuded cookstove soot was size-selected by a DMA and then sampled by the SP2 and a Centrifugal Particle Mass Analyzer (CPMA, Cambustion) [10]. The SP2 and CPMA masses agreed to within 10% (slope 1.08, $R^2 = 0.99$), indicating that the SP2 results presented in this paper are reasonably robust. (This calibration verification is presented in a manuscript currently under review at *Aerosol Science and Technology*: Saliba et al. (2016) [11].) The flow rate was measured in flight and was 180 ± 50 cc/min throughout the entire sampling campaign.

The mass missed was based on a lognormal fit as in Subramanian et al. (2010) [15] and is calculated for the small accumulation mode particles. Most of the particles missed were below the detection limit of the instrument because of their small size. The mass of particles missed was approximately 0.5-2% and an upper limit of 5% was stated, given the approximate nature of the analysis. For accumulation mode particles on the larger end, the gain settings of the SP2 incandescence detectors were set so BC particles up to a mass-equivalent diameter of ~ 800 nm (at a density of 1.8 g/cm³, 480 fg-BC/particle) were detected properly by the SP2. As the BC size distributions show (Figure S3), all the flare emissions were well below this limit. The mass-equivalent diameter range observed was 0.05-0.8 μ m.

Particle coincidence in the SP2

At the high source concentrations where our measurements were made, particle coincidence can be an issue. The code described in Holder et al. (2014) [6] was applied to correct for coincident particles. In

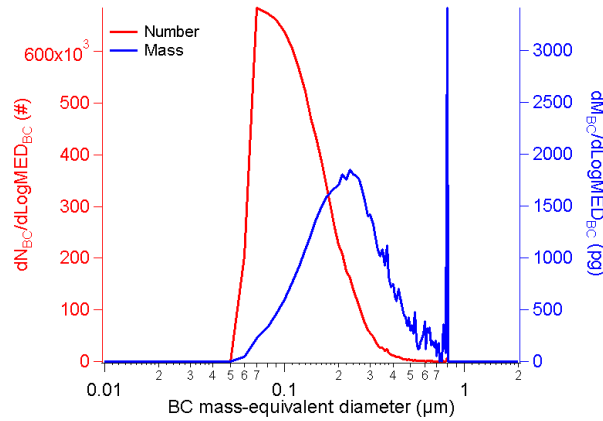


Figure S3: Size distribution of particle number and mass detected in the SP2 for flares on March 25.

that paper and in our analysis, the number concentration was more influenced by coincidence effects than the mass concentration (20% increase in number concentration and 8% increase in mass concentration in Holder et al (2014)). In our analysis, the mass was increased by 5-10% in individual flare passes when coincidence was corrected for. Figure S4 shows the shift in mass-equivalent size distribution of BC cores as originally processed, and after reanalysis with the code from Holder et al. (2014). The size distribution from reanalysis has more particles of small mass. This is expected because the original analysis chooses the largest incandescent peak, so the retrieved coincident particles must have smaller masses. This also explains why even though the coincidence effect is significant for number concentrations above 2000 #/cm³, the effect on BC mass is much smaller. Our measurements produce BC emission factors that may be under-reported by up to 10% and MAC that may be over-reported by up to 10% due to saturation in the SP2.

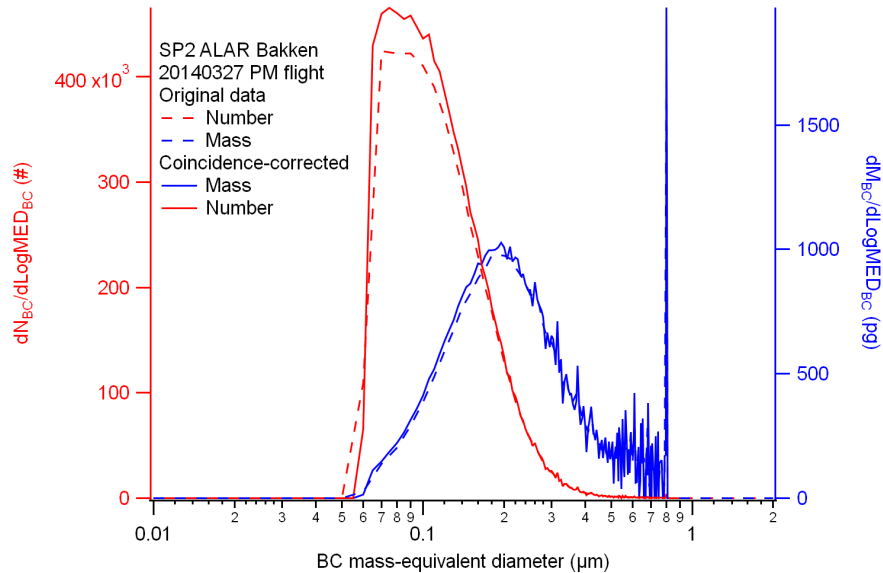


Figure S4: Coincidence corrected number and mass concentrations of BC measured in the SP2.

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