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# Black Carbon Emissions from the Bakken Oil and Gas Development Region

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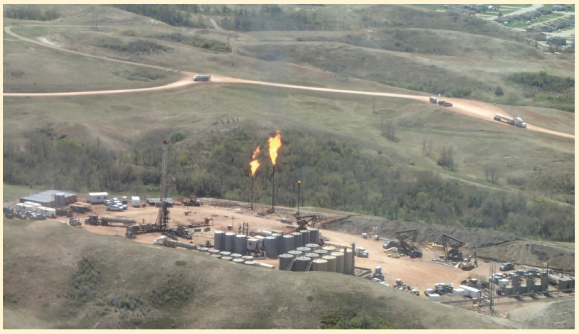
Supporting Information

**ABSTRACT:** Black carbon (BC) emission rates from the Bakken oil-producing region of North Dakota have been quantified with a NOAA airborne single-particle soot photometer (SP2). Flights in May 2014 led to six measurements of the BC emission rate in the region. Oil and gas operations (associated flaring, diesel engines associated with pumping and drilling, and oil production-related transport), limited agricultural burning, and sparse urban/transport sector activity contribute to these emissions. The BC emission rate was 1400 ± 360 t year$^{-1}$, implying that Bakken production activities are unlikely to contribute to large-scale biases in estimates of BC emissions. An upper limit on the BC emission factor from flaring based on these observations is 0.57 ± 0.14 g/m$^{-3}$. Flaring BC was not associated with optically significant internally mixed non-BC material or with significant emissions of non-BC-containing primary aerosol. BC in the outflow from the region was also generally externally mixed.



## INTRODUCTION

The Bakken region extending over northwestern North Dakota and southeast Saskatchewan hosts extensive fossil-fuel extraction activities, including both shale and tight-sand extraction of crude oil and associated petroleum (natural) gas. Gas and oil development in the region continues to see dramatic increases initiated in the past decade. For example, between May 2013 and May 2014, the number of wells and the amount of oil and gas production in the region increased by 30%.[1] At present, the mining activities in the region have outpaced the development of typical infrastructure, and thus, there is considerable associated flaring of gas. In May 2014, flaring consumed 28%[1] of the total gas produced from operations.

The large-scale nature of development raises questions about the emission of black carbon (BC), a product of incomplete combustion that is a major anthropogenic forcer of climate[2] and a focus for possible mitigation efforts to permit short-term reductions in climate forcing and for health co-benefits.[3] Modeling efforts suggest that flaring BC can have large impacts on climate,[4] but the emissions inventories they use are not yet well constrained with measurements in the ambient atmosphere.

Here we present the results of black carbon measurements made as part of a NOAA initiative to quantify oil production emissions. The TOPDOWN 2014 (Twin Otter Projects: Defining Oil/gas Well emissioNs) mission was based out of Minot, one of two central cities in the Bakken region of North Dakota, and involved flights in United States airspace. Figure 1 shows a map of the region in which the northwest corner of North Dakota abuts the southern Canadian and eastern Montana borders. Active oil and gas wells are shown with small black dots.[1] During the mission, in May 2014, some periods of steady high wind provided an excellent opportunity to evaluate the emission rate of black carbon from the area indicated by cross-hatching, which contains 80% by number of all ND wells. A single-particle soot photometer (SP2, Droplet Measurement Technology Inc., Boulder, CO) quantified black carbon atmospheric concentrations and also provided information about the microphysical state of this aerosol material. Experimental Methods describes the instrumentation, analysis, and relevant flight details of the mission, while the observations and contextual comparison to existing emissions inventories are provided in Results and Discussion. The Supporting Information provides relevant details.

## EXPERIMENTAL METHODS

The SP2, its data products, and its operation have been described extensively in the literature.[5,6] Briefly, the SP2, as

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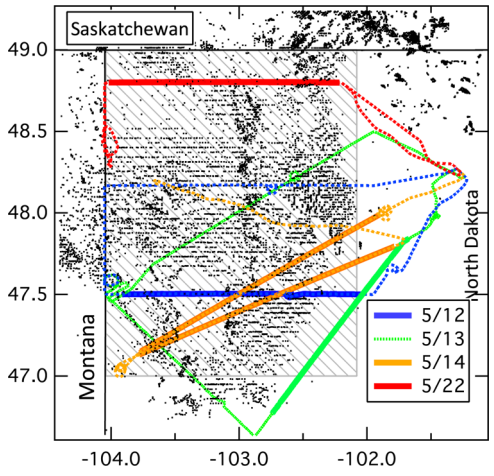


**Figure 1.** Map of the Bakken region, showing (hatched area) the region integrated in the NEI emissions inventory. The flight tracks on May 12 (blue), 13 (green), 14 (orange), and 22 (red), with mass balance transects in solid and other portions of the flights in dotted lines, and the location of oil and gas wells (black dots). Winds were from the north and northwest for May 12–14 and from the south for May 22.

configured for TOPDOWN, quantifies the refractory black carbon (rBC) content of individual aerosol particles in the mass range of ~0.7–160 fg [corresponding to an 80–550 nm volume-equivalent diameter (VED) assuming 1 g/cm$^3$ void-free density]. The SP2 also measures the total optical size of rBC-containing particles with 3–8 fg of rBC mass content,[5,7] thus allowing estimation of the amount of non-rBC material in an individual particle.[6] The size distribution of rBC observed here was fit with a log-normal function to evaluate the fraction of the accumulation mode rBC mass detected by the SP2. On the basis of this analysis, the observed mass mixing ratios were corrected upward to better represent the total accumulation mode rBC mass in the air. The correction ranged from 15% on most days to 30% on May 22. rBC in either smaller or larger sized modes would not be reflected in the corrected SP2 measurement; there was no indication of significant mass in such modes in the data set.

The SP2 was calibrated following community recommendations.[8] "rBC" is the accepted term for the material quantified by the SP2,[9] and this material has been shown to be equivalent to elemental carbon (EC), as measured under conditions minimizing possible biases, at the level of 15%.[10] "EC" is the term associated with BC measurements performed with a thermal decomposition technique that is often used for emissions inventories.[9] Details of the calibration, aircraft inlet, and sampling line configuration are provided in the Supporting Information.

## RESULTS AND DISCUSSION

rBC mass emission rate from primarily the North Dakota portion of the basin was calculated for flights on days with very steady and relatively high winds (May 12–14 and 22). On these days, the Twin-Otter flew in the boundary layer, with occasional climbs well above the top of the mixed layer and into the free troposphere to allow identification of the mixed layer height, and to evaluate how well mixed the boundary layer was. Mixed layer height was determined from analysis of vertical profiles of high-signal tracers, including methane, water vapor, ethane, ozone, and potential temperature, which were consistent with a well-mixed layer, including a discernible transition to the free troposphere. All flights reported here occurred in the afternoons after the boundary layer had been well established. Figure 1 shows the flight tracks for these flights; on May 22, the wind was out of the south (with two transects at different altitudes overlaid on the map), on May 12 out of the north, and on all other flights out of the north–northwest. The heavy portions of the tracks indicate the transects used here to determine BC emission rates; sampling on different days with different winds provides confidence that sources outside of the region explored are not strongly contributing to the emission rates calculated here (two transects on May 22 overlie each other). Transects were oriented within a small angle $\alpha$ from orthogonally to wind direction, and the average wind direction and wind speed ($S_W$) were averaged for each leg. Flight tracks were extended beyond the areas influenced by outflow from the basin to allow estimation of the mass flux that can be attributed to background air (i.e., the mass flux of rBC into the basin from more distant sources). The net rBC emission rate in the basin from each transect ($F_{rBC}$) was calculated using established mass balance techniques,[11,12] here presented in a simplified formulation:

$$F_{rBC} = S_W \cos(\alpha) \times lh\beta(C_O - C_{BG}) \quad (1)$$

where $l$ is the length of the flight track influenced by basin emissions, $C_O$ is the average ambient concentration of rBC in the outflow of that track length, $C_{BG}$ is the average concentration of rBC in the background air not influenced by local emissions, and $h$ is the height over which emissions are mixed. Hence, $S_W \cos(\alpha) \times lh$ is the volume of air containing the emissions that is swept over the region per unit time, and $C_O - C_{BG}$ is the enhanced concentration of rBC in that volume. $\beta$ is a correction factor to account for nonuniform mixing in the

**Table 1. Relevant Parameters for Six Transects Obtained with the NOAA Twin-Otter Aircraft**[a]

| 2014 date | local time (pm) | transect altitude (m) | $\alpha$ (rad) | $S_W$ (m s$^{-1}$) | $l$ (km) | $h$ (m) | $C_O$ (ng m$^{-3}$) | $C_{BG}$ (ng m$^{-3}$) | $\beta$ | $F_{rBC}$ (g s$^{-1}$) |
|---|---|---|---|---|---|---|---|---|---|---|
| May 12 | 5:45 | 1200 | 0.33 | 15.2 | 128 | 2308 | 30.4 | 18.1 | 0.89 | 49.5 |
| May 13 | 3:30 | 1020 | 0.20 | 13.1 | 144 | 2271 | 50.1 | 35.7 | 0.91 | 58.2 |
| May 14 | 4:10 | 1050 | 0.16 | 7.6 | 167 | 1956 | 65.5 | 46.5 | 0.96 | 47.5 |
| May 14 | 5:30 | 1350 | 0.14 | 8.1 | 168 | 2022 | 65.9 | 46.1 | 0.97 | 47.0 |
| May 22 | 4:45 | 1710 | 0.14 | 7.7 | 133 | 2112 | 80.1 | 49.5 | 0.91 | 39.2 |
| May 22 | 3:30 | 1140 | 0.08 | 7.1 | 132 | 2068 | 63.9 | 46.1 | 0.92 | 36.9 |
| mean | | | | | | | | | 0.93 | 45.8 ± 3.5 |

[a]The central columns provide parameters used in the mass emission rate calculation. The mean value of the rBC emission rate from the region is shown at the bottom of the table, with its statistical uncertainty. The ground level varied over ~600–800 m altitude. BC concentrations are corrected to represent average ambient pressure and temperature in the mixed layer.



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transition from the fully mixed layer to the free troposphere (further discussed in the Supporting Information). Here it averaged 0.93. Table 1 shows the values of the various parameters for the six transects suitable for emission analysis.

Observations of methane (a clear tracer of oil/gas production) during the climbs to the free troposphere mentioned above indicate that the layer was homogeneously mixed vertically up to a transition layer with <10% variability; hence, we simply use the average value observed in each track, corrected for temperature and pressure, to reflect the concentration throughout the layer, with the correction ($\beta$) mentioned above to address the transition to air free of these emissions. Flight tracks conducted at different altitudes within the mixed layer (as shown in Table 1) produce very consistent estimates for the emission rate.

On the first transect for May 22 shown in the table, the contributions of a single exceptional burning source encountered were excluded from contributing to the average observed rBC concentrations. The rBC particles in this plume were substantially larger than those generated from flares or observed in the outflow from the region; on the basis of characteristics of individual flare emissions in the area, it is not possible that this source was associated with a single "rogue" flare. If included, it would have increased the average emission rate for the data set by ∼10%. No other open burning plumes near this scale were observed, and generally, agricultural activity, including burns and field work with tractors, appeared to be very limited on the basis of assessments made by scientists flying on the aircraft.

The largest uncertainty is in SP2 calibration, which has been tied to ambient rBC sensitivity at the level of 15%.[13] On the basis of the sensitivities noted upon determination of mixed layer height above ground level, average enhanced concentration, and wind speed from the data, we conservatively estimate their uncertainties at 10% each, with an additional 10% to reflect uncertainty in the uniformity of the vertically mixed rBC concentration. Uncertainties associated with the wind angle and length of transect integration are negligible in this analysis. Summing the systematic sources in quadrature provides an overall relative systematic uncertainty estimate in the measured rBC emission rate from the measured sources of 25%. The statistical uncertainty in the determination, as shown in Table 1, is relatively small, only 8%, reflecting good consistency between estimates from different days and wind directions and providing confidence that the measurements do a good job covering the hatched area of Figure 1 and rejecting significant biases from other sources. After assumption of constant emission rates throughout the day and night, implying that diurnal emissions associated with agricultural activity and other anthropogenic sources are unimportant here, the data lead to an overall estimate of rBC emissions for the sampled portion of the North Dakota Bakken region of 1400 ± 360 t/year. To the extent that daily activities associated with non-oil/gas sector activity do wane at night, these contributions are overestimated.

The measurements bound the possible emission rate from associated gas flaring. To this end, we pair the state estimate of associated gas flaring for May 2014 in the region[1] to the total rBC emission rate that we measured here, scaled down by 20% to estimate gas emissions only from the wells in the hatched area of Figure 1. This generates an upper limit on the emission factor from flaring: 0.57 ± 0.14 g of rBC/($m^3$ of gas flared) (at STP).

The microphysical state of the rBC emitted specifically by flaring, and generally from all sources in the region, was determined. Figure S2 presents mass size distributions of rBC observed (1) under nascent conditions produced by individual flares and sampled within minutes of emission and (2) in the outflows used to produce the $F_{rBC}$ estimate (excluding the single outflow strongly influenced by open burning). The flare distributions were corrected to remove background air contributions concurrently sampled. The rBC mass median diameters average 166 ± 2 nm in the outflow (excepting the transect strongly influenced by open burning), which is larger than the rBC produced in U.S. metropolises,[14] and is more consistent with uncontrolled biomass burning rBC emissions.[15] This is consistent with substantial contributions to the total rBC emissions from flaring, which, when specifically analyzed, was associated with rBC with a mass median diameter of 192 ± 6 nm. However, unlike in uncontrolled biomass burning, the rBC generated by flares was not associated with optically detectable (by the SP2) amounts of non-rBC material, and without accumulation-mode rBC-free aerosols. This indicates that the flares produce nearly pure rBC that, on the basis of the work of Bond et al.,[2] exerts a positive climate forcing. The rBC-containing particles exported from the basin showed very similar characteristics, indicating that there was no substantial secondary production of aerosol materials condensing or coagulating with the rBC on time scales of hours and supporting our assumption that open burning, which typically produces and thickly coated rBC[15] and copious amounts of organic carbon aerosol, did not contribute substantially to the net emissions during the observations.

The most recent inventories of BC emissions from the Bakken region of North Dakota are provided by the Environmental Protection Agency's National Emissions Inventory Database (EPA NEI), which in 2013 released an inventory appropriate for 2011 emissions (NEI-2011).[16] The inventory includes oil and gas production sources but associates no EC with them in the Bakken, and hence obviously underestimates them; ∼70% of the NEI EC emissions are attributed to off-road diesel emissions from agricultural activities (tractors), with ∼15% each from agricultural open burning and on-road sources. The May EC emission rate in the NEI is 800 t/year over the hatched area of Figure 1, a factor of 2 less than we observed in 2014. Bakken oil (gas) production increased by a factor of 2.5 (2.9) between May 2011 (appropriate for the NEI estimate) and May 2014 (appropriate for the observations here). Flaring increased by a factor of 3.3 over this time.[1] Only limited agricultural activities were observed during the flights, so if oil/gas production-related sources are in fact the dominant emitters in this season, this would suggest that NEI estimates were reasonably accurate in 2011 but misapportioned emission sources. On the other hand, estimates of BC produced by gas flaring may be biased high. The GAINS model[17] has an emission factor of 1.6 g of rBC/($m^3$ of gas flared) (STP), which is a factor of 2.6 higher than the upper limit generated here. A more recent laboratory estimate of flaring emission factors,[18] which also was based on laser-induced incandesce measurements, produced a reduced estimate of 0.51 g of rBC/($m^3$ of gas flared); this was referenced as an estimate of flaring that was still consistent with Arctic flaring being significant for climate.[4] This value is also consistent with our upper limit. As we do not have measurements in other fields, it is not clear how well our


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estimate will translate to flaring conditions in the rest of the world.

Total anthropogenic and natural North American BC emissions were estimated in the year 2000 at 380000 t/year;[2] hence, the total yearly rBC emissions estimated here, which are only partially due to the exceptional amount of gas flaring and oil/gas production activities in the Bakken (nearly 40% of the U.S. flaring total[19]), make up <1% of that total. It follows that lack of accounting for oil/gas production sources does not very significantly contribute to any large-scale underestimate of BC emissions or BC direct radiative forcing from North America.

On global scales, North America makes up less than 10% of total flaring.[19] Thus, global-scale oil/gas production activities and associated flaring may produce significant amounts of BC, especially in the context of regional impacts.[4]

## ■ ASSOCIATED CONTENT

### Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.estlett.5b00225.

Two figures and expanded technical discussion about the SP2 calibration and installation in the aircraft and the determination of mixing height (PDF)

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### Author Contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript. J.P.S. wrote the primary manuscript. J.P.S., J.S.H., J.M.K., and J.P. analyzed data. J.P.S. and J.S.H. shared responsibility for the instrument. M.L.S. and J.P. contributed to operating the instrument on the aircraft. S.M. contributed modeling and inventory calculations. E.A.K., T.B.R., and C.S. contributed to project organization, flight planning, and logistics.

### Notes

The authors declare no competing financial interest.

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## ■ ABBREVIATIONS

BC, black carbon; SP2, single-particle soot photometer; VED, volume-equivalent diameter

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