Gas emissions, minerals, and tars associated with three coal fires, Powder River Basin, USA

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Abstract

Ground-based surveys of three coal fires and airborne surveys of two of the fires were conducted near Sheridan, Wyoming. The fires occur in natural outcrops and in abandoned mines, all containing Paleocene-age subbituminous coals. Diffuse (carbon dioxide (CO2) only) and vent (CO2, carbon monoxide (CO), methane, hydrogen sulfide (H2S), and elemental mercury) emission estimates were made for each of the fires. Additionally, gas samples were collected for volatile organic compound (VOC) analysis and showed a large range in variation between vents. The fires produce locally dangerous levels of CO, CO2, H2S, and benzene, among other gases. At one fire in an abandoned coal mine, trends in gas and tar composition followed a change in topography. Total CO2 fluxes for the fires from airborne, ground-based, and rate of fire advancement estimates ranged from 0.9 to 780 mg/s/m2 and are comparable to other coal fires worldwide. Samples of tar and coal-fire minerals collected from the mouth of vents provided insight into the behavior and formation of the coal fires.

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1. Introduction

Thousands of coal fires occur worldwide, in every major coal-bearing region and on all continents except for Antarctica (Stracher, 2010). Emissions from coal fires are a known, but poorly quantified, source of greenhouse gases; e.g. carbon dioxide (CO2) only and vent (CO2, carbon monoxide (CO), methane, hydrogen sulfide (H2S), and elemental mercury) emission estimates were made for each of the fires. Additionally, gas samples were collected for volatile organic compound (VOC) analysis and showed a large range in variation between vents. The fires produce locally dangerous levels of CO, CO2, H2S, and benzene, among other gases. At one fire in an abandoned coal mine, trends in gas and tar composition followed a change in topography. Total CO2 fluxes for the fires from airborne, ground-based, and rate of fire advancement estimates ranged from 0.9 to 780 mg/s/m2 and are comparable to other coal fires worldwide. Samples of tar and coal-fire minerals collected from the mouth of vents provided insight into the behavior and formation of the coal fires.

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Compounds found in the exhaust from coal fires, mentioned above, are similar to those found in the flue gas from coal-fired power plants, but are not subject to emission controls and technologies that minimize their release and potential impacts. The composition of the coal-fire exhaust gases has been shown to depend on the chemistry of the coal, oxygen (O₂) concentration, temperature, and physical factors such as fracturing of the overburden (Ide et al., 2010). The dominant transport mechanism for coal-fire exhaust gases includes the following: 1) convection and advection outward from gas vents and fractures; and 2) diffusion up into the soil column and eventually the atmospheric boundary layer (Engle et al., 2011). To adequately characterize emissions from a coal fire, estimates must be based on both mechanisms.

Dozens of natural and human-induced coal fires are active in the Powder River Basin (PRB) of southeast Montana and northeast Wyoming, the largest source of coal produced in the United States (Fig. 1). Coal fires exhibit surface features similar to those found above high-temperature geothermal systems including gas vents, elevated surface temperatures, expansion and subsidence related fractures, zones of dead vegetation, and geochemical alteration; i.e. clinker. The subbituminous coal beds of the Paleocene Fort Union Formation and Eocene Wasatch Formation are prone to ignition, and have burned throughout the past few million years (Reiners et al., 2011; Riihimäki et al., 2009) creating 3700 km² of reddish clinker outcrops that cap hills and ridges throughout the basin. The coal may ignite as a result of range and forest fires, lightning strikes, arson or carelessness, or spontaneous combustion. Four main types of coal fires occur: 1) fires in abandoned underground mines; 2) fires in highswords and spoil piles in active surface mines; 3) fires on natural coal outcrops; and 4) low-level smoldering of coal beds (Heffern and Coates, 2004). Scientists with the U.S. Bureau of Land Management (BLM) have documented 81 known historic and active coal fires in the PRB, using results from a 11–15 May 2009 field campaign that included ground- and aerial-based surveys of three coal fires located roughly 16 km north of Sheridan, Wyoming. The details concerning CO₂ and CH₄ emission estimates and comparison of ground-based and airborne methods for the Welch Ranch site were discussed in detail by Engle et al. (2011). Results from that work show that the two methods gave emissions estimates that agree within a factor of two: the total CO₂ emitted from the soil and vents was 6.8–9.0 t CO₂/d and 19–25 kg CH₄/d using the ground-based approach compared with airborne estimates of 3.5–4.1 t CO₂/d and 11–13 kg CH₄/d.

2. Description of the coal fires

The three fires examined in this paper occur along a southwest-to-northeast trend near the banks of the Tongue River in north-central Wyoming and have distinct geomorphic features (Fig. 1). The most northern fire investigated, the Ankney fire, occurs in a natural outcrop along the cut bank of the river and is of unknown origin but thought to have started circa 1990. The fire primarily consumes the Anderson coal bed and no attempt has been made to extinguish it. The surface expression of the Ankney fire is a series of arcuate fractures associated with the slumping/collapse of the surface that lay above voids left by the burned-out coal (Fig. 2). The fire is marked by the absence of vegetation in the most active areas, a consequence of the heat at the surface. A map of the burned-out area at the Ankney site (Fig. 3) shows that the fire front, as represented by fractures, has advanced to the southeast at an average rate of 6 m/yr over the past 7 years. Measurements by the BLM show that the fire-impacted area increased from 0.6 ha in May 2004 to 1.4 ha in September 2011 (Fig. 3).

The two southern fires, the Welch Ranch and Hotchkiss fires, have been burning in the Monarch coal (and Dietz 2 and 3 for the Welch Ranch fire) since the early to mid 1900s and are likely related to underground coal mining. The Acme mine, the northern edge of which is on the Welch Ranch now owned by BLM, operated from 1911 through 1940 (Fig. 1). Total disturbance from the mine includes >200 ha of underground operations (Dunrud and Osterwald, 1980), which eventually subsided. As the rooms between pillars collapsed, sinkholes allowed O₂-rich air to enter. Coal-bed fires sporadically ignite in those sinkholes, producing many of the coal fires in this region. The 4.7-ha Hotchkiss fire exhibited limited surface expression during the field campaign, and is constrained to a few active vents.

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![Fig. 1. Maps indicating location of study area and annotated aerial photograph noting locations of the coal fires and other major features.](image-url)
Conversely, the 1.7-ha Welch Ranch, like the Ankney fire, exhibited a chain of active vents and unvegetated patches and linear-to-arcuate fractures that vent the coal-fire gases. While there are barren areas associated with the Welch Ranch fire, much of the surface is vegetated and is used for livestock grazing. In September 2002, Wyoming’s Abandoned Mine Lands Division attempted to extinguish the Welch Ranch by injecting a sandy slurry into fissures along the north edge of the fire and bulldozing the fissures shut (Murja, 2002; Heffern et al., 2003). This intervention slowed the advance but did not extinguish the fire, which continues to burn. A second round of reclamation is being considered.

2.1. Coal quality

Fresh coal samples were not accessible at any of the coal fire sites, so published coal quality data from the region were used. Averages of all samples for the Acme 7 1/2-minute quadrangle, the location of all of our study sites, indicate that the coals generally have high moisture and low heating values, typical of subbituminous coals (Table 1). The major oxides and minor element concentrations and the generally low sulfur content of the coal samples have implications for the species of neoformed minerals precipitated at the vents.
3. Methods

The field procedure employed three teams: two on the ground responsible for diffuse CO₂ flux determination (Section 3.1), gas flow and chemistry (Section 3.2) and mineral and tar collection (Section 3.3); and one team in an airplane (Section 3.4). In addition, coal consumption estimates were calculated at the Ankney fire using the apparent rate of fire advancement based on multiple BLM land surveys (Fig. 3).

3.1. Diffuse CO₂ emissions

Point measurements of diffuse fluxes of CO₂ were performed using standard accumulation chamber techniques (Bergfeld et al., 2006) and a USGS-designed flux system described in Engle et al. (2011). The method consists of measuring the rate of CO₂ accumulation in the chamber (∂C/∂t) after the chamber is sealed to the ground surface; the soil flux (F) of CO₂ (g/m²d) is calculated as follows:

\[ F = \frac{\rho V \partial C}{A \partial t} \]  

(1)

where \( \rho \) is gas density, \( V \) is the chamber volume, \( A \) is the footprint of the flux chamber, and \( \partial C/\partial t \) is the CO₂ accumulation rate in the chamber. To examine for repeatability and small-scale heterogeneity, diffuse soil fluxes were measured at three locations within a 1-m radius at roughly 10% of the points (n = 45). Relative standard deviations from the three measurements ranged from 16 to 132% with a median of 48%. At each coal CO₂ flux point, soil temperature was measured at the surface using an infrared (IR) thermometer and at 10-cm depth using a K-type thermocouple immediately before or after each flux measurement.

Soil CO₂ flux points for the Welch Ranch and Hotchkiss coal fires were arranged in a N-S/E-W grid with additional measurement sites added in and around thermal anomalies within the grid (Fig. 4). For the Ankney fire, safety and access issues prevented layout of a grid, so CO₂ flux points were placed as equidistant as possible (Fig. 4). To determine total diffuse CO₂ emissions for each study area, the CO₂ flux data were interpolated using sequential Gaussian simulation (SGS). For interpolation calculations, negative fluxes (these can result from input of soil gas with a lower CO₂ concentration than that of the air originally in the chamber) and ∂C/∂t measurements not significantly different than zero (p < 0.05) were assigned a value equal to the method detection limit. The application of SGS to diffuse CO₂ emissions from coal fires is detailed in Engle et al. (in press). Briefly, a semivariogram model is developed for each fire based on Box–Cox transformed CO₂ flux data. This model is then used to generate 100 realizations of diffuse CO₂ flux for each fire, which are back-transformed into the original units. Integrating CO₂ fluxes across each site generates a histogram of total CO₂ emissions for that fire. The SGS modeling, variogram modeling, and integration were completed using SGeMS 2.1 (Remy et al., 2009).

Table 1
As-received coal quality and chemistry data for unweathered coal samples from the Acme 7 1/2-minute quadrangle. Mois = moisture; VM = volatile matter; FC = fixed carbon. Data taken from Bragg et al. (1997) and unpublished data from the U.S. Bureau of Mines BMALYT dataset.

| Heating value | Mois | VM | FC | Ash | H | C | N | O | S_total | Hg |
|---------------|------|----|----|-----|---|---|---|---|---------|----|
| BTU/MJ/kg     | %    | %  | %  | %   | % | % | % | % | %       | mg/kg |
| Avg.          | 9780 | 22.8 | 20.5 | 32.7 | 42.4 | 4.3 | 6.2 | 56.1 | 1.07 | 31.1 | 0.47 | 0.09 |
| Std. dev.     | 265  | 0.62 | 1.7 | 1.8 | 2.9 | 0.9 | 0.1 | 1.0  | 0.06 | 0.5  | 0.21 | 0.05 |
| Max.          | 10,100 | 23.5 | 23.4 | 36.8 | 47.2 | 6.0 | 6.2 | 56.3 | 1.10 | 31.4 | 1.16 | 0.20 |
| Min.          | 9130 | 21.2 | 18.1 | 30.3 | 35.9 | 3.11 | 6.1 | 55.0 | 1.00 | 30.5 | 0.25 | 0.04 |
| Number        | 17   | 17  | 17  | 17  | 17  | 3   | 3   | 3    | 3     | 17   | 9    |
measurement of background CO₂ emissions was not possible, it was estimated from median CO₂ flux for the lowest subpopulation observed in quartile-normality plots. The background estimates for each fire were subtracted from SGS-calculated emissions.

### 3.2. Composition of coal-fire gases and calculation of vent emissions

Concentrations of gases in active exhaust vents were determined both in near-real time, using in situ analyzers, and post sampling.
from laboratory analyses of collected samples (Fig. 5). At each vent, concentration data for the individual compounds are combined with the corresponding gas flow data to determine vent emissions such that:

\[ E_i = C_i V A \]  

(2)

where \( E_i \) is the emission of gas component \( i \), \( C_i \) is the concentration of component \( i \) in the vent, \( V \) is the velocity of the gas perpendicular to the vent (assuming the velocity is equivalent for each gas component), and \( A \) is the cross-sectional area of the vent at the surface (O’Keefe et al., 2010). Vent-gas velocities were determined using an S-type Pitot tube in conjunction with a Flow Kinetics FKT 1 IDP1A-SV flow meter (O’Keefe et al., 2010). The range of velocities measured in the vents (0.5–4.26 m/s) is within the limitations of the flowmeter/Pitot tube setup (0.5–12.9 m/s). Often large vents (>100 cm²) were broken into smaller subsections, measured individually, and emissions were summed across the entire vent.

Vent-gases were analyzed in situ for CO₂, CO, CH₄, H₂S, and Hg. The first four constituents were measured using an Industrial Scientific MX6 iBrid portable gas analyzer. The gas analyzer was calibrated with zero and span standards before and after each field campaign; no marked drifts were observed. Measurement accuracy for the iBrid instrument ranges from ±2–10% depending on the gas and concentration range. Mercury concentrations were measured with a differential Zeeman atomic absorption spectrometry Ohio Lumex RX-915 Portable Mercury Analyzer. The Lumex analyzer was calibrated at least once a day using an internal calibration cell; measurement accuracy for the Lumex is ±5–20%, depending on the concentration range. Within each vent (or each section of a vent) in situ gas measurements were made at a minimum of five locations.

Volatile organic compound (VOC) gas samples were collected in electropolished, stainless steel, evacuated canisters. The samples were analyzed for a suite of carbon-bearing gases (i.e., CO, CO₂, carbonyl sulfide, dimethylsulfide, and carbon disulfide), C₁-to-C₉ aliphatic compounds, and single-ring aromatics (benzene, toluene, ethylbenzene, and xylene) on a gas chromatographic system in the Rowland-Blake Group laboratory at the University of California-Irvine. Analytical methods are described in Barletta et al. (2008) following quality assurance/quality control protocols of Colman et al. (2001). Previous use of these techniques to produce VOC analyses of coal-fire exhaust gases is provided in Stracher et al. (2006) and Hower et al. (2009).

3.3. Coal-fire minerals and tars

Minerals and tar frequently deposit on the mouth of coal-fire vents, and are formed through isochemical and mass-transfer processes (Stracher, 2007). Samples were collected from each fire; the minerals were identified using X-ray diffraction (XRD), and the tars were characterized using organic analyses. Powder XRD analyses of the mineral samples were performed at the University of Georgia with a Bruker D8 Advance diffractometer using co-radiation at 40 mA and 40 kV with a step size of 0.01° 2θ and a scan rate of 2.5° per minute, with a Lynx-eye® 192 position sensitive detector. The samples were dry mounted in aluminum holders and scanned at 8–60° 2θ with Cu Kα radiation.

Tar samples were analyzed using gas chromatography/flame ionization detection (GC/FID) at NewFields Environmental Forensics to measure C₉-C₄₄ hydrocarbons (Environmental Protection Agency (EPA) Method 8015) and gas chromatography/mass spectrometry (GC/MS) using a modification of EPA Method 8270C to identify Polycyclic Aromatic Hydrocarbons (PAHs) with 2 to 7 rings, including parent and alkylated isomers and geochemical biomarkers (Stout and Emsbo-Mattingly, 2008).

3.4. Remote sensing

Airborne remote sensing of the coal fires was conducted with a Cessna 337 Super Skymaster (owned and operated by Airborne Research Consultants, Saunderstown, Rhode Island) equipped with a FLIR™ A320 camera, a non-contact thermal device operating in the spectral range of 7.5 to 13 μm, mounted under the aircraft wing for
the pre-dawn flights. The camera has an instantaneous field of view (FOV) of 25° and provides 16-bit images at an accuracy of ±0.2 °C or ±2%. The altitude of the aircraft was maintained at ~2900 m above mean sea level (MSL) compared with the ground elevation for the fire sites of ~1130 m MSL, yielding a pixel size of about 2.5 × 2.5 m. The FLIR™ software directly recorded surface kinetic temperature images by using modified MODTRAN code to perform atmospheric correction of the thermal data, based on local atmospheric conditions such as the target temperature, air temperature, and air density.

Data processing of the IR imagery to estimate energy emissions from the fire was focused on decomposing the data based on the dominant Gaussian features. An automated program was developed to detect fire-related thermal anomalies and determine corresponding boundaries for calculating the total energy for each fire. First, the program computes a temperature frequency distribution for the IR image which, in turn, is broken down by each local minimum as function of temperature. A Gaussian distribution is fit to each segment, giving standard deviation and other statistical properties. The segment with highest frequency amplitude is assumed to be the land segment. The second highest frequency distribution is assumed to be surface water, e.g. the Tongue River, if the standard deviation of that segment is significantly smaller, i.e. thermally monotonous, than that for the land segment. The remaining segments are assumed to be fire anomalies and are integrated to give the total fire energy emission. Each segment is assigned a numerical identity corresponding to land, water, or fire anomaly, allowing heat flux to be calculated from data for individual frames. The CO2 emissions were estimated from the heat emission data through application of the Stefan–Boltzman law for gray bodies (Engle et al., 2011); and the heating value and carbon content of the coals in the region, taken from published literature (Table 1). Estimation of the coal-consumption rate allows for calculation of CO2 emissions (Engle et al., 2011).

4. Results

4.1. Diffuse CO2 emissions

A total of 153 diffuse CO2 flux measurements were made at 119 sampling points across the three coal fires. Fig. 4 shows the location of the sampling points on maps that show nodal means from the 100 SGSs of the CO2 flux data for the three fires. Individual CO2 fluxes ranged from 34 g CO2/m²/d (negative fluxes can result from input of soil gas with a lower CO2 concentration than that of the air in the chamber at the start of the measurement). Multiple comparisons after a Kruskal–Wallis test indicate that median diffuse CO2 fluxes were significantly greater (p < 0.05) for the Hotchkiss coal fire (median = 71.2 g CO2/m²/d) relative to the Welch Ranch (median = 25.2 g CO2/m²/d) and Ankney (median = 10.3 g CO2/m²/d) coal fires. However, maximum diffuse CO2 fluxes were much greater at the Welch Ranch and Ankney coal fires relative to the Hotchkiss coal fire (29,500, 2460, and 757 g CO2/m²/d, respectively) indicating that the flux data from the former fires exhibit more skewed distributions (Fig. 6). For comparison, median background fluxes were 8.3, 4.4, and 1.7 g CO2/m²/d for the Hotchkiss, Welch Ranch, and Ankney coal fires, respectively; these estimates of background fluxes are typical for expected soil CO2 fluxes given the climatic conditions of the Powder River Basin (< 10 g CO2/m²/d; Raich and Potter, 1995). Correlations between soil temperature (at 10-cm depth) and diffuse CO2 fluxes are positive, varied (Spearman p = 0.69–0.90), and statistically significant for all three fires (p < 0.001). Significant relationships were also found between soil temperatures and diffuse gas fluxes for coal fires in Australia and Alabama (Carras et al., 2009; Engle et al., in press). Integration of the SGS results provides background-corrected diffuse CO2 emission estimates of 4.0 ± 0.48, 1.2 ± 0.2, and 0.91 ± 0.24 t/d for the Hotchkiss, Welch Ranch, and Ankney fires, respectively. The estimate for Ankney is considered a minimum value given the scarcity of data for active portions of the fire.

4.2. Composition of coal-fire gases and vent emission rates

In situ measurements of the composition of coal fire vent gases and emission rates were conducted at 12 vents at Welch Ranch and nine vents at Ankney (Tables 2, 3; Fig. 4). Additional vents at the Ankney fire were observed but were not sampled due to safety considerations. In situ gas chemistry was determined at the eight vents at the Hotchkiss fire (Table 2), but problems with velocity measurements prevented calculation of emission rates. Carbon dioxide was the dominant gas measured in the exhaust from the three fires. Although the highest exhaust gas temperatures were observed in vents at the Ankney fire (up to 319 °C), higher CO2 concentrations (up to 3.06%) were measured at the other two fires. Post Kruskal–Wallis multiple comparison tests show that median CO2 concentrations were greater (at p < 0.01) in exhaust from the Hotchkiss fire than for the other fires. Carbon monoxide, CH4, and H2S concentrations were detectable at roughly half of the vents, indicating large spatial variability in combustion behavior. Mercury concentrations in gas vents ranged by nearly four orders of magnitude from 15 ng/m³ (roughly ten times ambient air) to concentrations more typical of fumarolic vents (up to 12,100 ng/m³; cf. Engle et al., 2006). The large range in Hg concentrations in vent gas can be attributed to the heterogeneity
of Hg concentrations in the coal (Table 1), relatively low concentrations of Hg in ambient air, and large ranges in temperature across the fires (Table 2). Multiple comparisons after a Kruskal–Wallis test indicate that median Hg concentrations were higher in exhaust gases at the Ankney fire than for the other fires (p < 0.01).

Vents at the Welch Ranch fire were emitting about 3.8 t CO2/d and 130 mg Hg/d; roughly 50% of the total emission for both gases was emanating from the largest vent (Vent 1 area = 1.6 m2). Notably, CO2 emissions were greater for the Ankney fire than for the Welch Ranch and Ankney sites. Mercury concentrations in the coal (Table 1), relatively low concentrations of Hg in ambient air, and large ranges in temperature across the fires (Table 2). Multiple comparisons after a Kruskal–Wallis test indicate that median Hg concentrations were higher in exhaust gases at the Ankney fire than for the other fires (p < 0.01).

Due to the incomplete coverage of presumably high emission sites at the Ankney fire, discussions of average emission rates and comparisons of the Ankney and Welch fires are negatively biased. Given that context, total vent CO2 emissions from Welch Ranch were about three times higher than those at Ankney. However, most of the measured parameters, including vent temperatures (up to 319 °C), gas concentrations, and vent emissions of CH4, H2S, and Hg were higher for the Ankney fire than for the Welch Ranch fire (Tables 2, 3). Mercury concentrations in Ankney vents (up to 12,100 ng/m3) were particularly elevated and emissions from a single vent of up to 250 mg/d demonstrated that coal fires may be an important source of atmospheric Hg.

| Notes: Exhaust gases in vents with a “-2” suffix were remeasured on May 14, 2007 (1 day after the original survey), due to incomplete Hg data for the original survey. |
lower abundances of aliphatics, and benzene concentrations below 0.007 ppmv, suggesting more complete combustion of the source coal for these vents.

### 4.3. Composition of minerals and tars

Only one of the 2009 samples (collected near Welch Vent 12-B) contained high-temperature minerals in a quantifiable amount. The sample was a white crust recovered from a red, altered soil adjacent to the vent. X-ray diffraction analysis identified the presence of potentially coal-fire related minerals: cordierite ([Mg,F e]_2[Al,Si]_2O_9), osmilitite ([K,Na]_2[Al,Fe]_2[Si,Al]_12O_30·H_2O), and, possibly, cristobalite (higher temperature SiO_2 polymorph). Minerals commonly adjacent to the vent. X-ray diffraction analysis identified high-temperature minerals in a quantifiable amount.

Additional details about the mineralogy of coal-fire related minerals can be found elsewhere (Pone et al., 2007; Stracher, 2007; Silva et al., in press).

In 2009, eight samples of tar were collected from the mouth of vents at the Hotchkiss and Welch Ranch fires. The concentration of total extractable petroleum hydrocarbons (TEH) eluting between n-nonane (n-C_9) and n-tetratetracontane (n-C_44) ranged from 54,600 to 216,000 mg/kg in four Hotchkiss samples and 89,500 to 1,000,000 mg/kg in the four Welch samples (Table 6). Most of this mass consisted of large hydrocarbons and carbon molecules with masses in excess of n-C_44 (e.g., soot). The sum of the 16 EPA priority pollutant PAH concentrations ranged from 31.5 to 157 mg/kg in the samples from the Hotchkiss fire and 53.6 to 438 mg/kg in the samples from the Welch Ranch fire. These results indicate that the Hotchkiss fire generated more water hydrocarbon and PAH concentrations compared with the Welch Ranch fire, possibly as a consequence of more complete combustion.

High proportions of parent compounds relative to alkylated PAHs indicate that PAHs in the tars were pyrogenic in origin (Yunker et al., 2002). Geochemical biomarkers (i.e., sesquiterpanes, triterpanes, steranes, and triaromatic steranes) were not detected. The EPA priority PAHs generally comprised less than 0.060% of the TEH with elevated concentration in some samples from both the Hotchkiss (up to 0.074%) and Welch Ranch fires (up to 0.154%) (Table 6). The source ratio of fluoranthene relative to pyrene (FL0/Py0) ranged from 0.81 to 1.04 with little to no distinction in samples between the two fires (Table 6). These ratios of FL0/Py0 are similar to those for industrial tars carbonized from petroleum (Stout and Embsmo- Mattingly, 2008). Once exposed to the surface environment, lighter PAHs weather more rapidly than heavier PAHs; this relationship is quantified using the Pyrogenic Weathering Index (PWI) (Uller and Embsmo-Mattingly, 2006). The estimated PWI for the samples indicates that the coal tars are slightly to moderately weathered (Table 6) and it is evidence the PAHs may be mobile in the environment.

The saturated aliphatic compounds in the samples represent coal distillates from the original subbituminous coal that avoided carbonization due to non-uniform heating during combustion (Fig. 7). Patterns of these saturated aliphatics exhibit several common features. The concentrations of heavy normal alkanes with odd carbon numbers are markedly greater than the even carbon numbers. This pattern is indicative of a source of cuticular waxes from leaves and trees (Simoneit, 1984). Another common terrigenous feature is the high proportion of pristane (Pr) relative to phytane (Ph). By contrast, the variable proportions of isoprenoid hydrocarbons relative to normal alkanes likely indicate variation in the degree of biodegradation of the terrigenous organic matter during original coal deposition. The variable proportions of unidentified hydrocarbons (typically terpenoids) may also reflect the variety of petrogenic conditions or, perhaps, the rank of coals in the combustion zone.

### 4.4. Remote sensing applications to emission estimates

Airborne IR imagery collected over the same period as the ground-based measurements was used to estimate CO₂ emissions from the Ankney and Welch Ranch fires; results for the Welch Ranch fire were previously presented in Engle et al. (2011). As described in Section 3.4, the thermal data were reduced to a land signal, a water signal (constant at this scale), and a fire signal. The most intense fire signal at Ankney was confined to a relatively small, arcuate area, representing the fire front as of May 2009 (Fig. 8). For the 0.28 ha active fire area, the total heat emission was determined to be 0.68 ± 0.042 MW. For comparison, the Welch Ranch fire, described by Engle et al. (2011), was emitting 0.48 ± 0.20 MW of total heat over
an area of 0.78 ha. Normalized for the relative areas, Ankney, therefore, had a heat flux 3.9 times that of Welch Ranch. As such, the Ankney burn was considerably more vigorous than that of the Welch Ranch fire. Extrapolating from the estimated amount of coal combusted per day at Welch Ranch and assuming complete conversion of carbon to CO$_2$ (examining dominant carbon-bearing gases in vent measurements [CO$_2$, CO, and CH$_4$], 94% of the carbon measured in vents was emitted as CO$_2$), and that the heating value of the coal is 5.144 kJ/kg, the estimated combustion heat flux indicates an estimated combustion of 6.4–13.8 t coal/d and 12.9–27.1 t CO$_2$/d.

4.5. Rate of fire advancement to estimate CO$_2$ emissions

Data on the rate of fire advancement, i.e., coal consumption, for the Ankney fire allowed for a gross method to estimate CO$_2$ emissions. The maximum extent of the fire was mapped during several campaigns from May 2004 to September 2011; over that period the surface disturbed by the Ankney fire grew from 0.6 to 1.4 ha (Fig. 3). There are no drill holes adjacent to the fire, but the well log for a nearby coal bed methane well (0.4-km away) shows an 8 m thick coal bed at about the same elevation as the burning coal bed. Based on the increase in the area burned and assuming the 8 m coal thickness, about 80,800 tons of coal would have been burned, equal to 30 t/d over the previous 7 years. This is 3- to 6-times higher than the estimated combustion rate calculated for the Ankney fire in May 2009 based on the airborne IR data, assuming that the entire thickness of the coal bed burned.

5. Discussion

Comparison of gas chemistry, gas flow, and tar composition indicates that obvious differences exist both across individual fires and

### Table 4

| Common name     | Formula | Concentration* | Hotchiss | Welch Ranch | Ankney |
|-----------------|---------|----------------|----------|-------------|--------|
| Carbon dioxide  | CO$_2$  | ppmv           | 22,400   | 40,400      | 5780   |
| Carbon monoxide | CO      | ppbv           | 102      | 236         | 5780   |
| Carbon sulfide  | OCS     | pptv           | 725      | 4900        | 12,700 |
| Dimethyl sulfide| (CH$_3$)$_2$S | pptv | < dl        | < dl       | 157    |
| Carbon disulfide| CS$_2$  | pptv           | < dl     | 111         | 225    |

### Aliphatic Compounds

| Compound        | Concentration* | Sample color | X-ray identification | Depositional substrate of minerals |
|-----------------|----------------|--------------|----------------------|-----------------------------------|
| Ethane          | CH$_4$         | ppmv         | 610                  | White | Gypsum, sulfur (?) | Red cinder* |
| Ethene          | C$_2$H$_6$     | pptv         | 29,700               | Yellow | Methane, quartz | Red cinder |
| Propane         | C$_3$H$_8$     | pptv         | 314                  | Red    | Gypsum, quartz | Red cinder |
| Propene         | C$_4$H$_8$     | pptv         | 205                  | Yellow | Methane, quartz | Red cinder |
| Butane          | C$_5$H$_10$    | pptv         | 37,600               | White | Gypsum, sulfur (?) | Red cinder |
| Butene          | C$_6$H$_12$    | pptv         | 316                  | Red    | Methane, quartz | Red cinder |
| Pentene         | C$_7$H$_12$    | pptv         | 268                  | Yellow | Gypsum, sulfur (?) | Red cinder |
| Methylpentene   | C$_8$H$_12$    | pptv         | 28,100               | Red    | Gypsum, quartz | Red cinder |
| Hexene          | C$_9$H$_12$    | pptv         | 152                  | Yellow | Methane, quartz | Red cinder |
| Methylhexene    | C$_10$H$_12$   | pptv         | 4730                 | Red    | Gypsum, sulfur (?) | Red cinder |
| Heptene         | C$_11$H$_12$   | pptv         | 2970                 | Yellow | Methane, quartz | Red cinder |
| Methylheptene   | C$_12$H$_12$   | pptv         | 660                  | Red    | Gypsum, sulfur (?) | Red cinder |
| Octane          | C$_13$H$_12$   | pptv         | 1010                 | Yellow | Methane, quartz | Red cinder |
| Methyloctane    | C$_14$H$_12$   | pptv         | 219                  | Red    | Gypsum, sulfur (?) | Red cinder |
| Nonane          | C$_15$H$_12$   | pptv         | 660                  | Yellow | Methane, quartz | Red cinder |
| Methylnonane    | C$_16$H$_12$   | pptv         | 1010                 | Red    | Gypsum, sulfur (?) | Red cinder |

* ppmv = 1 part in 10$^6$; ppbv = 1 part in 10$^9$; pptv = 1 part 10$^{12}$.

### Table 5

| Coal-fire related minerals collected from the Welch Ranch fire by G.B. Stracher during an earlier trip in 2004. With the exception of the sulfur, inferred to be present based on appearance, all minerals were identified by X-ray diffraction. |
|---|---|---|---|---|
| Latitude | Longitude | Collection area | Sample color | Mineral X-ray identification | Depositional substrate of minerals (* ) |
| 44°56' | 106°57' | 24°N | hot spot | White | Gypsum, sulfur | Red cinder |
| 44°56' | 106°57' | 23°N | Gas vent | White | Tschermagite, quartz | Red cinder |
| 44°56' | 106°57' | 23°N | Gas vent | White | Cristobalite, quartz | Red cinder |

* Red cinder is primarily composed of baked shale.
among the three fires. In the case of the former, the most striking example is that of the Welch Ranch Fire where the vents align along a linear northwest–southeast trend (Fig. 4B) with a corresponding decrease in surface elevation towards the Tongue River (Fig. 9). Carbon monoxide, H2S, and CH4 concentrations increase from below detection limits at the upland vents (1–6) to elevated levels in the

Table 6
Organic composition of tar samples collected from vents at the Hotchkiss and Welch Ranch coal fires. Degree of pyrogenic weathering defined via: Light — NO>P0; Moderate — N0–P0>PY0.

| Compound | Hotchkiss coal fire | Welch Ranch coal fire |
|----------|---------------------|-----------------------|
| Source:  | Vent 1 | Vent 5 | Vent 7 | Vent 8 | Vent 1 | Vent 3 | Vent 6 | Vent 7 |
| Total Extractable Hydrocarbons (TEH; C9-C44) mg/kg | 216,000 | 205,000 | 108,000 | 54,600 | 124,000 | 85,500 | 1,000,000 | 901,000 |
| EPA PAHs: | | | | | | | | |
| Naphtalene (N0) mg/kg | 9.23 | 18.8 | 4.19 | 3.88 | 5.60 | 24.8 | 101 | 44.7 |
| Acenaphthylene (AY) mg/kg | 2.56 | 5.51 | 0.58 | 1.08 | 0.39 | 0.88 | 28.6 | 12.6 |
| Acenaphthene (AE) mg/kg | 0.87 | <4.30 | 0.58 | 0.38 | 0.73 | 0.99 | 15.4 | <6.50 |
| Fluorene (Fi) mg/kg | 3.34 | 8.78 | 1.55 | 0.91 | 1.14 | 2.05 | 27.3 | 18.4 |
| Phenanthrene (P0) mg/kg | 8.46 | 21.5 | 5.02 | 4.65 | 3.48 | 9.74 | 72.3 | 78.2 |
| Anthracene (A0) mg/kg | 2.27 | 5.25 | 0.69 | 1.04 | <0.25 | 0.96 | 14.6 | 13.4 |
| Fluoranthenone (FLO) mg/kg | 3.49 | 13.1 | 3.64 | 2.50 | 1.25 | 1.6 | 13.7 | 32.8 |
| Pyrene (Py) mg/kg | 4.20 | 13.3 | 3.80 | 2.65 | 1.26 | 1.54 | 16.9 | 31.6 |
| Benzo[a]anthracene (BA0) mg/kg | 2.64 | 6.20 | 1.47 | 1.16 | 0.27 | 0.72 | 13.1 | 8.59 |
| Chrysene (CY) mg/kg | 3.62 | 12.7 | 2.59 | 2.22 | 0.73 | 1.86 | 21.5 | 28.1 |
| Benzo[b]fluoranthenone (BBF) mg/kg | 3.09 | 9.05 | 2.34 | 2.15 | 0.63 | 0.92 | 18.5 | 11.5 |
| Benzo[k]fluoranthenone (BJKF) mg/kg | 3.71 | 7.22 | 1.52 | 1.85 | 0.60 | 0.86 | 22.6 | 9.54 |
| Benzo[a]pyrene (BAP) mg/kg | 3.64 | 6.10 | 1.31 | 1.97 | 2.08 | 0.88 | 0.66 | 27.9 | 15.4 |
| Indeno[1,2,3-cd]pyrene (IND) mg/kg | 4.59 | 8.64 | 1.63 | 1.92 | 0.53 | 1.05 | 19.0 | 10.8 |
| Dibenzo[a]anthracene (DA) mg/kg | 6.46 | 6.28 | 0.90 | 1.20 | 0.21 | 0.45 | 5.94 | 3.84 |
| Benzo[g,h,i]perylene (GHI) mg/kg | 4.38 | 7.45 | 2.02 | 1.86 | 0.60 | 1.05 | 19.6 | 10.3 |
| Sum of EPA PAHs: mg/kg | 66.7 | 152–157 | 34.5 | 31.5 | 53.6–53.8 | 138 | 438 | 330–336 |
| EPA PAHs/TEH % | 0.031 | 0.074–0.076 | 0.032 | 0.058 | 0.043 | 0.154 | 0.038 | 0.037–0.038 |
| FL0/PY0 Unitless | 0.83 | 0.98 | 0.96 | 0.94 | 0.99 | 1.04 | 0.81 | 1.04 |
| (BBF+BJKF)/BAP Unitless | 1.87 | 2.78 | 1.96 | 1.52 | 1.40 | 2.68 | 1.47 | 1.37 |
| Degree of pyrogenic weathering (PWI) | Light | Moderate | Moderate | Moderate | Moderate | Moderate | Light | Moderate |

Fig. 7. GC-FID chromatograms of total extractable petroleum hydrocarbons from tar samples collected from vents at the Hotchkiss and Welch Ranch coal fires. Some peaks left unidentified due to lack of confidence in matches with compounds in the data library. Pr = pristane; Ph = phytane.
vents at lower elevation (Table 2). Similarly, concentrations of aliphatic and aromatic compounds in samples of exhaust gas are considerably higher, by at least one to two orders of magnitude, at vent 10 vs. vent 1. Tars collected from vents 6 and 7 contain substantially higher concentrations of TEH relative to those from vents 1 and 2 (Table 6) and also show a notable increase in unresolved compounds in GC-FID chromatograms (i.e., a development of a positive mound in carbon numbers 20–40 (Fig. 7). All of these independent lines of evidence suggest that a spatial variation in combustion completeness is present at the Welch Ranch fire during the sampling campaign. Using the CO2/CO ratio as an indicator for combustion efficiency was also investigated, but Mann–Whitney U tests indicate no significant difference in exhaust gas temperature between the vents 1–7 and vents 8–12 (p > 0.05).

We propose two possible explanations for the variance in vent gas and tar composition along the elevation trend. The first explanation pertains to the fact that there are at least two coals burning at the Welch Ranch site (Fig. 9). Higher concentrations of rapidly depositing species, e.g., H2S, in the lower elevation vents suggest that the lower coalbed, the Dietz 3 coal, has a fairly direct conduit to the vents on the slope closer to the Tongue River. The burning of the Dietz 3 coal may be heating the upper coal, the Dietz 2 coal, dewatering and otherwise, to some extent, devolatilizing the coal. In this case, CO2 in vents 1–6 could be derived either from combustion of the upper coal or advection and diffusion through the upper coal of gas emitted from the lower coal. Ultimately, the heating and drying, possibly augmented by repeated wetting and drying cycles, of the upper (Dietz 2) coal would result in its combustion, but at a burn front offset from the lower (Dietz 3) coal. A second explanation for the observed variation in combustion efficiency is that more complete combustion occurs on the upwind, upslope portion of the site, and efficiency decreases toward the downwind portion of the site, closer to the river. Assuming that the dominant subsurface direction of airflow in the fire also moves in the same direction, lower combustion efficiency in the lower elevation vents (i.e., vents 9–12) may be a result of upgradient oxygen depletion leading to O2-poor conditions near the end of a subsurface flowpath.

The Hotchkiss, Welch Ranch, and Ankney coal fires emit large quantities of CO2; volatile toxic, mutagenic, and carcinogenic organic species; and Hg. We have noted for coal fires elsewhere (Hower et al., 2009; O’Keefe et al., 2010), that combustion is less complete compared with the combustion that occurs in coal-fired utility power plants. However, unlike the power plant environment, the emissions from a coal fire are uncontrolled. Additionally, Hg concentrations in vents at the Ankney fire suggest that coal fires may be an important source of atmospheric Hg, at least locally, especially when considering the number of coal fires (thousands) thought to exist internationally.

As more coal-fire emission data are generated, comparison of emissions and fluxes should provide insight into natural, spatial, and temporal variability and data scalability. The CO2 emission estimates from airborne remote sensing and estimation of fire advancement.
methods indicate that the Ankeny fire emits up to 80 t/d, roughly 1.5 to 20 times more CO₂ than either of the other two fires (Table 7). For comparison, a 514-MW coal-fired boiler in Eastern Kentucky produces >1000 t CO₂/d and roughly 100 g Hg/d (Center for Global Development, 2007; O'Keefe et al., 2010). Normalizing the emissions to the area of each fire, i.e. flux, there is a nearly a 3-order of magnitude variation in CO₂ fluxes between sites, despite their close proximity and the similarity in the coal composition (Table 8). These differences highlight variations in the rate at which the different fires consume coal. Our flux data indicate that the Ankeny fire was consuming coal ~10–100 times faster than Welch Ranch fire and ~25–750 times faster than the Hotchkiss fire. These results also generally follow the difference in temperatures measured at the fires; the hottest fire, i.e. the fire consuming the most coal, produces the largest flux of gases. By comparison, a gob pile fire near Mulga, Alabama, exhibited a CO₂ flux on the low end of estimates for the Ankeny fire. Given surface features, temperatures, and the estimated fire intensity, our estimates of total CO₂ flux for the fires correspond well with average CO₂ equivalent fluxes for coal mine spoil piles in Australia under varying degrees of combustion (Carras et al., 2009). The flux estimates compiled in Table 8 may serve as a first step in providing a range of possible values for coal-fire CO₂ emissions for national and international inventories.

One major caveat to the potential extrapolation of coal-fire emission estimates is the impact of temporal variability. As discussed previously in this paper, vent CO₂ emissions at the Welch Ranch fire decreased by ~50% in measurement sets collected within 48 h. Finer resolution time series data presented by Hower et al. (2011) demonstrate that the concentrations of coal-fire gases vary substantially within daily and hourly time scales. The impact of temporal variability in fires has important implications for scaling emission estimates to longer time scales and is currently being investigated further.

The composition and potential mobility of the tar samples and the compounds that comprise them show that gaseous emissions are not the only potential mechanism for release of coal-fire contaminants into the environment. Querol et al. (2011) examined the leachability of inorganic elements from coal-fire tars and minerals and found that freshly formed samples produced acid (pH 1.6–4) solutions, capable of dissolving trace elements. However it is unclear how general these results are to other fires and environments. The availability and transport of organics from coal-fire tars are much less well known and require further study.

6. Summary

Ground-based surveys of three coal fires and airborne surveys of two of the fires in the Wyoming portion of the Powder River Basin were conducted in May 2009. The Hotchkiss fire has the lowest coal consumption rate per area (based on total CO₂ flux of 0.9–1.1 mg/s/m²) releasing roughly 4.0 t CO₂/d through diffuse soil emissions. Hotchkiss vent gases contained the highest CO₂ concentrations of any vent in the study (3.06%) and the lowest Hg concentrations (mean = 58 ng/m³). The composition of exhaust gas and tar chemistry at the Welch Ranch fire varied along a transect parallel to the vents. Results show that combustion efficiency, i.e. higher CO₂/CO and lower relative abundance of aromatics, is greater in gases in the high elevation vents and decreases with elevation, closer to the river. We suggest the following explanations for this trend: 1) chemical variations between the two burning coal beds; and 2) consumption and decrease in O₂ along the subterranean flow path. The estimated total CO₂ flux for the fire (1.9–6.0 mg/s/m²) was similar to that of non-venting coal mine spoil piles in Australia. The Ankeny fire burns more intensely than the two other fires (CO₂ flux = 25–780 mg/s/m²). Ground-based measurements over portions of the fire indicate Hg concentrations in vents exceeded background ambient air concentrations by up to ~4 orders of magnitude (12,100 ng/m³) and benzene concentrations ranged up to 3.81 ppmv.

The emissions and concentrations of CO, CO₂, volatile gases, such as benzene, and volatile metal species, primarily Hg, are at levels that may be of potential human health and environmental concern near all three of the fires. The amount of coal being consumed by the fires is less than at a coal-fired power plant, but, in contrast to the latter setting, the natural coal fires do not necessarily represent complete combustion and the emissions from natural fires are uncontrolled. While sizable strides on investigating the products of coal fires have occurred in the last several years, more research is required to adequately assess and understand the role of coal fires with respect to emission inventories and human and environmental health.

Table 7
Comparison of CO₂ emission estimates for the Hotchkiss, Welch Ranch, and Ankeny coal fires. ND = not determined.

| Fire            | Ground-based data | Airborne data | Rate of fire advancement | Data source |
|-----------------|-------------------|---------------|--------------------------|-------------|
|                 | Diffuse t CO₂/day | Vent t CO₂/day | Total t CO₂/day | total t CO₂/day | total t CO₂/day | Notes | Source |
| Hotchkiss       | 4.0±0.48          | ND            | >3.5–4.5               | 3.5–4.1     | ND            | Mine fire | Subbituminous | This study |
| Welch Ranch     | 1.2±0.2           | 6.7±0.9       | 6.8–9.0               | ND          | ND            | Mine fire | Subbituminous | Engle et al. (2011) |
| Ankeny          | >0.91±0.24        | 1.6±0.09      | >2.2–2.8              | 12.9–27.1   | 36–80         | Outcrop fire | Subbituminous | This study |

Table 8
Summary and comparison of total CO₂ fluxes for coal fires in the United States and Australia.

| Fire            | Area (ha) | Total CO₂ flux (mg/s/m²) | Estimate type | Notes | Coal rank | Source |
|-----------------|-----------|--------------------------|---------------|-------|-----------|--------|
| Hotchkiss, USA  | 4.7       | 0.90–1.1                 | Diffuse only  | Mine fire | Subbituminous | This study |
| Welch Ranch, USA| 1.7       | 1.9–6.0                  | Airborne and ground-based | Mine fire | Subbituminous | Engle et al. (2011) |
| Ankeny, USA     | 0.58      | 25–780                   | Airborne and rate of fire advance | Outcrop fire | Subbituminous | This study |
| North, USA      | 3.5       | 0.28–0.51               | Subsidence rate, chimney modeling, and ground-based | Outcrop fire | High volatile bituminous (?) | Ide and Orr (2011) |
| Mulga, USA      | 9.1       | 27–48                    | Diffuse only  | Gob pile fire | Low volatile bituminous | Engle et al. (2011) |
| Combustion fire | 9.1       | 260±290*                 | Diffuse only  | –      | Bituminous | Carras et al. (2009) |

* Sum of diffuse CO₂ and CH₄, in terms of equivalent CO₂ greenhouse impact.
Disclaimer

Equipment brand and trade names are for descriptive purpose only and do not imply endorsement by the U.S. Government.

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