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The Tiptop coal-mine fire, Kentucky: Preliminary investigation of the measurement of mercury and other hazardous gases from coal-fire gas vents

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ABSTRACT

The Tiptop underground coal-mine fire in the Skyline coalbed of the Middle Pennsylvanian Breathitt Formation was investigated in rural northern Breathitt County, Kentucky, in May 2008 and January 2009, for the purpose of determining the concentrations of carbon dioxide (CO_2), carbon monoxide (CO), and mercury (Hg) in the vent and for measuring gas-vent temperatures. At the time of our visits, concentrations of CO_2 peaked at 2.0% and >6.0% (v/v) and CO at 600 ppm and >700 ppm during field analysis in May 2008 and January 2009, respectively. For comparison, these concentrations exceed the U.S. Occupational Safety & Health Administration (OSHA) eight-hour safe exposure limits (0.5% CO_2 and 50 ppm CO), although the site is not currently mined. Mercury, as Hg^0, in excess of 500 and 2100 $\mu g/m^3$, in May and January, respectively, in the field, also exceeded the OSHA eight-hour exposure limit (50 $\mu g/m^3$). Carbonyl sulfide, dimethyl sulfide, carbon disulfide, and a suite of organic compounds were determined at two vents for the first sampling event. All gases are diluted by air as they exit and migrate away from a gas vent, but temperature inversions and other meteorological conditions could lead to unhealthy concentrations in the nearby towns.

Variation in gas temperatures, nearly 300 °C during the January visit to the fire versus <50 °C in May, demonstrates the large temporal variability in fire intensity at the Tiptop mine. These preliminary results suggest that emissions from coal fires may be important, but additional data are required that address the reasons for significant variations in the composition, flow, and temperature of vent gases.

1. Introduction

Coal fires consume coal reserves, pollute the environment, and emit potentially toxic gases. In addition, they often produce land subsidence due to volume-reduced coal during burning in the subsurface (Stracher and Taylor, 2004; Pone et al., 2007). At this time, we know of six active or recently active coal-fire sites in eastern Kentucky. Two, the Laura Campbell and Ruth Mullins[1] fires, were previously investigated by Stracher et al. (2008a,b). A third active site, the Highway 80 fire, also known as the Truman Shepherd fire, was investigated on 21 February 2009 and the results of this work will be presented elsewhere. To date, legal access to two other fires has not been obtained. The sixth, the Tiptop fire in northern Breathitt County, Kentucky, is the subject of this short communication. In addition to documenting the occurrence of this fire, another objective of the Tiptop investigation was to test the field measurement of mercury (Hg) and other gases in vent emissions from an active coal fire.

The Tiptop coal-mine fire[2] is located at 37° 35.911′N latitude and 83° 03.586′W longitude in the Tiptop 7.5′ geologic quadrangle (Danilchek, 1977), northern Breathitt County, Kentucky (Fig. 1). According to this map, the fire is the Middle Pennsylvanian, Breathitt Formation, Skyline coalbed. Underground mining dates to the 1930s. The cause and starting date of the Tiptop fire are unknown although, to the best of our knowledge, the start of the fire post-dates the underground mining activity. Gas from the fire exits from fractures in the sandstone overlying the underground mine and from vents in the collapsed roof overlying abandoned mine portals. Our sampling was at the latter sites. A view of the hillside and gas vents is shown in Fig. 2.

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[1] The Ruth Mullins fire was examined on the west side of the coal outcrop in March 2007. The outcrop was buried in an attempt to smother the fire prior to a visit in May 2007. We later examined the fire on the east side of the outcrop in September 2007 and in March, June, and July 2009. It is still active.

[2] Previous Kentucky mine fires investigated were named after the person first reporting the fire. We do not know of any formal name for the fire of this study, so it is convenient to name it after the nearby settlement of Tiptop, the namesake of the 7.5′ quadrangle.

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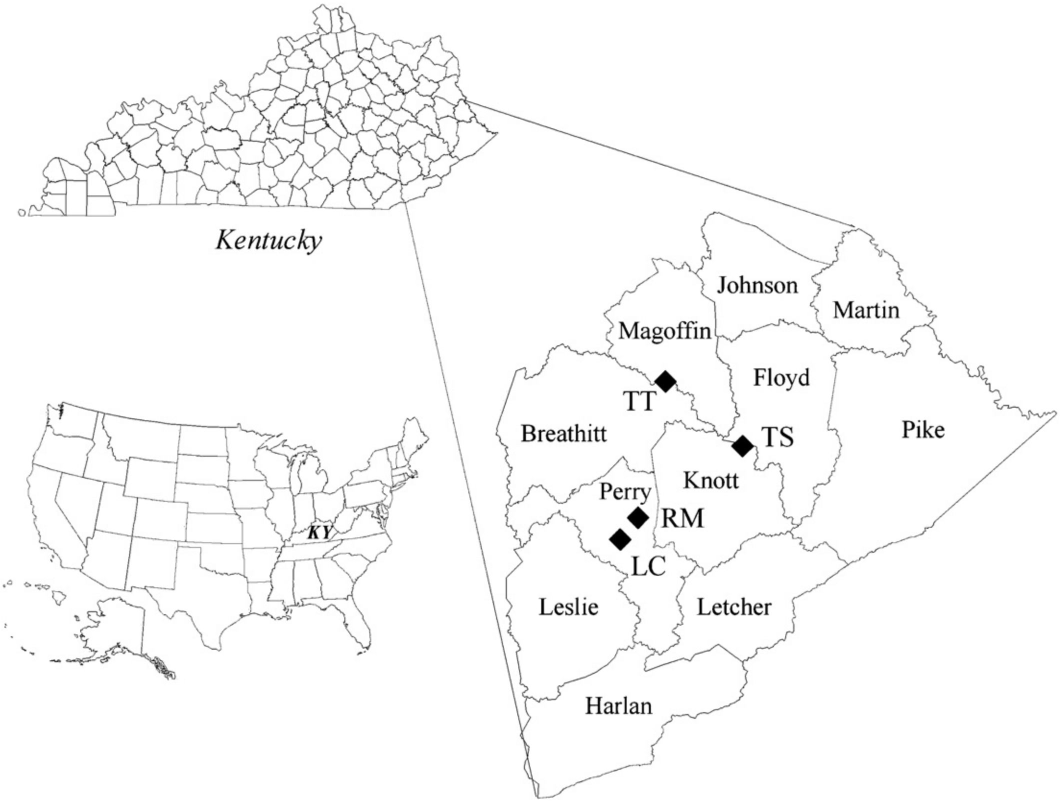

Fig. 1. Location of the Tiptop coal fire (TT). Locations of the Laura Campbell (LC), Ruth Mullins (RM), and Highway 80 (80) fires are also shown.

Based on a limited number of analyses (three samples each of the upper and lower splits, two samples of the middle split) of the Skyline coalbed in the Tiptop 7.5′ quadrangle, the high volatile A bituminous (0.77% R_{max}) coal has a moderate-ash (8.33% as determined) and low sulfur (0.75% as determined) content, with negligible Cl (0.01–0.02% as determined). The coalbed also has 64 µg/g As (ash basis) (5.3 µg/g (whole coal basis) and 0.05 µg/g Hg (whole coal basis).

2. Methods

Two visits were made to the Tiptop coal-mine fire: 7 May 2008 and 15 January 2009. Temperature was measured with a Vernier thermocouple probe linked to a TI-84 calculator. The point of temperature measurement was generally a few cm below the edge of the vent in order to avoid interferences from atmospheric air flow. The thermocouple was calibrated

Fig. 2. View of south-facing slope, about 30 m high, with several vents emitting smoke from the Tiptop coal fire in an abandoned underground mine. Co-author Kevin Henke is at left.

against a muffle furnace before each use in the field. Carbon dioxide (CO_2) concentrations were measured with a Dräger CH-23501 detection tube containing a chemical reagent calibrated for measuring 0.1–6.0 % (v/v) CO_2 within a temperature range of 0–30 °C. Carbon monoxide (CO) was measured with a Dräger CH-20601 detection tube containing a chemical reagent calibrated for measuring 10–3000 µg/m^3 CO within a temperature range of 0–50 °C.

Gaseous Hg^0 concentrations were measured using an Arizona Instruments Jerome 431-X Hg Vapor Analyzer. Mercury values reported herein are from gas samples collected within the mouth of active gas vents. The instrument utilizes a gold-film sensor to amalgamate gaseous Hg. The operating range extends to 40 °C, a limitation for analyzing most *in situ* coal-fire emissions, and the sensitivity range is 3 to 999 µg/m^3 Hg ($<$0.001 ppm to 0.12 ppm) with a $\pm$5% standard deviation at 100 µg/m^3 (Brookhaven National Laboratory, 2007). The instrument can be sensitive to interferences from acidic gases, so an acid gas scrubber was used in the sampling line. Because the instrument is only measuring Hg^0 concentrations, we are missing the data on other Hg forms, something we hope to address in future studies.

A Dräger CH-23101 detection tube was also used to measure Hg emissions. The detection tube is one of several methods recommended according to the US Occupational Safety & Health Administration's (OSHA) guidelines for the detection of Hg vapor (OSHA, 2004). The Dräger CH-23101 tube uses the $Hg + CuI \rightarrow Hg\text{–}Cu$ complex reaction to detect Hg. The reaction reagent in the tube has an upper limit of 0.24 ppm (2000 µg/m^3) with a $\pm$30% standard deviation and a maximum operating temperature of 40 °C. While halogens will interfere with the Dräger CH-23101 detection of Hg, there is no reported interference from arsine, phosphine, hydrogen sulfide, ammonia, nitrogen dioxide, sulfur dioxide, and hydrazine (Brookhaven National Laboratory, 2007).

On the 7 May 2008 visit, two gas samples were collected in electropolished, stainless steel, evacuated canisters: canister DE2486 for the east vent and canister DE2534 for the west vent. A stainless steel bellows valve was opened slightly to fill each canister to ambient pressure. Gas analyses for a full suite of simple carbon-bearing gases (i.e., CO, CO_2, carbonyl sulfide, dimethylsulfide, and carbon disulfide), aliphatic compounds (methane to nonane) and aromatics (benzene, toluene, ethylbenzene, and xylene) were performed on a gas chromatographic system in the Rowland-Blake Group laboratory at the University of California — Irvine. Quality assurance and quality control description are given in Colman et al. (2001) and more detailed analytical methodology is found in Barletta et al. (2008).

3. Results and discussion

Sampling at the Tiptop fire focused on two vents (west vent and east vent) emitting gas and particulate matter. Since coal-mine fires are dynamic, multiple data collecting expeditions to the fire are desirable where feasible. During the visit in May, the air temperature was about 20 °C and the gas vent temperatures in the two vents were 32 °C and 47 °C (Table 1). The temperature gradient during the January visit was more extreme with an air temperature of −8 °C and a maximum vent temperature of 291 °C. Smoke was observed exhausting from gas vents during the January visit (Fig. 2), but not during the May visit. One possibility is that the lower outside temperature could have accelerated the heat flow, enhancing the draw on the underground fire, increasing the vent temperature in January. It is also likely that the position of the fire front moved in the intervening eight months, possibly contributing to the difference in temperature.

In contrast to the Ruth Mullins fire, no condensed minerals or only minor amounts of coal tar were observed adjacent to gas vents during either visit to the Tiptop fire. The lack of condensates around the Tiptop vents may be due to the generally low temperatures (often around 40 °C). In contrast, condensates were more common around the vents of the hotter (400 °C) Ruth Mullins fire. This does not preclude the formation of minerals at higher temperatures in inaccessible coal-mine voids.

Table 1

Vent temperature, CO_2 (vol.%), CO (parts per million (ppm) [one part in 10^6 parts]), Hg (ppm), and Hg:C molar ratio for two vents for each of two visits.

Sampling date	Vent	T (°C)	CO_2 (vol.%)	CO (ppm)	Hg (µg/m^3)	Hg (µg/m^3)	Hg:C Molar ratio
			[Draeger CH-23501]	[Draeger CH-25601]	[Jerome 431-X]	[Draeger CH-23101]	
May-08	West	32	0.3	600	580		1.8E − 05
	East	47	0.3	500			1.9E − 05
			2.0	300	390		2.2E − 06
				400			2.2E − 06
Jan-09	West	40	0.5	100	<9		<2.0E − 07
	East	291	0.5	100			<2.0E − 07
			4.0	***700***	[1]	***>2100***	>5.8E − 06
			6.0	500			>3.9E − 06
			6.0				>3.9E − 06

[1] Vent temperature exceeded limits of instrument.

Bold italics — upper detection limit.

Multiple concentration per vent and event indicate replicate analyses.

During the January 2009 sampling event, CO_2 and CO concentrations as high as 6.0% (v/v) and 700 ppm (Table 1), respectively, occurred at the hotter east vent. Vent temperature during these measurements exceeded the recommended operating temperature-detection range for the Dräger tubes, casting suspicion on the validity of the values. Even at a 32 °C vent during the May expedition, CO concentrations reached a maximum of 600 ppm. The limited data from the Tiptop site suggest a correlation of CO and CO_2 concentrations with vent temperature, although further investigation, potentially following techniques of Singh et al. (2007), Xue et al. (2008), and Wessling et al. (2008), is necessary.

All of the CO_2 and CO concentrations measured in vents at the Tiptop fire far exceed the eight-hour OSHA limits of 5000 ppm (0.5 % (v/v)) CO_2 (NIOSH, 2005) and 50 ppm CO (OSHA, undated (a)). CO_2 and CO downwind of the vent would likely be diluted below hazardous levels, although dangerous conditions could exist during temperature inversions or other unfavorable meteorological conditions. Unlike the fires near Hazard, Kentucky, or the Mulga, Alabama, gob fire (Stracher et al., 2008b), the Tiptop fire is not located immediately adjacent to a populated area. With the exception of a haul road, the current surrounding surface-coal-mine workings are greater than 500 m from the vents.

Mercury concentrations, as Hg^0, in vent gases (measured with the Jerome 431-X instrument) were 390 and 580 µg/m^3 at the two vents in May. In January, the Hg concentration measured in the west vent was at the lower detection limit of the instrument (Jerome 431-X, $<$9 µg/m^3). In the east vent, the Hg concentration was measured as $>$2100 µg/m^3 (Dräger CH-23101 detection tube). The latter value is suspect for the same reason the Jerome 431-X could not record a value in that vent; the gas-emission temperature exceeded the upper temperature limit for reliable measurement. Variations in the Hg could be related to the gas temperature and the subsequent partitioning among Hg species as well as to differences in the coal burning at the two sampling times. The OSHA (undated (b)) ceiling limit for Hg exposure, which no one should be exposed to, is 100 µg/m^3 (0.011 ppm), with lower limits for an eight- or ten-hour exposure period. Therefore, the Hg values measured in May (and, if valid, the high January value) exceed the recommended exposure limit and are several orders of magnitude higher than background tropospheric Hg concentrations (~1.5 ng/m^3). Molar ratios of C:Hg in vent gas, where C is defined as the sum of C in CO_2 and CO, ranged almost two orders of magnitude from $<2.0\times10^{-7}$ to 1.9×10^{-5}

(Table 1). The significant range in these ratios, which are in the range exhibited in volcanic and geothermal systems (Engle et al., 2006) suggests that the composition of gases emitted from the Tiptop fire varies spatially and temporally.

The analytical techniques presented in this investigation are useful for providing preliminary Hg^0 concentration data for coal-fire gas vents. However, the two methods have several limitations that must be considered when gathering data at coal-fire gas vents. Mercury detection using the Jerome 431-X instrument can be impacted by interference from hydrogen sulfide and other acidic gases if acid traps are not used (Schroeder et al., 1985) and the analyzer exhibits questionable reliability at temperatures greater than 40 °C. The Dräger CH-23101 detection tubes can be affected by halogen gas interference, provide semi-quantitative output, and lack the ability to collect continuous measurements. Both methods have detection limits well above the ambient concentrations making Hg measurement in more dilute systems difficult (9 $\mu g/m^3$ for the Jerome 431-X instrument and 50 $\mu g/m^3$ for the Dräger CH-23101 detection tube).

For improved quantitative analysis of Hg concentrations in coal-fire gas, is recommended the use of either a Zeeman atomic absorption spectrometer (Witt et al., 2008a,b) or dual gold-trap amalgamation followed by cold vapor-atomic fluorescence (Aiuppa et al., 2007) is recommended. These methods were successfully used to measure Hg concentrations in geothermal and volcanic gases, which provide similar analytical issues (Engle et al., 2006).

Gas analyses (Table 2) show a wide range of volatile organic species and carbon–sulfur compounds. Among the organics are many that are harmful, including known carcinogens such as benzene. Benzene is linked to several forms of leukemia, Hodgkin's disease, multiple myeloma, and non-Hodgkin's lymphoma (Agency for Toxic Substances and Disease Registry, 2000). Although the two samples were ostensibly from the same fire, albeit different vents, the relative concentrations varied considerably between the vents. For example, sample DE2486, representing the East vent, has considerably less benzene, carbonyl sulfide, dimethyl sulfide, and carbon disulfide, among other compounds, than sample DE2534. The order is reversed, with DE2486 having a greater concentration than DE2534, for other compounds, such as *i*-pentane and *n*-pentane, although not in the several-order of magnitude range as for the previous set of compounds. For the east vent, the benzene concentration of 14 ppb (one part in 10^9 parts), or 38 ng/g, is below the National Institute for Occupational Safety and Health's (NIOSH) eight-hour exposure limit of 100 ppb (Eco-USA, undated) and the 50 ppb 10-minute maximum during an eight-hour work shift (OSHA, 2005). In general, the gas analyses indicate that the gaseous emissions from this coal fire are potentially hazardous and prolonged exposure could be harmful to people in close proximity to the vents.

Table 2

Methane, carbon monoxide, and carbon dioxide and carbon–sulfur compounds and a suite of organic compounds.

Common name	Chemical formula	Units	West vent	East vent
			DE2534	DE2486
Carbon monoxide	CO	ppmv	87	1.9
Carbon dioxide	CO_2	ppmv	5600	1700
Carbonyl sulfide	OCS	ppbv	35	1.9
Dimethyl sulfide	$S(CH_3)_2$	pptv	290	19
Carbon disulfide	CS_2	pptv	500	13
Aliphatic compounds				
Methane	CH_4	ppbv	220000	24000
Ethyne	C_2H_2	ppbv	12000	49000
Ethene	C_2H_4	ppbv	230	360
Ethane	C_2H_6	ppbv	2600	6000
Propene	C_3H_6	ppbv	140	89
Propane	C_3H_8	ppbv	890	1800
1,3-butadiene	C_4H_6	ppbv	2.8	1.6
trans-2-butene	C_4H_8	ppbv	21	8
cis-2-butene	C_4H_8	ppbv	14	4
1-butene	C_4H_8	ppbv	18	9
i-butene	C_4H_8	ppbv	20	10
i-Butane	C_4H_{10}	ppbv	110	300
n-Butane	C_4H_{10}	ppbv	260	520
Isoprene	C_5H_8	ppbv	1.1	0.8
1-pentene	C_5H_{10}	ppbv	6.5	2.4
i-pentane	C_5H_{12}	ppbv	97	180
n-pentane	C_5H_{12}	ppbv	120	200
Methylcyclopentane	C_6H_{12}	ppbv	36	66
Cyclohexane	C_6H_{12}	ppbv	10	21
2,3-dimethylbutane	C_6H_{14}	ppbv	3.6	10
2-methylpentane	C_6H_{14}	ppbv	26	60
3-methylpentane	C_6H_{14}	ppbv	10	25
n-hexane	C_6H_{14}	ppbv	50	49
Methylcyclohexane	C_7H_{14}	ppbv	15	35
2,4-dimethylpentane	C_7H_{16}	ppbv	1.1	2.1
2,3-dimethylpentane	C_7H_{16}	ppbv	5.3	12
2-methylhexane	C_7H_{16}	ppbv	5.8	14
3-methylhexane	C_7H_{16}	ppbv	8.1	15
n-heptane	C_7H_{16}	ppbv	23	1.2
2,2,4-trimethylpentane	C_8H_{18}	ppbv	5.1	12
n-octane	C_8H_{18}	ppbv	13	0.8
n-nonane	C_9H_{20}	ppbv	6.5	<d.l.
Aromatic compounds				
Benzene	C_6H_6	ppbv	14	0.8
Toluene	C_7H_8	ppbv	35	0.9
Ethylbenzene	C_8H_{10}	ppbv	7.6	0.4
m&p-xylene	C_8H_{10}	ppbv	13	0.6
o-xylene	C_8H_{10}	ppbv	4.0	0.2

Ppmv = 1 part in 10^6; ppbv = 1 part in 10^9; pptv = 1 part 10^{12}.

4. Conclusions

The Middle Pennsylvanian Breathitt Formation Skyline coalbed is burning in an abandoned underground coal mine in northern Breathitt County, Kentucky. The date of inception and cause of this fire, the Tiptop fire, are unknown.

Two visits were made to the fire site, 7 May 2008 and 15 January 2009, to measure gas-vent temperatures and to determine the concentration of CO_2, CO, Hg, carbon-sulfur compounds, and organic compounds in the vent gases. The vent temperatures during the May 2008 visit were less than at other Kentucky fires (Stracher et al., 2008a,b), thereby permitting the measurement of CO_2, CO, and Hg within the operating temperature limits of the Jerome and Dräger instruments, although interferences are still a concern. CO_2 and CO concentrations peaked at 2.0 vol.% CO_2 and 600 ppm CO for the May 2008 analysis, exceeding OSHA safety limits for the geologists at the site. Mercury, as Hg^0, in excess of 500 ppm, about five times the OSHA eight-hour exposure limit, is also a hazard in the immediate vicinity of the vent. A number of carbon-sulfur and organic compounds in the vented gases may pose health threats. While compounds such as benzene are present at levels below the safety limits, prolonged exposure would be a health hazard.

While coal-fire gas is usually diluted with air as it exits and migrates away from a gas vent, temperature inversions or other meteorological conditions may concentrate gaseous toxins, resulting in a potential health hazard in populated areas. Hotter gas temperatures during the January visit to the Tiptop fire exceeded the temperature limits necessary for reliable measurements using Dräger tubes and the Jerome analyzer. In addition, the mixing of coal-fire gas from different vents during exhalation at the surface could limit the reliability of Hg vapor determinations. *In situ* gas-analytical techniques utilized for geothermal and volcanic studies (Engle et al., 2006; Aiuppa et al., 2007; Witt et al., 2008a,b) would be applicable to coal-fire gas vents and should be adapted for future investigations of these vents. Additional investigations should also examine the speciation of the Hg in vent emissions due to the drastically different deposition rates and bioavailability of elemental Hg, reactive gaseous Hg, and particulate Hg.

Disclaimer

Use of brand or trade names is for descriptive purpose and does not imply U.S. Geological Survey endorsement.

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