Re-entering a sealed heating: How gas indicators change

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ABSTRACT

Coal self-heating events have in the past been difficult to detect. Once a heating is detected, it is typically managed by injecting nitrogen or another inert gas into the area to reduce the oxygen supply to the heating and/or sealing the area.

Sometimes, it is necessary to re-enter a previously sealed area in which a heating has taken place, for example, to recover a longwall. Small-scale research suggests that when a coal heating which has previously been extinguished is re-exposed to air, the gas indicators for a given temperature differ to what they were when the coal was first exposed to air.

A heating was allowed to develop in a medium-scale test reactor containing approximately 50 kg of coal and then extinguished with nitrogen. Air was then again introduced into the column. This paper will present the gas data for coal undergoing a heating, a heating being inerted, and the gas data from when the coal was re-exposed to air. The effects of inerting a heating upon the gas indicators will be discussed.

INTRODUCTION

Coal self-heating events have posed a problem for hundreds of years (Walters, 1996). The detection of coal self-heating events has been aided by the discovery and use of gas indicators and ratios (Chamberlain, Hall and Thirlaway, 1970). Since Graham's ratio was developed in the early 1920's (Morris, 1988), there have been few significant developments in the methods used for the detection of coal self-heating events. Industry has made use of these indicators and treated many heatings successfully. Additional information will improve the ability to manage a coal self-heating event. Recent developments have shown that volatile organic carbons (VOCs) and aldehydes may be a useful tool to help a management team discriminate at what temperature a heating is at. In addition, there is research that suggests that the gas indicators may change after a coal self-heating event has been inerted (Taraba and Pavelek, 2016). This information would be important for mine personnel who need to manage an underground area after it has undergone a heating.

This paper documents an investigation into the use of natural gas components, VOCs and aldehydes to detect a heating. A two metre column apparatus which holds

approximately 50 kg of coal was used to step heat a sub-bituminous coal from central Queensland.

TEST EQUIPMENT

Simtars has built a two metre column for testing bulk coal samples. The column has an internal diameter of 200 mm, and is 2 m in length. It has an internal capacity of 62 L, which equates to 40 kg – 70 kg of coal depending on the packing density used. The column has 10 individually controlled heaters spaced along its length. It is wrapped with insulation to minimise radial heat loss. There are 10 thermocouples equally spaced along the length of the column, which measure the core temperature. There are another 10 thermocouples which measure the wall temperature corresponding to the location of each core thermocouple. This enables each heater to be individually controlled in one of two modes: set point or semi-adiabatic. In set point mode, the wall temperature at each location is maintained at a set temperature. In semi-adiabatic mode, the wall temperature is maintained at no more than 0.5 °C below the core temperature as the core temperature increases. A gas port is located adjacent to each core thermocouple which enables gas samples to be taken. Figure 1 shows a schematic of the column layout.
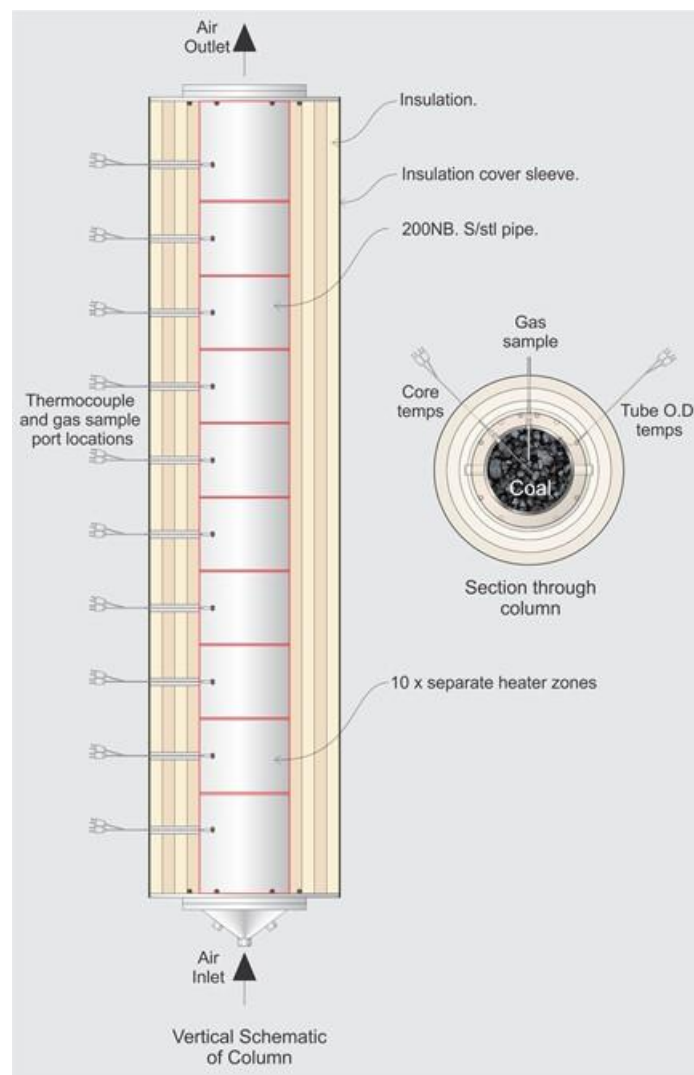

Figure 1: Schematic cross section of the Simtars 2-Metre Column

TEST PROCEDURE

Run of mine coal was obtained from a central Queensland mine. It was not crushed or treated in any way. Three 20 L buckets of coal were loaded into the column. Samples for R_{70} and moisture testing were collected. The column was then sealed for 2.5 days. A gas sample was taken from the exhaust of the column to simulate a sealed goaf sample. Air was then applied at 250 mL/min to the base of the column and allowed to exhaust out the top of the column. The coal was heated to 40 °C and then step heated in 5 °C increments every 24 to 48 hours until the coal temperature reached 90 °C. Gas samples were collected in Tedlar bags from the exhaust and every second port along the length of the column when the coal reached key temperatures. These samples were analysed for the standard gases, natural gases and VOCs using Agilent CP490 micro gas chromatographs. Sorbent tubes were used to collect samples that could be analysed by high performance liquid chromatography (HPLC) for volatile organic carbons (VOCs) and aldehydes. The types of sorbent tubes used were SKC 226-01 and SKC 226-119 respectively. This paper only presents data to this point. Further research will investigate the effects of higher coal temperatures and nitrogen inertisation upon the gas indicators.

R_{70} AND MOISTURE CONTENT RESULTS

A representative coal sample from the column was milled to less than 250 µm, dried under nitrogen at 110 °C for 16 hours and then 150 g was tested under oxygen in an adiabatic oven. The R_{70} value is the average rate at which the coal heats from a starting temperature of 40 °C until 70 °C is reached and is expressed in units of °C/h. The R_{70} value determined from this test was 7.68 °C/h. The initial moisture content of the R_{70} sample, prior to it being dried, was 13.62%.

PERMANENT GAS EVOLUTION

The Tedlar bag gas samples collected were analysed by the Simtars gas laboratory for the permanent gases. These gases are helium, hydrogen, oxygen, nitrogen, methane, carbon monoxide, carbon dioxide, ethylene and ethane.

Figure 2 shows the change in CO and CO_2 concentration at the exhaust as the coal temperature increased. The CO_2 concentration was initially over 13% as this sample was taken while the column was sealed. After the air was applied, it can be seen that the CO_2 concentration dropped to just under 1%, at 45 °C. The concentration then steadily increased as the coal temperature increased. It is important to note that any indicators that rely upon CO_2 will be affected by CO_2 present as a seam gas.

The CO concentration also initially increased with increasing coal temperature. When the coal temperature reached 50 °C, the CO concentration began decreasing with increasing coal temperature. The CO concentration did not begin to increase again with temperature until 65 °C was reached. This CO trend was consistent along the length of the column. It is both unusual and unclear why this occurred.
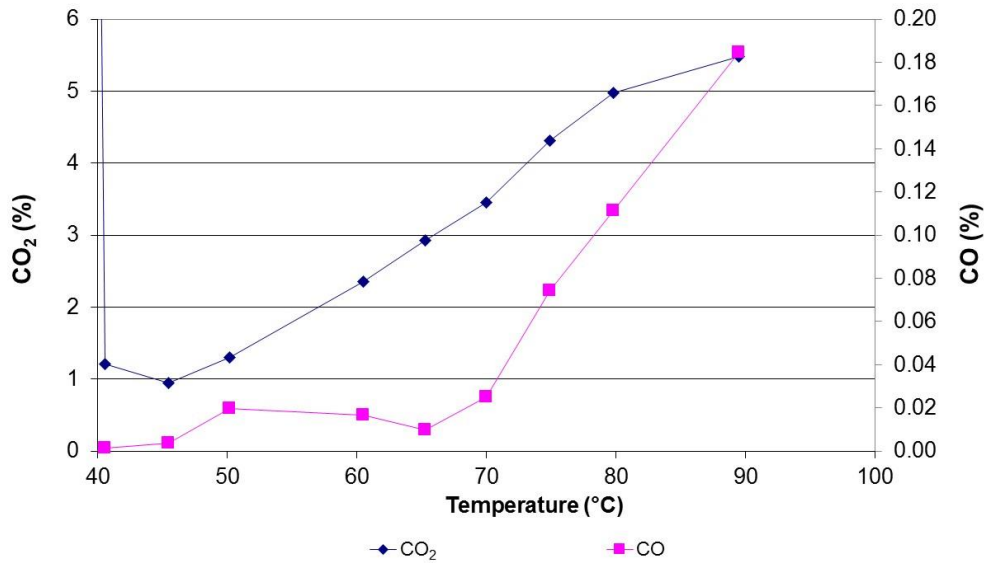
Figure 2: Change in CO and CO$_2$ concentration at the exhaust with increasing coal temperature.

Figure 3 shows the change in CO$_2$ concentration at various distances from the air inlet and at the column exhaust as the coal temperature increases. When the coal had reached a temperature of 90 °C, the CO$_2$ concentration had reached 3.6% at a distance of 0.55 m from the air inlet. At the exhaust, the CO$_2$ concentration was 5.5%. Over 65% of the CO$_2$ was generated in the first 25% of coal. Whilst the column was step heated and the temperature profile across the column was relatively even, the coal towards the air inlet of the column would have been drier due to higher rates of moisture evaporation than the coal further downstream. When the coal temperature was set to 90 °C, it was noted that the coal temperature was decreasing at a distance of 0.36 m from the air inlet. This indicates the development of a zone at this location with a higher rate of moisture evaporation resulting in an increased availability of oxidation sites and hence increased gas generation.
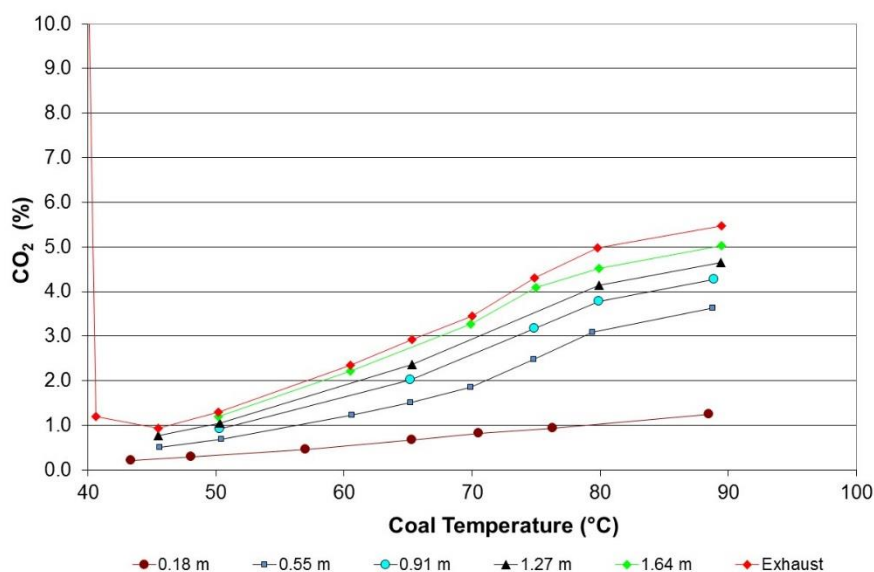

Figure 3: Change in CO$_2$ concentration within the column at various distances from the air inlet including the exhaust as the coal temperature increases.

Figure 4 shows the change in methane, ethane, ethylene and hydrogen concentrations with increasing coal temperature. The gas sample taken when the column was sealed had a methane concentration of 0.0107% (107 ppm). The methane concentration then dropped to 0.0002% when air was applied to the column. The methane concentration did not trend with increasing temperature. It appears that it is present in small concentrations as a seam gas.
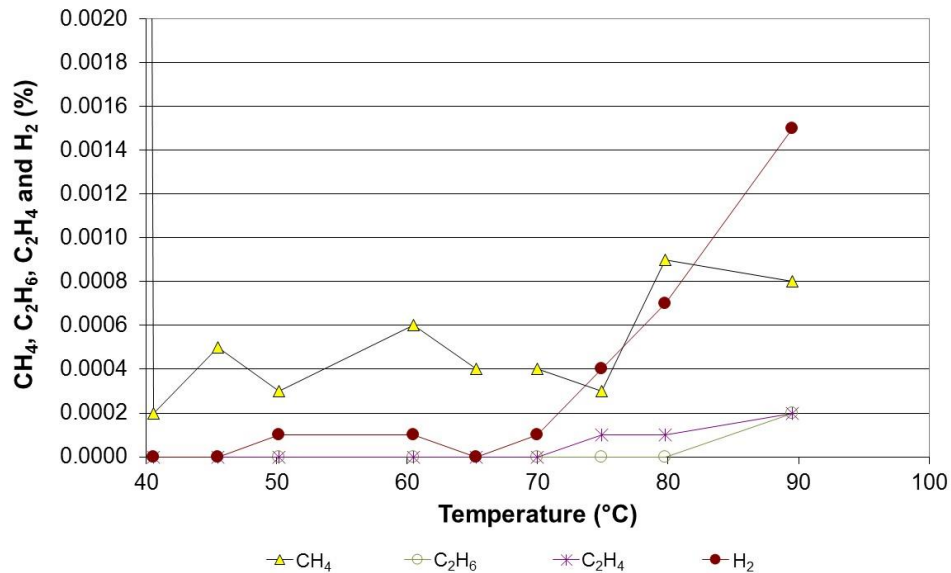

Figure 4: Change in CH_4, C_2H_6, C_2H_4 and H_2 concentration with increasing coal temperature.

Trace amounts of hydrogen were detected at coal temperatures below 65 °C. Once the coal temperature reached 65 °C the hydrogen concentration began to rapidly increase as the coal temperature increased, reaching a concentration of 0.0015% when the coal reached 90 °C.

Trace amounts of ethylene and ethane were detected once the coal had reached a temperature of 75 °C and 90 °C respectively. The trace amounts of ethylene being detected at 75 °C are a reflection of the coal rank being sub-bituminous. It should be noted that ethane was not detected when the column was initially sealed and therefore is not a seam gas for this coal.

Figure 5 shows the change in Graham's, Young's and Trickett's ratios with increasing coal temperature. The Graham's ratio increases initially with increasing coal temperature until the coal temperature reached 50 °C. The Graham's ratio then decreased with increasing coal temperature. This is because the CO concentration decreased as shown in Figure 2. When the coal reached a temperature of 65 °C, Graham's ratio and CO began to increase with increasing coal temperature.

The Young's and Trickett's ratios were almost identical in value. They did not increase or decrease significantly as the coal temperature increased. They are therefore not useful indicators for this coal below 90 °C.

Figure 6 shows the change in O_2 deficiency, CO/CO_2 and H_2/CO ratios with increasing coal temperature. The O_2 deficiency was initially 20.6% when the column was sealed. After air was applied, the O_2 deficiency dropped to 2.0% and then

steadily increased with increasing coal temperature. The CO/CO_2 ratio was affected by the drop in CO concentration between the coal temperatures of 50 °C and 65 °C and is identical in trend to the Graham's ratio. The H_2/CO ratio was also affected by the drop in CO. Once the coal temperature reaches 65 °C, the H_2/CO increases with increasing temperature.
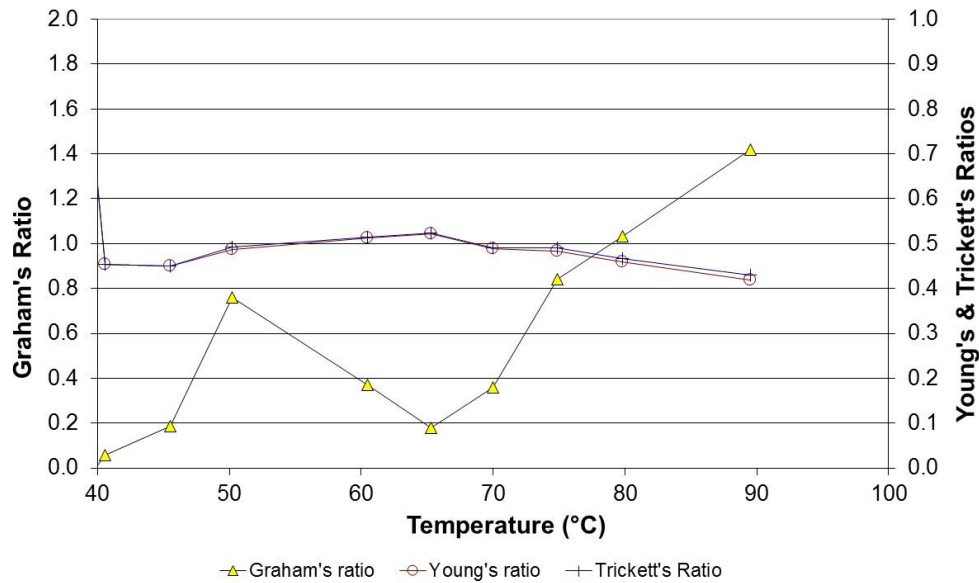

Figure 5: Change in Graham's, Young's and Trickett's Ratio at the exhaust with increasing coal temperature.
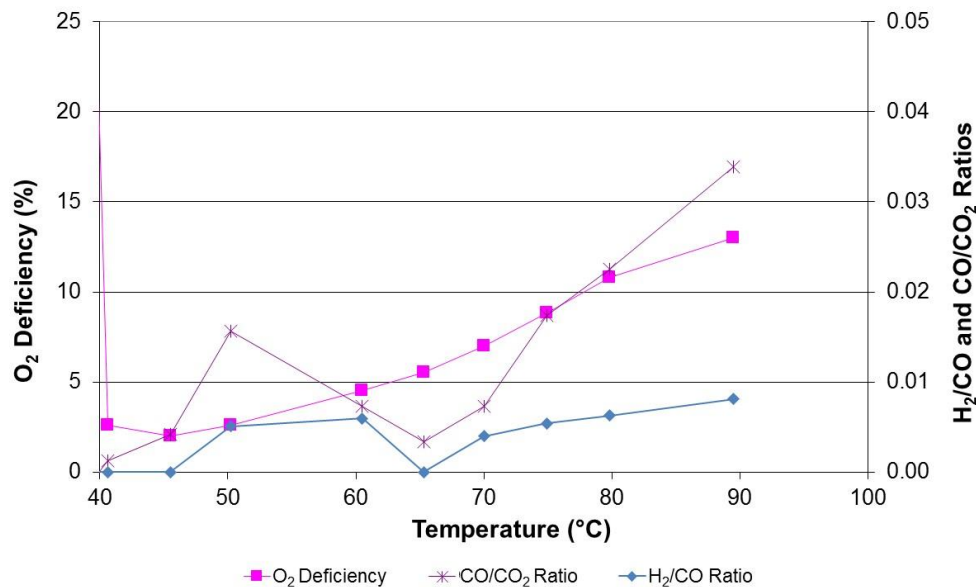

Figure 6: Change in O_2 Deficiency, CO/CO_2 and H_2/CO Ratios with increasing coal temperature.

NATURAL GAS EVOLUTION

The Tedlar gas samples collected during the test were also analysed for natural gases. The gases reported on were: propane, iso-butane, n-butane, neo-pentane, iso-pentane, n-pentane and hexane. They were only detected at temperatures

greater than 60 °C. All gas concentrations less than 0.0020% (20 ppm) are indicative only.

Table 1 shows that iso-butane is detected when the coal reaches temperatures in excess of 65 °C. It appears that the iso-butane was produced at the point where the coal was drier towards the air inlet and was subsequently scrubbed out by either the moisture or coal further downstream. This effect can be seen in the drop in iso-butane concentration at a distance of 0.55 m from the air inlet to a distance of 0.91 m from the air inlet for the coal temperatures of 70 °C to 80 °C.

Table 1: Change in iso-butane and ethylene concentration within the column at various distances from the air inlet including the exhaust as the coal temperature increases.

Distance from Air Inlet		0.18 m	0.55 m	0.91 m	1.27 m	1.64 m	Exhaust
Nominal Temperature	**Gas (%)**						
65 °C	Ethylene	0	0	0	0	-	0
	iso-Butane	0.0001	0	0	0	-	0
70 °C	Ethylene	0	0	-	-	0	0
	iso-Butane	0.0002	0.0001	-	-	0	0
75 °C	Ethylene	0	0.0001	0.0001	-	0.0001	0.0001
	iso-Butane	0.0002	0.0004	0.0002	-	0	0
80 °C	Ethylene	-	0.0001	0.0001	0.0001	0.0002	0.0001
	iso-Butane	-	0.0007	0.0005	0.0000	0.0001	0
90 °C	Ethylene	0.0001	0.0001	0.0002	0.0002	0.0002	0.0002
	iso-Butane	0.0002	0.0011	0.0011	0.0011	0.0011	0.0005
				Note: All concentrations below 0.0020% are indicative only.			

The presence of iso-butane was detected within the column at lower coal temperatures than ethylene. The rate of increase of the iso-butane concentration at the exhaust is more than twice that of the ethylene concentration at the exhaust when the coal temperature has reached 90 °C.

Figure 7 shows iso-pentane is detected when the coal reaches temperatures in excess of 65 °C. In a similar fashion to iso-butane, the iso-pentane concentration profiles show evidence of the iso-pentane being scrubbed out of the gas stream by either the coal or moisture within the column. In contrast to iso-butane, however, the presence of iso-pentane is detected in the exhaust at the lower temperature of 75 °C and the concentration of it increases with increasing coal temperature. When the ethylene concentration in the exhaust is approximately 1 ppm, the iso-pentane concentration is also approximately 1 ppm. However, when the coal temperature increases from 80 °C to 90 °C, the iso-pentane concentration increases by a factor of five, whereas the ethylene concentration only increases by a factor of two.

The other natural gases reported on are not useful indicators for this coal. At coal temperatures of 90 °C and below, n-pentane and hexane were not detected at all. The presence of n-butane and neo-pentane was detected at concentrations of less than 0.5 ppm when the coal temperature reached 90 °C. Propane was also detected at concentrations of up to 1 ppm when the coal temperature reached 90 °C.
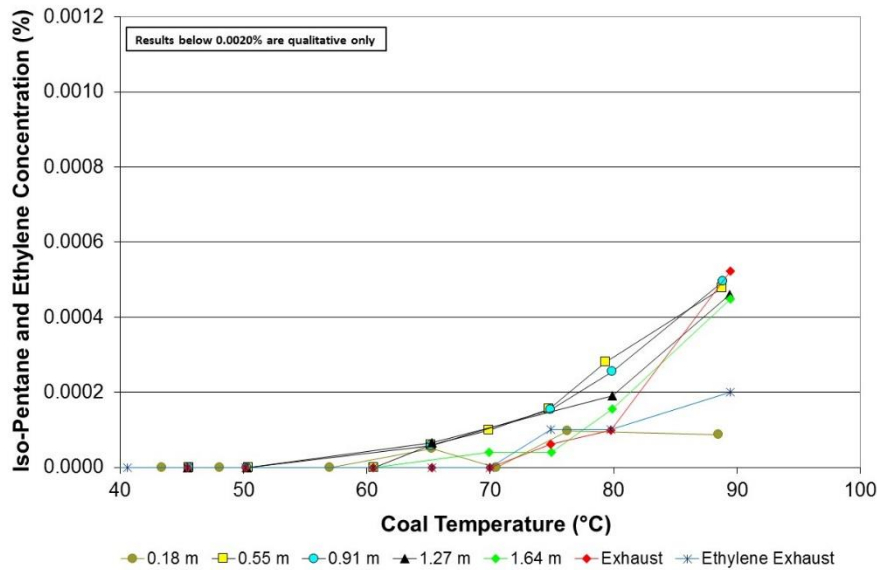
Figure 7: Change in iso-pentane concentration within the column at various distances from the air inlet including the exhaust as the coal temperature increases. The ethylene concentration at the exhaust is also shown for comparison.

VOC AND ALDEHYDE EVOLUTION

SKC sorbent tubes were used to collect samples for VOC and aldehyde analysis. Tube types were 226-01 and 226-119 respectively. The tubes were then analysed using high performance liquid chromatography (HPLC). Of the VOC suite analysed for, only methyl ethyl ketone (MEK) and acetone were detected. Figure 8 shows MEK was first detected when the coal temperature was 80 °C. It was detected within the column but not at the exhaust. It would appear that the MEK can be scrubbed out from the air stream by either moisture or coal. When the coal had reached 90 °C, MEK concentrations were consistently measured as being between approximately 1.6 mg/m^3 to 1.8 mg/m^3, both within the column and at the exhaust.

Acetone was able to be detected using both types of sorbent tubes. The graph in Figure 9 shows the acetone concentrations at the exhaust as measured by both tube types as the coal temperature increased. While there is a difference between the values given by the two methods it is important to note that the trends are similar. The change in ethylene concentration at the exhaust is also plotted in Figure 9 for comparison to the acetone trend. Between 80 °C and 90 °C the ethylene trend increases by a factor of two, whereas the acetone trend increases by a factor of at least four.

The acetaldehyde and propionaldehyde concentrations at a distance of 1.82 m from the air inlet are shown in Figure 10 as well as the gas concentrations at the exhaust. At a distance of 1.82 m from the air inlet, acetaldehyde and propionaldehyde are detected when the coal has reached 80 °C whereas they were not detected in the exhaust. When the coal had reached a temperature of 90 °C, acetaldehyde and propionaldehdye are able to be detected in the exhaust. It would therefore appear

that the aldehydes are being scrubbed out of the air stream by either the coal or moisture within the column.
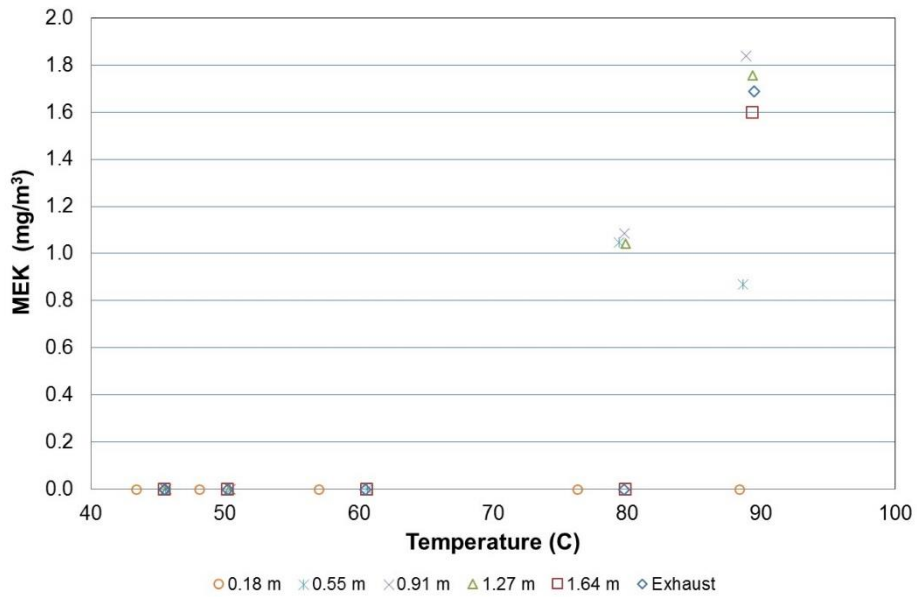

Figure 8: Change MEK concentration within the column at various distances from the air inlet including the exhaust as the coal temperature increases.
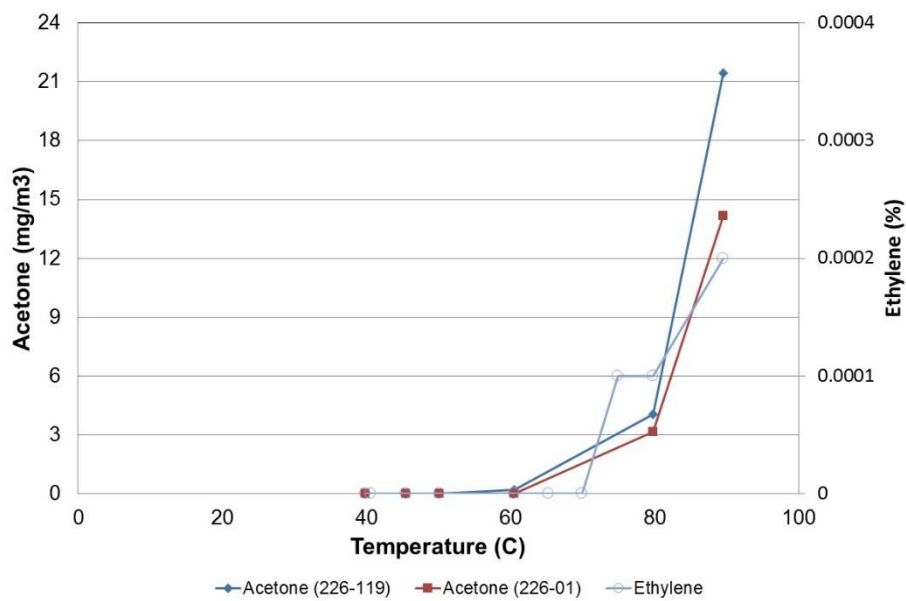

Figure 9: Change in acetone concentration at the column exhaust as the coal temperature increases. The values obtained from both the VOC tubes (226-01 and aldehyde tubes (226-119) is shown. The ethylene concentration at the exhaust is also shown for comparison.
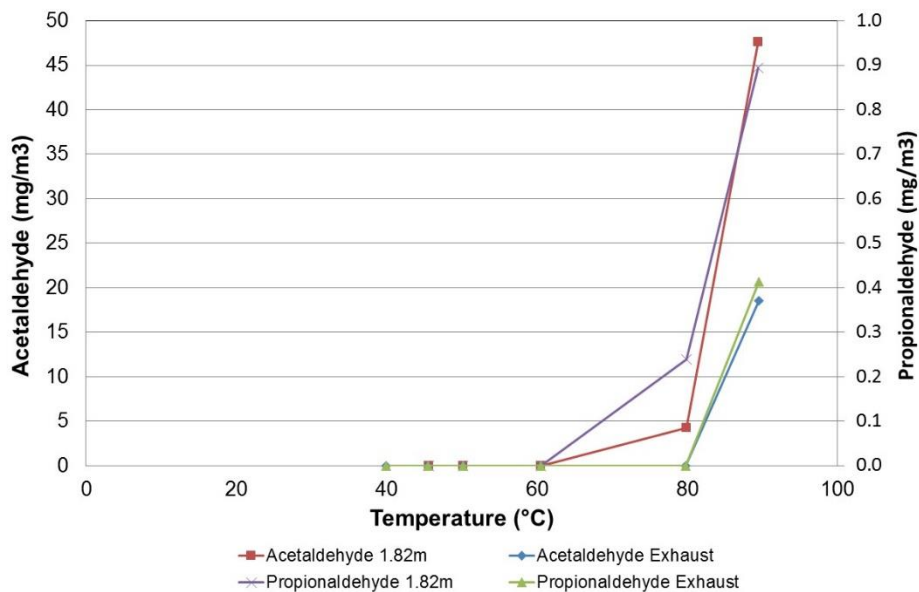
Figure 10: Change in acetaldehyde and propionaldehyde within the column and at the exhaust as the coal temperature increases.

CONCLUSIONS

- 2-metre columns can be used to enable a better understanding of traditional and non-traditional gas indicators of coal self-heating events.

- Iso-butane and iso-pentane are detected at coal temperatures greater than 65 °C and are more responsive to increasing coal temperature than C_2H_4.

- Natural gases, VOCs and aldehydes may potentially be scrubbed from the air stream by either the coal or moisture.

- High moisture content blocks access of oxygen to the coal reactive sites and thus inhibits coal oxidation.

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