

VOLATILE ORGANIC COMPOUNDS FROM COAL WASTE ROCKS SUBJECTED TO SELF-HEATING

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Introduction

Coal-waste dumps, a common landscape feature in coal mining regions, are a potential source of hazardous substances emitted to the atmosphere and leached to surface- and ground waters. Moreover, dumps are commonly subject to self-heating or even open fire, processes that increase their negative environmental impact by releasing gases and water-soluble inorganic- and organic compounds. Since coal-waste dumps are commonly located in highly-populated industrial regions, they should be considered a significant environmental- and health hazard. Noxious odours and dust associated with self-heating constitute, at the very least, a nuisance to the nearby residents. The major compounds emitted are CO₂, accompanied by CO and lighter organic compounds, partially derived from gaseous compounds trapped in the pores of organic matter and partly from pyrolysis, depending on the degree of heating, coal rank, and oxygen availability (Fabiańska et al. 2013).

Gas samples, 57 in all, were collected from seven coal waste dumps: 32 samples in the Upper Silesian Coal Basin (USCB; 12 at the Wełnowiec dump, 10 at the Rymer Cones, 5 at the Anna dump and 5 at the Czerwionka-Leszczyny dump) and 25 in the Lower Silesian Coal Basin (LSCB; 21 at the Słupiec dump, 3 at the Nowa Ruda dump and 1 at the Przygórze dump). The number of samples depended on the number of active heating sites and their intensity. Gas samples were taken using syringe samplers that sucked in 100 cm³ of gas. Samples were collected a few centimetres below the surface or deeper (< 1.5 m), if possible. Additionally, temperature was measured at each sampling site.

The molecular composition of self-heating gases (CH₄, C₂H₆, C₃H₈, *i*-C₄H₁₀, nC₄H₁₀, C₅H₁₂, C₆H₁₄, C₇H₁₆, unsaturated hydrocarbons, CO₂, O₂, H₂, N₂) and their concentrations were determined on an Agilent 7890A gas chromatograph equipped with a set of columns, flame ionization (FID) and thermal conductivity (TCD) detectors.

Results

Based on the temperatures measured at the sampling sites, and the degree of heating, gas samples were divided into (1) samples from sites with no known history of fire or where thermal activity had ceased and (2) samples from sites where self-heating was on-going.

The typical gaseous phases at a site without thermal activity, as might be expected, atmospheric gases, i.e., nitrogen and oxygen (Table 2). Average percentage contents for the USCB and LSCB are as follows: N₂—8.6 and 80.1% vol., respectively, and O₂—18.8 and 20.8 % vol., respectively. Carbon dioxide at these sites, i.e., 1.085 (USCB) and 0.151 (LSCB) % vol., is elevated compared to the average (0.035 % vol.) atmospheric content. However, at

some sites without thermal activity, emitted CO₂ was not evident. The atmospheric gases were accompanied by small amounts of organic compounds, among which methane (0.0061 and 0.0040 % vol.) and ethylene (0.0030 and 0.0011 % vol.) predominate. It is notable that CH₄ contents are 22 and 34 times higher than those in the air (0.00018% vol.), despite the absence of thermal activity. This indicates that coal waste should be considered an important source of greenhouse gases even when self-heating is not evident. Heavier aliphatic hydrocarbons from *cis*-2-butene to *n*-hexane were also detected, but in traces (0.0001 - 0.0005 % vol.).

The general composition of gases from sites with on-going self-heating show a significant decrease in O₂ content; values may be < 5.5 times lower than atmospheric composition. Of the atmospheric gases, carbon dioxide was the predominating component measured, averaging 3.7350 (USCB) and 5.2447 (LSCB) % vol. In these sites, organic gases included also methane (1.3233 and 0.1432 % vol., respectively), the saturated aliphatic hydrocarbons ethane, propane, *n*-butane, *n*-pentane, *n*-hexane, *n*-heptane, *iso*-butane, and *iso*-pentane, together with the unsaturated aliphatic hydrocarbons ethylene, acetylene, propylene, *trans*-, and *cis*-2-butene. Mostly their concentrations decrease with increasing molecular weight but, in some LSCB samples, there are elevated contents of propane and *n*-butane. Thermal activity also led to elevated hydrogen contents in the gases, 0.2125 (USCB) and 0.0186 (LSCB) % vol. These values many times exceed average atmospheric H₂ concentrations (0.0000055% vol.). Both unsaturated hydrocarbons and hydrogen should be considered pyrolytical products of self-heating. They are absent in natural gas whereas common in such industrial gases as refinery gas and gas from thermal cracking of coal.

Conclusions

Gaseous emissions from self-heating coal-waste dumps should be deemed a major environmental hazard. Gas composition is related to the stage of self-heating; at initial- and waning stages of heating, CH₄ increases relative to CO₂ whereas, at the peak stage, emitted gases are dominated by CO₂. Two major components formed due to self-heating, namely, methane and CO₂, are greenhouse gases. Even though it is usually considered that CH₄ values of >1.0% vol. should lead to immediate action when the compound is found in wells or vents, in the case of coal waste dumps, no action is undertaken. Mostly, the methane is oxidized to CO₂, as shown by a good substrate-product correlation, but some of it may contribute to the general temperature elevation. Both ethylene and methane are also produced at sites without any recent thermal activity, possibly due to bacterial decay of coal wastes.

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References

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