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SELF-HEATING COAL WASTE FIRE MONITORING AND RELATED ENVIRONMENTAL PROBLEMS: CASE STUDIES FROM POLAND AND UKRAINE

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Abstract

The self-heating of coal waste dumps is considered as a serious environmental issue, wherever active or inactive coal mining has been present. This issue is introduced from two active coal mining regions from Poland (Upper Silesian Coal Basin) and Ukraine (Donetsk Coal Basin) based on mineralogy, organic petrography and geochemistry, and remote sensing techniques. Thermally affected coal wastes reveal changes recorded by organic and mineral matter. Irregular cracks and fissures appear within and at the edges of organic matter particles, which are oxidised, devolatilised and plasticised. Mineral phases underwent oxidation, dehydration, structure rebuilding and recrystallisation. Highest temperatures generated during the fire cause melting and paralava formation. During self-heating, some chalcophile elements like Hg (mostly present as HgS), Pb, Zn can be enriched and released, or different organic pollutants like phenols (originated from vitrinite particles), different PAHs with alkyl substitutes, chlorinated PAHs, or sulphur heterocycles are formed. The introduced remote sensing techniques helped to localise and monitor hot spots with different temperature ranges. Applying SWIR bands of Landsat hot spots from extremely burning dumps in Ukraine were successfully localised, however, only night-time scenes with SWIR can be used. The sun's disturbing effects should be considered as an influential factor for both thermal imaging camera or satellite images. Thermal cameras can reveal the most detailed signs of low to high temperature anomalies with different cracks and line shapes.

Keywords: coal waste dumps, self-heating, remote sensing, organic – inorganic pollutants

INTRODUCTION

Coal wastes are produced during coal mining and coal processing. Usually, such wastes contain 5–30% organic matter and ca 70–95% of minerals, mainly sulphides (e.g., pyrite or sphalerite), alumino-silicates, carbonates, and other minerals rich in heavy metals (Skarżyńska, 1995; Finkelman, 2004; Huang, 2004; Sütő et al., 2007; Zhao et al., 2008a, 2008b; Kang et al., 2011; Xiao et al., 2015). After deposition in dumps located in close vicinity of coal mines, the waste starts to weathering what might lead to the exothermic oxidation of the organic mater known as self-heating, which may lead to self-ignition (Zhang and Kuenzer, 2007; Carras et al., 2009; Masalehdani et al., 2009; Misz-Kennan and Fabiańska, 2011). Susceptibility to self-heating is increased by air temperature, wind, organic-matter rank, ash content, surface area exposed to air, particle size, moisture and oxygen content, shape, layering and compaction of the dumped pile (Skarżyńska, 1995; Lohrer et al., 2005; Pone et al., 2007). In the case of bituminous or sub-bituminous coals and coal wastes, the temperature increases slowly until a threshold temperature of 60–80°C, and then the temperature rises fast, followed by self-ignition and burning of the waste. The temperature inside a coal waste dump is typically highly variable ranging from ambient to ~ 500°C, however, at the end stages of heating in the subsurface, the self-combustion temperatures can reach up to 1000–1300°C (Carras et al., 1994, 1999; Skarżyńska 1995; Heffern and Coates 2004; Sütő et al., 2007; Querol et al., 2008; Ribeiro et al., 2010; Jendruś 2016).

Coal-waste dumps, are typically occupying large areas and are located nearby urban areas, thus they have potential health and environmental impacts (Querol et al., 2008; Zhang, 2008; Zhao et al., 2008b; Li, 2010; Liu and Liu, 2010; Li et al., 2011; Zhou et al., 2014; Nádudvari et al., 2020a, 2021b). Toxic elements in coals are associated with organic matter and minerals, e.g., clays and sulphides (Vassilev et al., 2001; Ciesielczuk et al., 2014). Such waste dumps emit enormous quantities of greenhouse gases: CO_2, CH_4, and such harmful compounds as NO_x, NH_3, SO_x, H_2S, HCl, CH_2O, benzenes, phenols, PAHs, heavy metals (Kruszewski et al., 2018, 2020; Nádudvari et al., 2021b). In the dumps elevated Hg (~100–1078 mg/kg) and Pb concentrations (~600–2000 mg/kg) can be reached, reflecting evaporation of these metals from deeper parts of the dumps. The acidic pH levels (3.0–4.5) may help to mobilise these elements (Nádudvari et al., 2021b).

The aim of the paper is to describe and understand the complex processes occurring within coal waste dumps based on organic petrography, mineralogy, and organic geochemistry and combine remote sensing techniques. On the selected Polish dumps the ongoing self-heating was well documented e.g. Misz-Kennan and Fabiańska (2011), Nádudvari (2014), Fabiańska et al. (2019), Kruszewski et al. (2018) and Nádudvari et al. (2018, 2021, 2021b) therefore these researches were provided good insight about the state of self-heating, pollution levels – especially heavy metals e.g. Hg, Pb or organic pollutants: benzene, phenols, PAHs. In Poland and Ukraine many of those coal waste dumps are surrounded by urban areas (Fig. 1), so their self-heating pose a serious threat to the local community through, among others, environmental pollutions, odorous smells and surface collapsing. Therefore the application of remote sensing techniques and thermal imaging cameras are crucial to protect society - prevent self-heating, conduct its proper monitoring and warn about the hazard.

STUDY AREA

The self-heating phenomenon of coal waste dumps most often occurs in coal basins, where the large number of dumps are created near the coal mines. In this paper, we focus on some of the largest coal basins in Europe: Upper Silesian Coal Basin (USCB; southern Poland) and Donetsk Coal Basin (DCB; eastern Ukraine) (Fig. 1). In both basins, large-scale coal mining began in the 19th century. Since then, many dumps have been created there, which have subsequently undergone numerous processes. As the waste in both cases came from coal mines, their composition was relatively similar. The coals of the DCB and USCB are from bituminous coal to anthracite and have high ash yields and high sulfur contents (Panov et al., 1999; Kędzior, 2019). The dumps in USCB are characterised by enormous diversity in the area, volume, and shape. These objects often fit well into the highly urbanised landscape (especially in the Katowice district). In the DCB case, dumps have large-area with a typically conical structure (for this reason such dumps are often called as Donetsk type – Fig. 1D). These dumps stand out above the landscape, towering over the surrounding settlements. Regardless of the differences and similarities between the two basins, the self-heating phenomenon was detected in both regions, and its impact on the natural environment and society was determined (Panov et al., 1999; Fabiańska et al., 2019; Nádudvari et al., 2018, 2020a, 2021b; Abramowicz et al., 2021).

METHODS

Approx. 500–1000 g were taken from the dumps up to 10 cm-s depth and collected/stored in alumina foil to prevent other organic matter contamination. Samples were manually cleaned from roots, wood tissues and dried at room temperature (ca 22°C) for ca. 5 days. After they were uniformed and about 200 g milled and powdered in a rotary mill very fine grain size. These samples were used for GC-MS analysis.

For petrographic analyses the samples were air dried, crushed to < 1mm, embedded in epoxy resin and polished blocks were prepared. Sometimes also oriented fragments of coal wastes were embedded in epoxy resin, and polished blocks were prepared. Samples were examined in reflected white fluorescent light. That allows to identify various macerals and forms of alteration of organic matter. The identification was carried out at 500 points in each sample according to ISO 7404-3 (2009). At the same time, minerals and mineral phases were identified. The aim of petrographic investigations was to establish the maceral composition of the wastes and their degree of alteration. Reflectance measurements were carried out at 50–100 points on each sample depending on

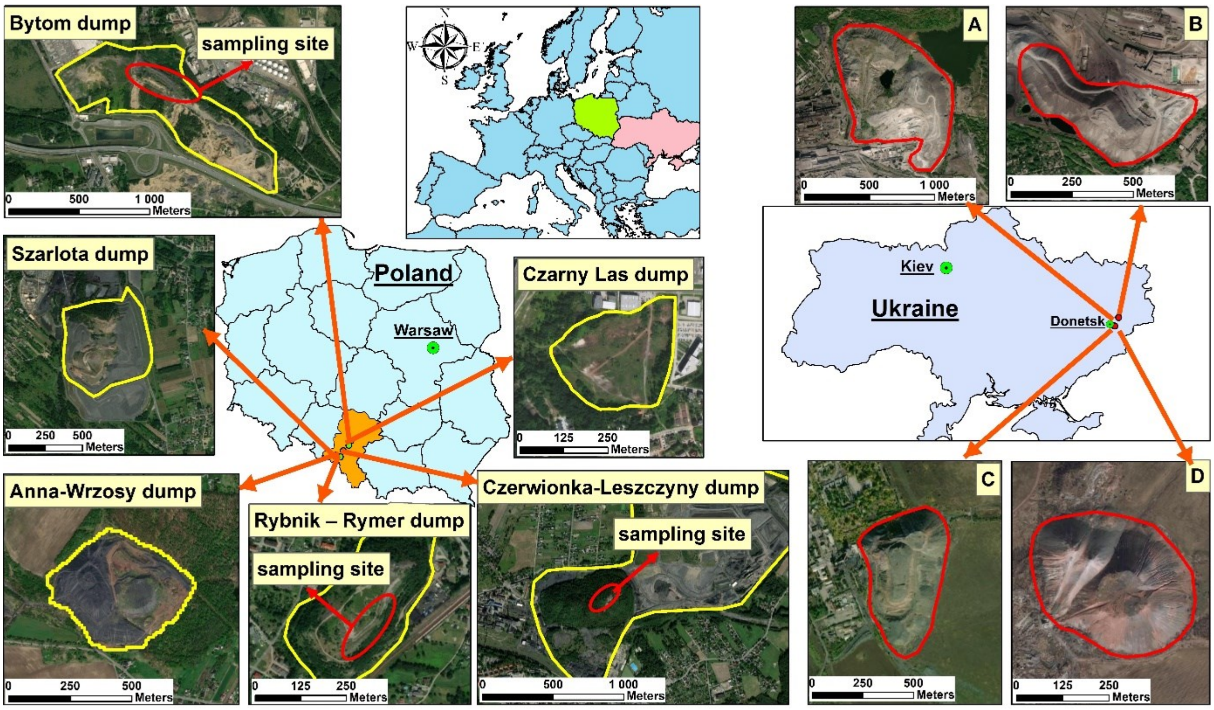


Fig. 1 Location of the studied coal wastes dumps from Poland and Ukraine. Background map: GoogleEarth – 2021

the number of available particles and according to ISO 7404-5 (2009). The vitrnite reflectance was aimed to determine the rank of original organic matter and their thermal alteration. It was done using Axioplan II optical microscope equipped with immersion objective and applied magnification of 500X.

Gas chromatography-mass spectrometry (GC–MS) samples were powdered (ca 18–20 g) and extracted using dichloromethane (DCM) and methanol (1:1) with an ultrasonic bath at 30–40°C for 15–20 min. The extracts were pooled, filtered, and the solvents evaporated at room temperature. The dry residue was dissolved in 0.5 ml of DCM and analysed by GC-MS an Agilent Technologies 7890A gas chromatograph and Agilent 5975C Network mass spectrometer with a Triple-Axis detector (MSD) at the Institute of Earth Sciences, Faculty of Natural Sciences, University of Silesia. Helium (grade 6.0) was used as a carrier gas at a constant flow of 2.6 ml/min. Separation was obtained on fused silica capillary column, DB-5 (60 m × 0.25 mm i.d.; film thickness 0.25 µm) coated with a chemically bonded phase (5% phenyl, 95% methylsiloxane), for which the GC oven temperature was programmed from 45 (1 min) to 100°C at 20 °C/min, then to 300°C (held for 60 min) at 3°C/min, with a solvent delay of 10 min. The GC column outlet was connected directly to the ion source of the MSD. The GC–MS interface was set at 280°C, while the ion source and the quadrupole analyser were set at 230 and 150°C, respectively. Mass spectra were recorded from 45 to 550 da (0–40 min) and 50 to 700 da (> 40 min). The MS was operated in the electron impact mode, with an ionisation energy of 70 eV. The results were obtained in full scan mode and processed with the Hewlett-Packard Chemstation software. The studied compounds were identified according to their mass spectra (using peak areas acquired in the manual integration mode), by comparison of peak retention times with those of standard compounds, by interpretation of MS fragmentation patterns, and from mass spectral databases GEOPETRO, NIST17 (National Institute of Standards and Technology) and Wiley (W10N11) (Philp, 1985; McLafferty and Stauffer, 1989).

Mineral phase identification was established using X-ray diffraction and scanning microscopy. Powdered samples were determined using a fully automated X-ray Philips PW 3710 diffractometer operated at 45 kV and 30 mA, CuKα radiation, and equipped with a graphite monochromator. Mineral morphologies and spatial relationships between components were examined in thin sections and samples fragments using a Philips XL 30 ESEM/TMP scanning electron microscope in environmental mode, coupled with an energy-dispersive spectrometer (EDS; EDAX type Sapphire). Analytical conditions of the SEM included: accelerating voltage of 15 kV; a working distance of ca 10 mm; counting time of 40 s. All these researches were carried out at the Faculty of Natural Sciences, University of Silesia, Sosnowiec, Poland.

The remote sensing and thermal camera monitoring possibilities of self-heating coal waste was applied by Landsat satellite and thermal camera images. Application of remote sensing for such hot spots must be taken into account solar influences (e.g., different albedo, slope, aspect or vegetation cover) can significantly influence thermal anomalies (Zhang and Kuenzer, 2007). However, extended observation permits persistent heat sources due to self-heating to be recognised, as well as their migration, intensification, and disappearance, when they are hot enough for detection with satellite sensors (Nádudvari, 2014). Using the Thermal Infrared Sensors (TIRS) of satellite images, i.e., Landsat series, ASTER is the cost-effective and time-saving technique for monitoring coal waste fires and detecting their thermal anomalies. Several authors have successfully applied Landsat, ASTER night-time and winter-time images for localising such coal- or coal-seam fires worldwide (e.g., Voigt et al., 2004; Gangopadhyay et al., 2005; Chatterjee, 2006; Zhang and Kuenzer, 2007; Prakash et al., 2011; Guha and Kumar, 2012). Therefore, to monitoring such coal waste fires, only night-time and/or daylight snow-covered images should be used (Nádudvari et al., 2021a). The most effective sensor for hot spot detection is Landsat 7 ETM+ as the TIRS band has 60 m resolution compared with Landsat 4-5 TM (120 m) or Landsat 8 OLI (100 m) because ETM+ can detect properly smaller hot spots which Landsat 8 OLI, or TM cannot. However, adding the SWIR (short wavelength infrared) bands (30 m) can precise the TIRS temperatures (Nádudvari et al., 2020b). Lower temperature phenomena, such as small ground thermal anomalies, must be studied in the thermal infrared (TIR) window (8–14 µm) (Bonneville et al., 1985; Bonneville and Kerr 1987). Self-heating can also be classified using the self-heating intensity index (SHII) introduced by Nádudvari et al. (2021a). It can be calculated from the highest (pixel max), and lowest (pixel min) values based on the area of a coal waste dump taken from Landsat, ASTER (TIRS) bands. This index can help to classify the fires of ongoing self-heating where snow-covered or night-time Landsat, ASTER images are available (Nádudvari et al., 2021a). However, in the case of daylight cold, snow-covered images, the albedo, solar radiation, slope and aspect can significantly impact the thermal anomalies. That can indicate low thermal activity, which is usually visible on Landsat thermal map with a 2–3°C difference between pixel max (hot spot)—pixel min (cold, snow-covered surface with no thermal activity). Hot spots with lower thermal activity or their extensions smaller than the TIRS satellite sensor capabilities making difficult to detect such hot surfaces (Nádudvari and Ciesielczuk, 2018; Nádudvari et al., 2021a).

Pictures from the camera mounted on the drone can reach a resolution of several centimetres, therefore such measurements provide the best option to monitor self-heating dumps. It is mainly due to the distance of the measuring device from the measured surface and the specification of used devices. Pictures from the drone can be planned depending on weather and terrain conditions. On the other hand, generally available satellite images are taken at specific times and under a fixed lens setting, regardless of weather and terrain conditions. It introduces additional uncertainty to the results presented by the satellite images and necessitates appropriate corrections (e.g., radiometric calibration, atmospheric correction, etc.).

RESULTS AND DISCUSSION

Coal petrography

All components present in coal wastes were divided into mineral and organic particles. Mineral particles are composed of minerals and various mineral phases and quantitatively usually dominate in coal wastes. Organic particles are composed of organic matter that was altered to various degrees (Misz-Kennan et al., 2020). With regard to various levels of alteration organic matter is further divided into three groups: unaltered (1), altered (2) and newly formed (3).

The first group in coal wastes is representing the original organic matter i.e. huminite/vitrinite, liptinite and inertinite maceral groups (Fig. 2A) showing the same optical features (colour in reflected light, reflectance, fluorescence, morphology) as unaltered macerals. For their further division the terminology described in, ICCP (1998, 2001), Sýkorová et al. (2005) and Pickel et al. (2017) was used.

In second group (altered particles), the particles showing such signs of the alteration as paler colour, paler/darker colour oxidation rims, cracks and fissures, plasticised edges and higher vitrinite reflectance (Misz et al., 2007; Misz-Kennan, 2010; Misz-Kennan and Fabiańska, 2010, 2011; Ribeiro et al., 2016; Misz-Kennan et al., 2020). Cracks and fissures occur within the particles, and their size varies from a few to a few tens of µm (Fig. 2B, C and F). Cracks can also occur perpendicular to the edges of particles (Fig. 2C). Oxidation rims that are paler with higher reflectance or darker with lower reflectance comparing to the central part of the particles occur around the edges and/or around the cracks and fissures (Fig. 2F). With prolonged oxidation, gradually, the whole particle becomes paler in colour (Fig. 2E). Some particles have rounded edges caused by the plasticity of organic matter that was a consequence of a higher heating rate. Oxidation rims can also occur around such plasticised edges (Kus and Misz-Kennan, 2017). Another form of alteration are pores caused by devolatilisation that are usually round or oval, and their size varies from a few to a few tens of µm. Their presence causes the classification of the particle as porous, while the absence of pores makes the particle massive (Misz-Kennan, 2010; Misz-Kennan et al., 2020).

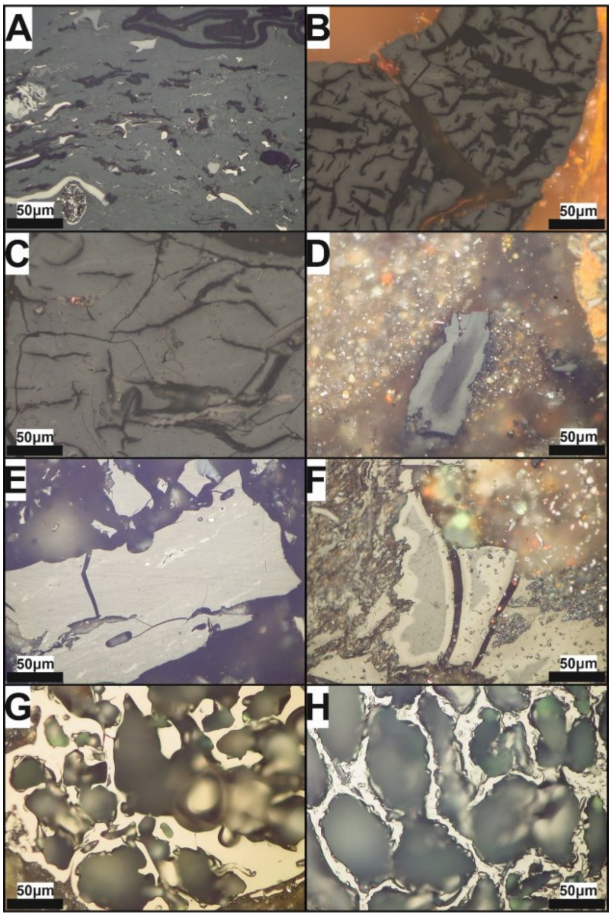


Fig. 2 Characteristic forms of self-heated coal wastes from Upper Silesian Coal Basin. A: well-preserved trimacerite particle; B – C: irregular crack with oxidation rims; D – F: thermally altered particles with paler oxidation rims; E: particle with devolatilisation and plasticised edges in paler colour caused by temperature; G – H: chars containing randomly distributed pores.

The third group contains particles representing products of low-temperature oxidation, self-heating or self-combustion. The following forms are included in this group: pyrolytic carbon forming from condensation of organic compunds previusly released from decomposition of organic matter (Kwiecińska and Pusz, 2016); bitumen being a product of expulsion of hydrocarbons from organic matter, mostly liptinite and vitrinite, during self heating, occurring as small droplets and /or thread like structures (Misz et al., 2007; Misz-Kennan, 2010; Misz-Kennan and Fabiańska, 2010, 2011) or can fill cracks and various empty spaces (Gentzis and Goodarzi, 1989; Alpern et al., 1992; Jacob, 1993; Sýkorová et al., 2018) and appearing in fluorescent with yellow – oragne to light green colour (Nádudvari and Fabiańska, 2016). Chars derived largely from vitrinite and inertinite as product of self-heating or spontaneous combustion and containing randomly distributed pores (Kwiecińska and Petersen, 2004; Lester et al., 2010). Graphite mostly formed from precipitation of carbon-saturated C–O–H fluids as product of self-heating or spontaneous combustion (Kwiecińska and Petersen, 2004). Coke formed from organic matter heated in the strongly limited access of air and hardened into coke as a product of self-heating or spontaneous combustion (Kwiecińska and Petersen, 2004; Suárez-Ruiz and Crelling, 2008).

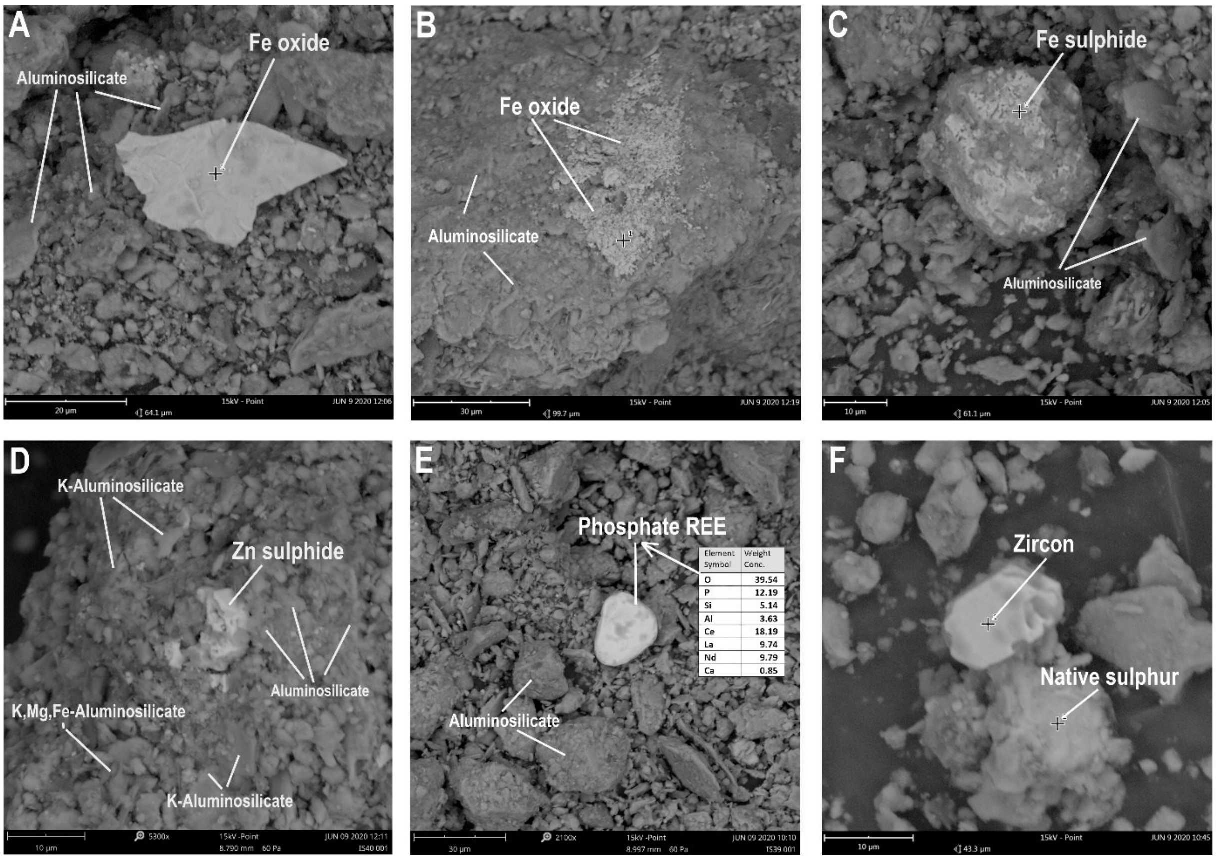
The presence of individual forms of organic matter gives an indication of the heating history of the dumps (Fig. 2). The presence of unaltered organic matter suggests that the wastes were recently deposited or were not influenced by oxidation/weathering (Fig. 2A). The paler the colour of the forms and higher reflectance indicates thermal alteration (Fig. 2D and F). Massive forms suggest that the heating rate was low, while the presence of porosity caused by devolatilisation and plasticised edges suggests a higher rate of temperature increase within the dump (Fig. 2E). Moderately altered organic matter in the wastes has irregular cracks and fissures, cracks perpendicular to the edges of particles, paler/darker in colour oxidation rims forming around the edges and/or cracks (Fig. 2B and C), and chars formed by high temperatures during self-heating – combustion (Fig. 2G and H).

Mineralogy

The mineral composition of coal wastes deposited in dumps in the Upper Silesian Coal Basin reflects the primary composition of gangue rocks and mineral matter associated with coal seams. They are quartz, clay minerals as illite, montmorillonite or kaolinite, chlorite, micas, mainly muscovite with associated dispersed organic matter. Subordinately occur Na- and K-feldspars, carbonates such as calcite, dolomite, siderite or siderite-magnesite, framboidal pyrite, and accessory zircon, monazite and xenotime. Self-heating and self-ignition processes that led to short or long-termed coal-waste fire with or without oxygen access can modify the primary mineral composition. Structures of mineral phases which are not stable at higher temperatures can be destroyed, and amorphous and metastable phases formed. Highest temperatures can lead to melting and paralava formation (Ciesielczuk et al., 2014, 2015; Pierwoła et al., 2018). Overburned parts of the dumps dominate in combusted phases as cristobalite-tridymite, celsian, anorthite, cordierite, indialite, mullite, olivine, augite, spinel group, hematite, and glass. Minerals as anatase, lead, zinc, iron sulphides and chalcopyrite occur in traces. Exhalating phases with sal ammoniac, native sulphur and K-, Mg-, Fe-, NH_4-, Al-sulphates, and chlorides are blooming around vents. Additionally, gypsum, bassanite and goethite can form due to weathering (Fig. 3-4).

Heavy metals in coal wastes are associated with silicates (especially clays), carbonates, sulphides, oxides, and phosphates (Swaine, 1994). Chalcophile elements, i.e. As, Se, Cd, Cu, Pb, Zn, Hg, are associated with sulphide minerals as chalcopyrite ($CuFeS_2$), galena (PbS), and sphalerite (ZnS) or as organically-bound species (Taylor et al., 1981; Raask, 1985; Finkelman, 1995; Monterroso and Macias, 1998). Pyrometamorphic and weathering processes influence the chemical composition of the wastes. During combustion, As, Hg, Se, and Cd can be released from sulphide minerals as these elements are highly volatile (Zhou and Ren, 1992; Querol et al., 1995; Luo et al., 2002; Głodek and Pacyna, 2009; Pirrone et al., 2010; Langner et al., 2013). According to Nádudvari et al. (2021b) the concentration of Pb, Cd, Zn, Hg, As in thermally-altered coal waste and samples rich in pyrolytic bitumen is enriched. Important to note is that clinkers, the burnt-out product of coal waste fire considered for reusage, may also contain higher concentrations of them.

Mercury in bituminous coals is mainly related to pyrite or other Fe-sulphides, whereas, in lower rank coals, it tends to be organic-bound (Kolker et al., 2006; Hower et al., 2008). Hg concentration in coals from USCB ranges from 0.1 to 0.4 mg/kg. During combustion, Hg starts to be released at 100–150°C, and ~90% of it in coal is emitted into the atmosphere (Liu et al., 2000; Guo et al., 2003; Wagner and Hlatshwayo, 2005). Therefore in self-heated coal wastes at low temperatures, Hg enrichment is not significant and ranges from 0.2 to 0.5 mg/kg (Hlawiczka et al., 2003; Abramowicz et al., 2021), whereas at high



Element Symbol	Weight Conc.
O	39.54
P	12.19
Si	5.14
Al	3.63
Ce	18.19
La	9.74
Nd	9.79
Ca	0.85

Fig. 3 SEM-BSE images of mineral phases from the thermally affected part of the Bytom dump (coordinates: 50.377516N, 18.900200E)

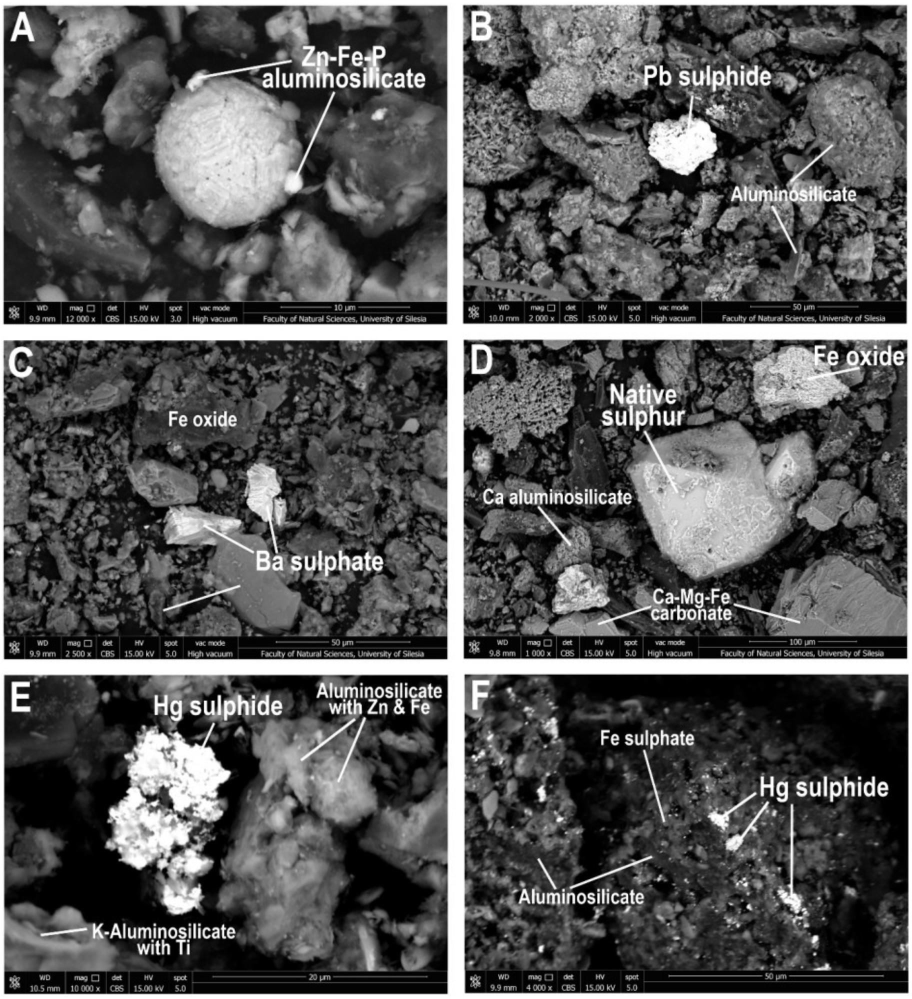


Fig. 4 SEM-BSE images of mineral phases affected by self-heating of coal wastes at the Bytom (A-E) and Czerwionka-Leszczyny (F) dumps (coordinates: 50.159672N, 18.679375E)

temperatures reaches the value of 1078 mg/kg (Nádudvari et al., 2021). In Poland, it is estimated that 24% of Hg emissions come from coal which is burnt in power plants and individual homes (Hlawiczka et al., 2003; Głodek and Pacyna, 2009).

Organic geochemistry of coal wastes

Phenols (phenol C1 - methylphenols and C2 - dimethyphenol, and ethylphenols) are typical products of self-heating dumps and are usually commonly identified in coal waste samples (Fig. 5). That reflects the thermal destruction of vitrinite via coking conditions they may represent the relatively early stages of self-heating (Skręt et al., 2010; Misz-Kennan and Fabiańska, 2011; Nádudvari et al., 2015). Especially high concentrations occur in samples containing pyrolytic bitumen or thermally affected wastes, whereas burned out coal wastes (clinker) usually contains no or small concentrations since phenols are easily evaporable by higher temperatures or leached by water (Nádudvari et al., 2015, 2016). Therefore, the identification of these toxic compounds is important around those dumps, especially in Ukraine, where a large number of burning coal waste dumps pose a serious environmental-pollution threat (Nádudvari et al., 2021a). Phenols easily dissolve in water and are prone to leaching. Potential environmental hazards of phenols are related to their toxicity; they irritate skin and cause necrosis, damage of kidneys, liver, muscle, eyes, and are carcinogenic (Clayton and Clayton, 1994; EPA, 2000).

PAHs are another significant pollutants formed by self-heating (incomplete combustion) see (Fig. 5). Usually, 2- and 3-ring PAHs dominate in these wastes (Nádudvari et al., 2021b). The majority of PAHs are carcinogenic, mutagenic, generally stable and tend to accumulate in the environment (Wagoner, 1976; Grimmer et al., 1983; Achten and Hofmann, 2009; Prus et al., 2015). In the study of Nádudvari et al. (2021b) phenanthrene, anthracene, chrysene, benzo[*a*]anthracene, benzo[*a*]pyrene, benzo[*b*]fluoranthene, and benzo[*k*]fluoranthene in many samples several times surpassed the acceptable values of Polish limits for post-industrial areas (Dz.U., 2016). Lifetime cancer risks in the vicinity of the dump are high due to the heavy metal and PAH pollution, especially when there are elevated Hg concentrations (~100–1078 mg/kg), Pb (~600–2000 mg/kg) considered. Additionally, the low pH levels (3.0–4.5) may help to mobilise these elements (Nádudvari et al., 2021b). Such coal wastes are also a source of other toxic compounds such as pyridines, quinolines, light sulphur compounds like thiophenol, benzo[*b*]thiophene or

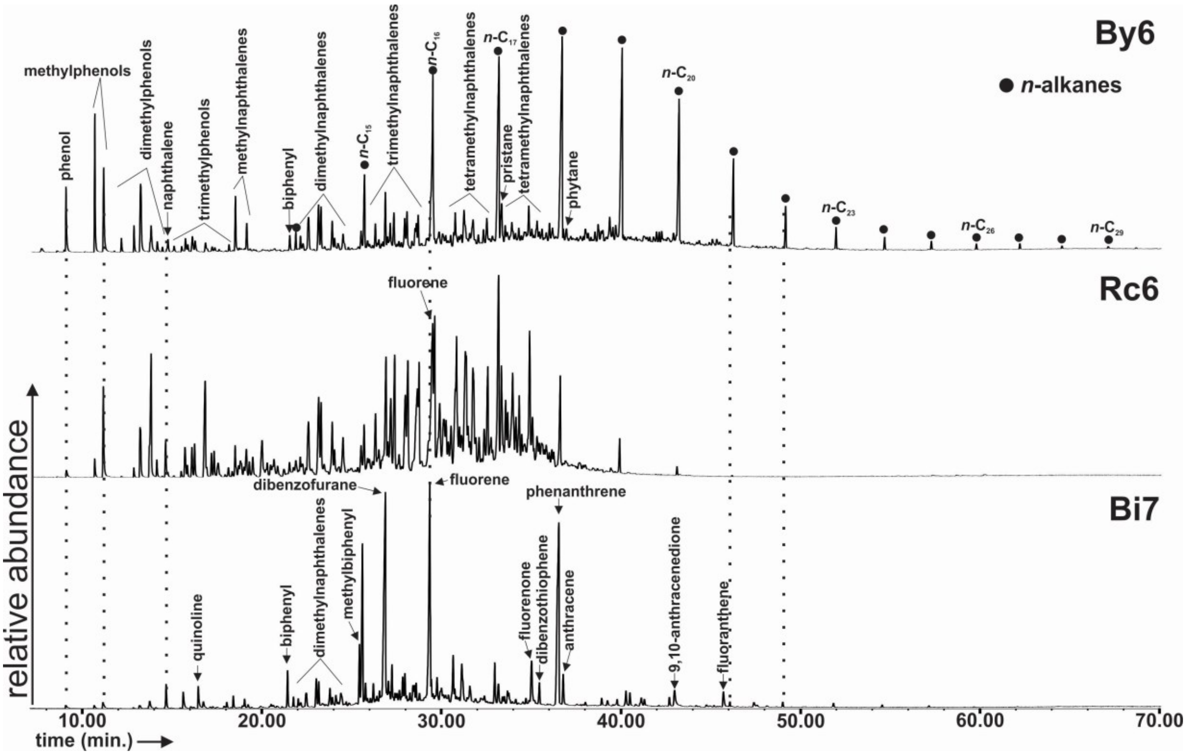


Fig. 5 Representative TIC - Total Ion Chromatograms of self-heated coal wastes in Poland. By6 (Bytom dump): thermally altered, Rc6 (Rybnik – Rymer dump): precipitated (pyrogenic) bitumen crust, Bi7 (Bytom dump): thermally altered coal waste.

more harmful chlorinated PAHs present in the bitumen (Fabiańska et al., 2014; Nádudvari et al., 2018), and also identified in emitted gases (Kruszewski et al., 2018). Chlorinated compounds formed during self-heating, e.g. dichloropropane, dichloromethane, chloroethene, chlorobenzene, dichlorobenzenes, and trichlorobenzene were previously reported by Kruszewski et al. (2018) or Nádudvari et al. (2018, 2021b). These compounds are considered persistent organic pollutants (POPs) (Nádudvari et al., 2018). These compounds, e.g. phenols, quinolines, pyridines, chlorinated PAHs are commonly identified in waste water from coking plants (Sun et al., 2018; Gao et al., 2019). Abundant naphthalene together with alkyl derivatives is characteristic for the expelled pyrogenic bitumen (Fig. 5) that lacks heavier PAHs and nitrogen heterocycles because of their higher boiling temperatures (Nádudvari et al., 2018). Also, oxidation products of PAHs can be recognised (Fig. 5) e.g. transformation fluorene to fluorenone, or anthracene to anthracenedione.

Typical characteristical feature of organic matter caused by pyrolysis during self-heating is the monomodal Gaussian distribution of short to middle chain n-alkanes (Fig. 5 - By6) and the high contents of alkylbenzenes and alkylmethylbenzenes (Nádudvari et al., 2020a). According to Radke and Willsch (1993), large quantities of n-alkanes (n-C_{15}–C_{26}), long-chain alkylbenzenes, alkyltoluenes, and benzo[b]thiophenes are produced between 170 and 360°C through the primary cracking of the coal matrix. Also, self-heating hot spots are characterised by high concentrations of emitted gases like H_2, CO_2, CH_4, C_2H, C_3H_8, C_2H_4 and with two, three times less O_2 content (Fabiańska et al., 2019; Nádudvari et al., 2020a).

Remote sensing and thermal camera monitoring possibilities

Self-heating hot spots on coal waste dumps usually appear as high-temperature surface anomalies (Figures 6 and 7). According to the self-heating intensity index (SHII), burning dumps in Figure 6 (SHII: A: 21; B: 14.5, C: 11, D: 7.5) and Figure 7 (SHII: 9) can be classified as extreme self-heating with extended high surface temperatures. The index is also suitable to describe a long-term self-heating evolution (see Nádudvari et al., 2021a). The highest surface temperatures were detected in Ukrainian dumps (Figure 6B and D) reaching 327 – 376°C. It may indicates opened fires or strong smouldering on the surface. Such surface temperature anomalies can be visible on day-time–night-time Landsat SWIR bands in case of cooling lava flows (Rothery et al., 1988; Pieri et al., 1990; Oppenheimer 1991; Nádudvari et al., 2020b) or burning coal seam fires (Chatterjee, 2006; Huo et al., 2014). The SWIR bands (Fig. 6) were counted according to Nádudvari et al. (2020b), who applied e.g. SWIR and NIR, red, green, blue, panchromatic bands for lava flow surface temperature detections. However, applying SWIR or TIR bands of night-time Landsat images for burning coal wastes similarly to lava flow temperature estimations seems to be the best way to avoid sun disturbance. The NDSI vs TIRS is also a good option for self-heating hot spot determinations (Fig. 7). Therefore the availability of night-time or snow-covered Landsat scenes always limits the application of such satellite images.

Remote sensing measurements using a thermal imaging camera are one of the possibilities of thermal monitoring of coal-waste dumps. This kind of camera

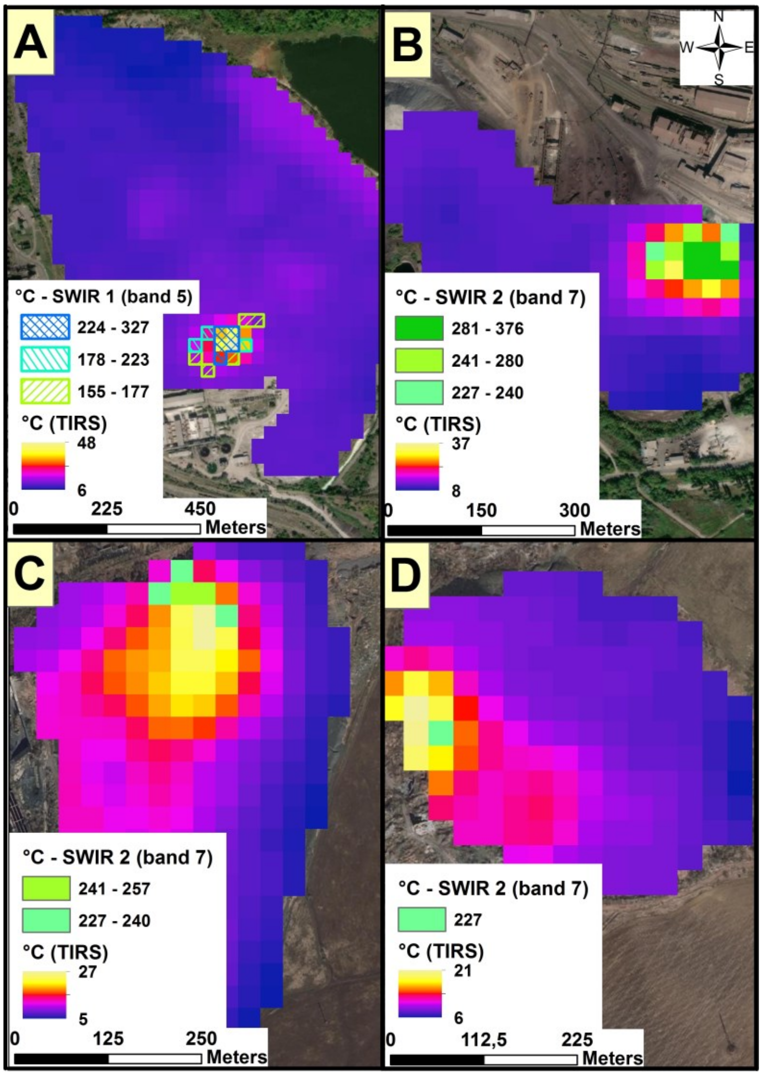


Fig. 6 Ukranian coal waste dumps from the Donetsk Coal Basin. Location of the dumps:
A: 48,225887N, 38,243042E; B: 48.213413N, 38.244355E; C: 47.999718N, 37.947060E; D: 47.985495N, 37.986345E.
The Landsat 7 ETM+, night-time satellite image acquired: 13.05.2002 – scene center time: 18:52:12.
Background map: GoogleEarth – 2021.

operates based on infrared radiation in the wavelength range from 780 nm to 1 mm. These devices provide data in the form of a raster image in which all pixels have assigned thermal values. The accuracy of measurements varies depending on the class and type of equipment and how it is used. Thermal imaging cameras can be collected in fieldwork directly from the ground or placed on an unmanned aerial vehicle (Abramowicz and Chybiorz, 2020). However, to obtain reliable results, photos should be taken in appropriate conditions - in the absence of wind, fog, rain, air pollution, and with stable air temperature and total cloud cover (alternatively before sunrise or after sunset). In the case of very diverse objects, such as dumps, using the camera should be adapted to the shape and land cover of the measured object. Using a handheld camera from the ground makes it possible to reach surfaces that are difficult to get from the air, e.g., areas covered by dense vegetation or with difficult relief. However, taking such photos is more labour-intensive and time-consuming compared to aerial photos. Photos from the air are a good way, especially in massive objects, the surface of which cannot be accessed in any other way.

Infrared images (satellite, aerial and terrestrial) enable only mapping the surface temperature of coal-waste dumps. Invasive methods should be used to measure the actual temperatures at the epicentre of a subsurface fire, including a pyrometer with a probe (Abramowicz and Chybiorz, 2019). Unfortunately, unlike camera images, measurements with a pyrometer provide only point data that require subsequent interpolation. Interfering with the body of the dump by making holes to measure the subsurface temperatures carries additional risks because such interference adds air to the interior of the dump. If higher targets do not require it, it is worth staying with the surface temperature analyses based on infrared images. On their basis, it is possible, inter alia, to the determination of the impact of a fire on the vegetation cover (Abramowicz et al., 2021b) or land development (Abramowicz et al., 2021a). Subsurface fires not only affect the place of their location but also interfere with the

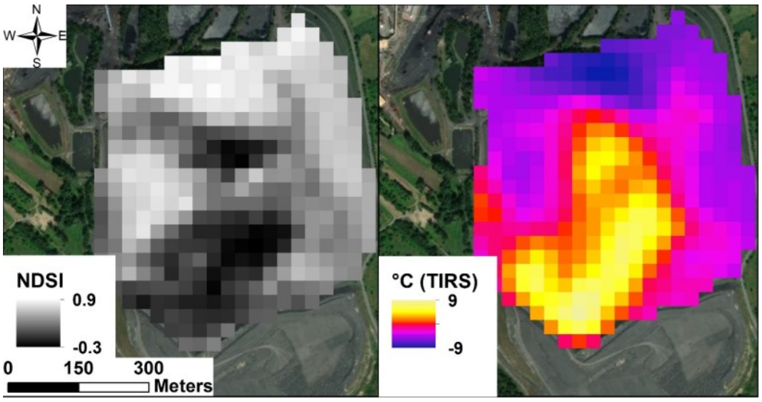


Fig. 7 An example of a self-heating dump (Szarlota) from Poland, Upper Silesian Coal Basin (coordinates: 50.062546N, 18.441930E; satellite image: Landsat 7 ETM+, data acquired: 26.02.2001 – daylight snow-covered image).NDSI - Normalised Difference Snow Index. Southern part of the dump is constructed after the image acquisition. High NDSI values indicate abundant snow cover and below 0.4 melted snow which correlating with the TIRS image. Background map: GoogleEarth – 2021.

elements of the environment directly below, above, or next to the thermally active zones, including soil, water, air, and vegetation (Homoki et al., 2000; Abramowicz et al., 2021b; Lewińska-Preis et al., 2021; Nádudvari et al., 2021a).

In Figure 8 the burning Anna dump (Poland) is presented, and the image was taken by a drone. The dump was flattened, and currently, the fire was extinguishing as the surface temperatures indicate it (<30°C). However, this dump underwent a strong self-heating on the eastern and southern part in 2010 or 2018 as detected by archive satellite images (Nádudvari, 2014, Nádudvari et al., 2021b). The drone image reveals more details, e.g. erosion gullies could promote self-heating, as it is seen on the southern part or initial stages of self-heating on the west side of the dump (Fig. 8 and 9B). Also, such fires usually are visible as cracks, line shapes (Fig. 8 and 9A-B) or extended spots (Fig. 9C) in such detailed thermal images. After burning out, blocks can erode or slide down (Fig. 9B), which may intensify the fire by more oxygen access to the deeper hot spots.

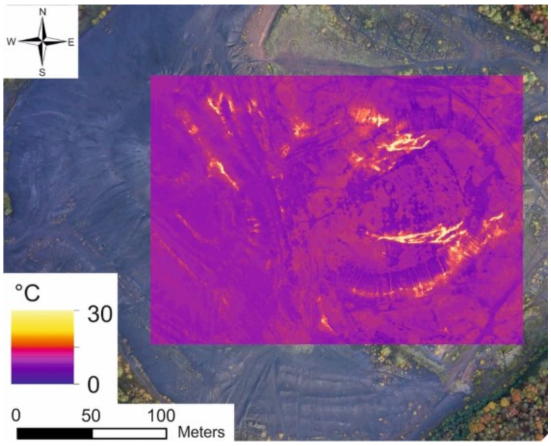


Fig. 8 Thermal image of Anna-Wrzosy dump (Poland) taken with the IR camera on a drone - resolution 20 cm (background: orthophotomap with resolution 3.5 cm). The drone operations were carried out before sunrise on 05.11.2020 in windless and rainless weather and with an air temperature of 5°C. (coordinates: 50.045445N, 18.421978E)

CONCLUSIONS

Self-heating and self-ignition processes affect poorly constructed coal waste dumps all over the world. They are uncontrolled, hard to extinguish and difficult to predict. Examples from the Upper Silesian (Poland) and Donetsk Coal Basins (Ukraine) were studied in detail to help to understand the phenomena.

Thermal alteration of coal particles causes their devolatilisation and plasticisation. Moderate heating causes the formation of irregular cracks and fissures perpendicular to the edges of particles, paler/darker in colour oxidation rims forming around the edges and cracks. Additionally, expelled pyrogenic bitumen is also a common form around self-heated particles. Oxidation rims are usually paler than the central part of the particles. With prolonged oxidation, gradually, the whole particle becomes paler in colour up to whitish.

The primary mineral matter of coal wastes is composed of quartz, clay minerals as illite, montmorillonite or kaolinite, chlorite, micas, mainly muscovite with subordinate feldspars, carbonates, pyrite, chalcopyrite and accessory zircon, monazite, apatite and xenotime. Unstable minerals under higher temperature phases rebuild their structures, releasing hazardous elements like Pb, Cd, Zn, Hg, As. These elements migrate from the hot spots and enrich in the cooler surface in e.g. sulphide minerals as HgS.

Different organic pollutants like phenols (originated from vitrinite particles), different PAHs with alkyl substitutes or oxidised PAHs, chlorinated PAHs, or sulphur heterocycles are formed. Therefore the high concentrations of PAHs and heavy metals making these coal waste dumps are potential risk for humans as they might contribute to cancer.

The introduced different remote sensing techniques helped to localise and monitor hot spots with different temperature ranges despite disadvantages of these methods. The application of SWIR bands of Landsat contribute to the successfull localisation of hot spots of extremely burning dumps in Ukraine. However, only night-time scenes with SWIR can be used, but these night-time data are rarely available and usually cloudy. The snow-covered images can be used to localise the hot spots;

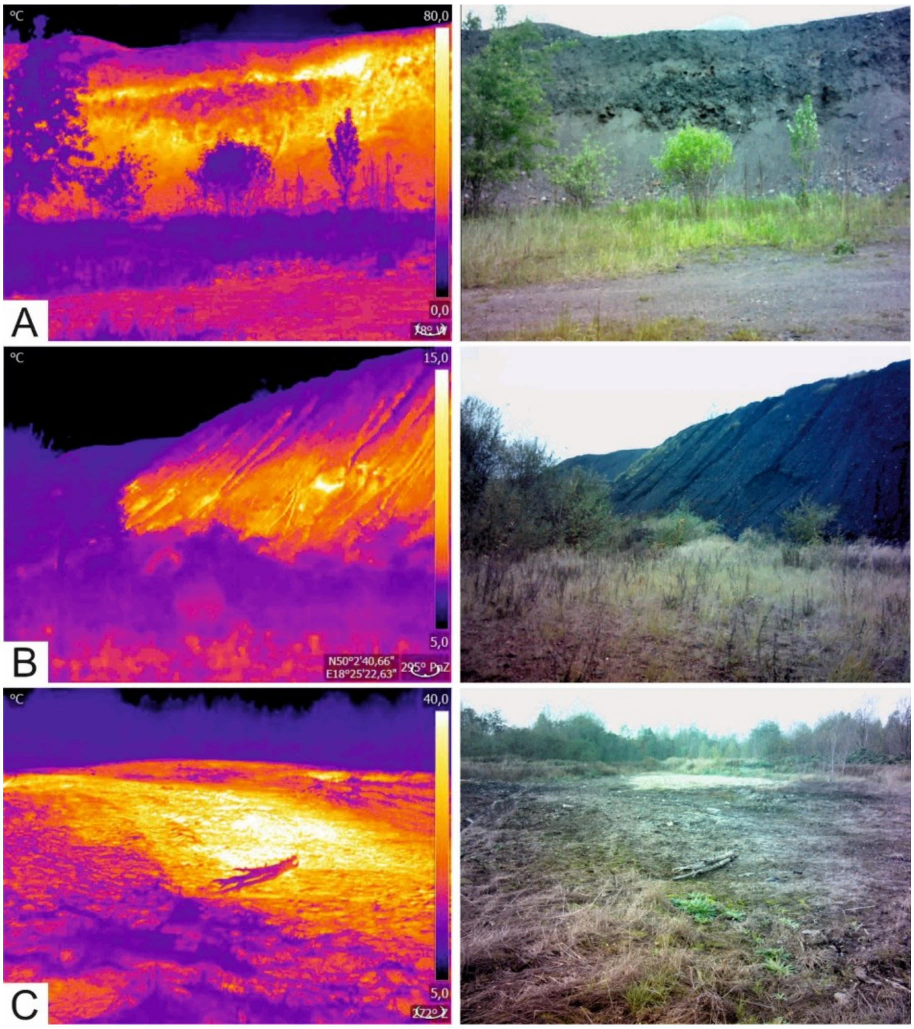


Fig. 9 Infrared photos of selected coal-waste dumps in the Upper Silesian Coal Basin (Poland): A: Bytom dump (27.05.2021, air temperature: 17°C), coordinates: 50.377516N, 18.900200E; B: Anna-Wrzosy dump in Pszów (01.11.2020, air temperature: 6°C); C: Czarny Las dump in Ruda Śląska (02.11.2019, air temperature: 10°C), coordinates: 50.280664N, 18.850826E

however, better TIRS band resolution of Landsat 7 ETM+ is more advantageous. The sun's disturbing effects should be considered as an influential factor for both thermal imaging camera or satellite images. Thermal cameras can reveal the most detailed signs of low to high temperature anomalies with different cracks and line shapes. Our research indicates the only by combining the different investigation techniques, such as applied here, the understanding of complex processes occuring within coal waste dumps can be achieved.

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