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SPECIAL FEATURE: REGIONAL CASE STUDY

Mercury cycles and their management

Mercury emission from underground coal fires: a typical case in China

Qingyi Cao[1,3] · Yingchao Cheng[2] · Taketoshi Kusakabe[2] · Yahui Qian[1,3] · Handong Liang[1,3] · Masaki Takaoka[2]

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Abstract
Mercury, a highly toxic environmental pollutant with a global circulation, must be controlled worldwide. Taking the Wuda underground coal fires, one of the most severe coal fire disaster areas in China, as a typical case, this paper systematically introduces the serious environmental output and strong environmental pollution of mercury from underground coal fires. Smoke with unusually high mercury concentrations was released from surface vents and cracks, resulting in significant enrichment of mercury in the air and surface sediments. A portion of mercury (particulate and reactive gaseous mercury) was deposited near the fire zones, but the positive high mercury fluxes of the surface soils indicated that mercury would again escape from the soil–air interface. The annual gaseous mercury emissions from the underground coal fires in China were estimated to reach 4.85 tonnes. Underground coal-fired mercury can be identified as an essential part of the global mercury cycle. Although some remediation measures were implemented, the development of coal fires proved difficult to control and was destined to be accompanied by the continuous release of mercury. Given the widespread distribution of coal fire cases worldwide, mercury pollution from underground coal fires deserves attention in the future.

Keywords Underground coal fire · Mercury · Index gas · Migration · Wuda

Introduction

Underground coal fire is a kind of environmental disaster buried in the underground coal seam that can ignite low-temperature smoldering or even high-temperature burning [1]. The burning of coal may rapidly change the climate, which may have negative consequences on the ecology [2, 3]. When the buried underground coal seams are exposed to air through rock fractures or faults, heat accumulates in the coal seams due to oxidation reactions of sulfides in coal, which then ignite as temperature rises [4, 5]. In addition, coal fires can also occur in scenarios, such as mining shafts, exposed coal seams, coal storage and transportation, and coal waste piles (gangue) [6, 7]. In geological terms, evidence from paleo-coal fires can define coal-seam fires as natural disasters. Traces of coal fires can be traced back to 4 million years ago, as evidenced by coal fires in the Powder River Basin, United States [8]. However, large-scale mining after the Industrial Revolution has dramatically exacerbated the global expansion of coal-seam fires, such as in China [9, 10], USA [1, 11], India [12], South Africa [13], and Australia [4]. The perennial combustion of underground coal fires not only causes a massive loss of coal resources, destroys the mining landscape, and subsides the bedrock surface, but also releases high concentrations of toxic gases (e.g., SO_2, CO, H_2S, F, Hg, and volatile organics), endangering the health of residents [14, 15].

Mercury (Hg) is a highly toxic persistent pollutant of global concern that is prone to long-range transportation. It has been recognized as a worldwide threat to human and environmental health [16, 17]. Hg emission from coal combustion is considered to be one of the most important sources of atmospheric Hg [18]. The assessment of Hg emission has received attention. Still, it tends to focus on anthropogenic impacts, such as the Hg emissions from coal-fired power plants and the scattered coal utilization. Hg emissions

✉ Handong Liang
hdl6688@vip.sina.com

[1] State Key Laboratory of Coal Resources and Safe Mining, Beijing 100083, China

[2] Department of Environmental Engineering, Graduate School of Engineering, Kyoto University, Kyoto 615-8540, Japan

[3] College of Geoscience and Surveying Engineering, China University of Mining and Technology (Beijing), Beijing 100083, China

from underground coal fires are generally not included in emissions inventories due to a lack of understanding or even hearing of underground coal fires. Therefore, the behavior of Hg from this source should be more examined and clarified. Hg concentration in an underground coal fire in vents from Tiptop, USA, can reach 2100 μg m^{-3} [19], which is 40 times higher than the U.S. Occupational Safety & Health Administration (OSHA) 8-h safe exposure limits (50 μg m^{-3}) [20], and much higher than the global atmospheric Hg background value (1.5–1.8 ng m^{-3}) [21]. Moreover, underground coal fire Hg is emitted almost freely, because it cannot be controlled.

Coal fires mainly occurred in the vast area of northern China, from Xinjiang Uygur Autonomous Region in the west to Heilongjiang Province in the east, stretching 4800 km longitudinally [22]. More than 50 coal fields in northern China have been affected by fires, with a cumulative burning area of ~ 700 km^2, direct burning of coal reserves of ~ 20 million tonnes per year, destruction of coal reserves of ~ 200 million tonnes per year due to operational difficulties, and harmful gas emissions of about 1.06 million tonnes per year [10, 23–25]. The Wuda underground coal fire has been burning continuously for more than 60 years and is one of the most severe coal fire disaster areas in China and even the world. Spontaneous combustion signs have been found in several upper coal seams (Nos. 1, 2, 4, 6, 7, 9, and 10), and the spread of coal fires has been a concern as the coal seams are only a few meters apart [26]. It is of urgent significance to deepening the understanding of the potential environmental impact of coal-seam fires on the ecological environment.

Hg emissions from underground coal fires have not been included in the Hg emission inventory and are a new source of emissions to the environment. As a key research topic, studies on the migration process and quantitative assessment of Hg release from underground coal fires need to be further supplemented. In this study, taking the Wuda coal fires in China as a typical case, by integrating the Hg content parameters in various environmental media, the gaseous Hg emissions from China's underground coal fires were evaluated and the migration process of Hg from underground coal fires was clarified for the first time.

Materials and methods

Case study area

The Wuda coalfield (39° 28′ N–39° 34′ N, 106° 36′ E–106° 40′ E) is located in Wuhai City, Inner Mongolia, in northern China, with a total area of 35 km^2 and an average elevation of 1150–1300 m (Fig. 1). Carboniferous–Permian coal is in the area, with coal reserves of 630 million tonnes, and the remaining recoverable coal reserves are about 190 million tonnes. The region has a typical continental arid climate with an extremely hot and dry environment. The evaporation is 3500 mm $year^{-1}$, which far exceeds precipitation (170 mm $year^{-1}$). The northwest wind prevails in the area, with an average annual wind speed of 4.8 m s^{-1}. The terrain is dominated by low mountains and hills, and with the insolation duration of ~ 3000 h $year^{-1}$ [15, 27].

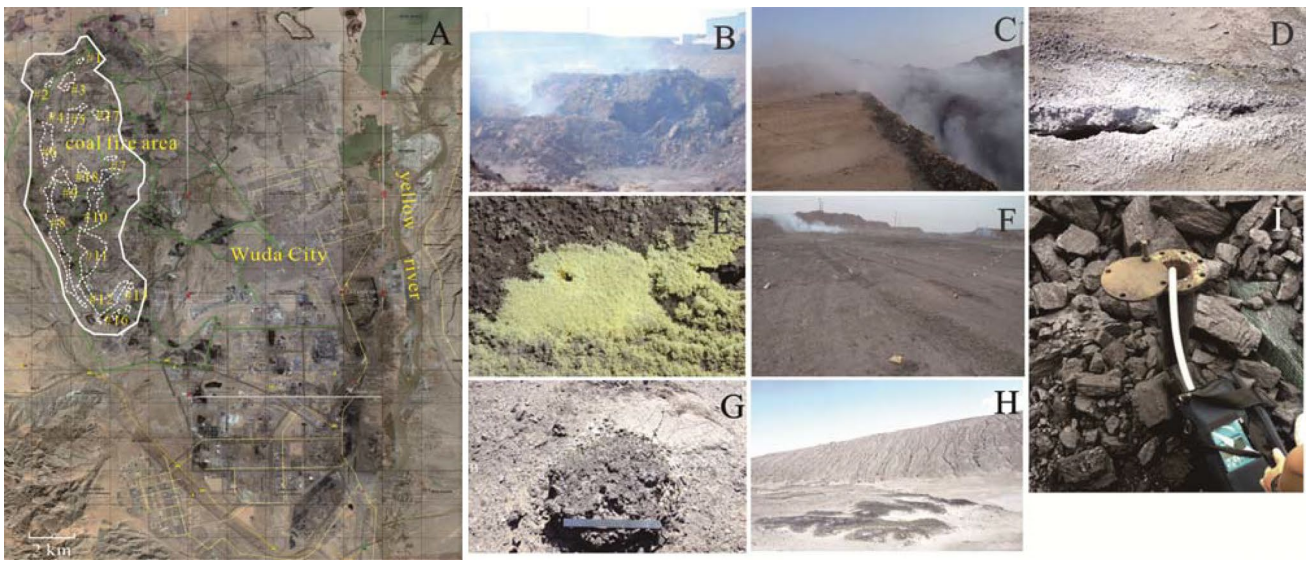

Fig. 1 Wuda coal field and surface landscape caused by underground coal fires: **A** Wuda coal field and coal fire areas; **B**, **C** surface smoke landscape; **D**, **E** smoking vents and surface cracks, and surrounding sulfur and mirabilite; **F** collection scene of surface soil; **G**, **H** coal fire sponge; **I** boreholes used to monitor underground coal fires The Fig. 1I was modified from Shan et al. [26]

The outbreak of the Wuda coal fire can be traced back to 1961, when the coal seams of No. 9 and No. 10 in the Suhaitu minefield took the lead in the spontaneous combustion of coal seams. Since then, the development of coal fires has intensified, from 6 surface fire areas in 1978 to 26 surface fire areas in 2004, with a total area of about 4 km^2 [28, 29]. In 2009, Shenhua Remote Sensing Exploration Co., Ltd. re-investigated the area covered by the Wuda fire, which covered an area of 4.86 km^2. Since 2009, the local government has vigorously curbed the development of the Wuda coal-seam fire and extinguished the visible surface flames. However, despite the staged governance results achieved in the distribution of the Wuda fire area from discrete to concentrated, the development trend that the fire area rebound is a cause for concern. Surface vents and fissures, with or without smoke, are scattered throughout the coal fire zone, and heavy smog in the near-surface air can be observed year-round.

Collection of index parameters in boreholes

To monitor the development of coal fire in a specific coal seam between the main roadway (80–120 m underground) of the Suhaitu minefield and the ground, and to ensure the safety of underground transportation, the borehole monitoring project was implemented. The function of setting the boreholes was to collect the gas in the borehole for component analysis, and then evaluate the underground coal fire and its development and corresponding countermeasures. The drilling depth is 13–42 m, the diameter is 10 cm, and there are 44 drilling holes in total. Most of the boreholes were set in the area where the coal seam had not been mined (the depth is 1 m from the coal-seam floor rock), and several drilling holes were set in the Goaf. These boreholes were equipped with steel pipes with air inlets on the pipe walls to allow the inflow of gas released from the coal seam, while the near-surface outlet ends of the steel pipes were sealed with steel caps to isolate air exchange [30]. Gas acquisition and on-site tests were performed after opening the steel caps.

Results and discussion

Hg release from Wuda underground coal seams

The Hg content in the raw coal of the Wuda coalfield has been evaluated many times. For example, reports from Wang [31], Hong [30], and Li [32] et al. indicated that the average contents were 261 ng g^{-1} (112–450 ng g^{-1}, $n=4$), 227 ng g^{-1} (48–589 ng g^{-1}, $n=30$), and 317 ng g^{-1} (273–346 ng g^{-1}, $n=7$), respectively. Thus, the average Hg content in the raw coal can be estimated to be 246 ng g^{-1}, which is lower than the average Hg concentration of Chinese coal (290 ng g^{-1}) [33]. In addition, Wang et al. [31] tested the average Hg content of burned coals in the Wuda underground coal seam to be 14 ng g^{-1} (9–24 ng g^{-1}). These data are obtained by comparing the Hg content of raw coal—about 94% of the Hg was released due to coal fires.

Due to coal fires, the buried coal seams provided a continuous Hg source, and gas composition analysis data from the borehole monitoring project can reveal this hidden behavior. The average Hg concentration was 637 ng m^{-3} ($n=144$) at the drill holes in the #2 fire zone, 3694 ng m^{-3} ($n=432$) in the #3 fire zone, 1095 ng m^{-3} ($n=144$) in the #4 fire zone, 4481 ng m^{-3} ($n=396$) in #5 fire zone, and 6364 ng m^{-3} ($n=468$) in #9 fire zone, respectively. The overall mean Hg concentration was 4165 ng m^{-3} (34–62,513 ng m^{-3}, $n=1584$), which was 45 times higher than the concentration measured in the near-surface atmosphere around the boreholes, indicative of high Hg released from the boreholes [30]. Nine boreholes in the #10 fire zone were used to monitor the dynamics of the No. 7 coal-seam goaf. Because most of the coal has been mined, gaseous Hg concentrations were relatively low, ranging from 5.69 to 158.19 ng m^{-3}, with an overall average of 49 ng m^{-3} (detected in July, September, and October 2018) [25]. The high Hg concentrations in the boreholes can indicate the smoldering of underlying coal seams.

Hg pollution caused by Wuda underground coal fire

Atmospheric Hg pollution

Gaseous mercury The most direct and significant Hg emission channels for coal fire areas are smoking vents and surface cracks. These channels transport underground coal-fired Hg into the air, diffusing and advecting into the surrounding area. The Hg concentration in the fumes from smoking vents and surface cracks at 30 spots in the central area is 200–1350 ng m^{-3}, with an average Hg of 464 ng m^{-3} [34]. The temperature of smoking vents and cracks' surface were mostly 150–280 °C, with several exceptions exceeding 300 °C. The extremely high concentration values and high-temperature data obtained from smoking vents and surface cracks undoubtedly demonstrate the vigorous development of underground coal fires, accompanied by uncontrolled Hg emissions. In addition, coal gangue piles stacked on the surface were also burning, and the average Hg concentration in the vents reached 5908 ng m^{-3} (1022–31,750 ng m^{-3}) [35]. The resulting consequences are doomed to high levels of Hg in surface air. The Hg content in near-surface air in the coal fire central area was 257 ng m^{-3} (211–375 ng m^{-3}), and that in the peripheral area was 89 ng m^{-3} (23–211 ng m^{-3}) [34]. This result reveals that underground coal fires can lead to severe Hg pollution, as evidenced by Hg monitoring data from coal fire area vents in the Wyoming coal fires,

Powder River Basin, USA (12,100 ng m^{-3}) [36], Kentucky coal-seam fires, USA (7000–610,000 ng m^{-3}) [37], and Witbank and Sasolburg coal fires, South Africa [38]. Furthermore, atmospheric Hg concentration in the downwind urban area reached 33 ng m^{-3}, which was much higher than previously reported data in other cities or regions, e.g., Guiyang (9.72 ng m^{-3}) [39], Guangzhou (5.4 ng m^{-3}) [40], and Changchun (18.4 ng m^{-3}) [41] in China, as well as Kagoshima City in Japan (3.5 ng m^{-3}) [42].

At the same time, surface Hg fluxes were potentially and continuously occurring in an invisible form. A dynamic flux chamber method [43] was used at the Wuda coalfield to measure Hg flux in the surface soils of the fire zone. The measurement principle is to determine whether Hg is emitted or deposited and the corresponding flux, according to the difference between the outlet Hg concentration and the inlet Hg concentration (the Hg concentration in the ambient air) of the flux chamber within the delineated surface soil area. The Hg fluxes in the #3 fire zone were 76–174 ng m^{-2} h^{-1}, with an average value of 99 ng m^{-2} h^{-1}. The Hg fluxes in the #6 fire zone were 80–318 ng m^{-2} h^{-1}, with an average value of 177 ng m^{-2} h^{-1}. The no-fire area, located upwind of #3 and #6 fire zones, had lower values of 4–29 ng m^{-2} h^{-1} and 14–62 ng m^{-2} h^{-1}, respectively, with an average of 19 and 32 ng m^{-2} h^{-1} [44]. These Hg flux values were positive, indicating that the soil was transporting more Hg to the air than it was deposited. In addition, the significant differences in flux values across the different zones indicated the inhomogeneity of Hg fluxes and potential effects from underground coal fires. Based on data available for comparison, the exchange flux of Hg between soil and air in the Wuda fire area was much higher than in many sites, such as forest areas (− 2.5 to 27.2 ng m^{-2} h^{-1}) [45], wetlands (~ 3.5 ng m^{-2} h^{-1}) [46], landfills (~ 20.0 ng m^{-2} h^{-1}) [47], Hg mines and volcanic areas (17.1 ng m^{-2} h^{-1}) [48], bare soil (6.5 ± 0.2 ng m^{-2} h^{-1}) [49], urban areas (7.8 ± 7.1 ng m^{-2} h^{-1}) [50], forests (~ 2.2 ng m^{-2} h^{-1}) [51], and grasslands (1.0 ± 0.7 ng m^{-2} h^{-1}) [52].

Particulate mercury Generally, gaseous elemental mercury (GEM) is the primary form of atmospheric mercury, and atmospheric particulate mercury (PHg) often contributes less than 10% to the total atmospheric mercury [53, 54]. The regional sedimentation and water solubility of PHg are stronger than that of GEM [55], and it can enter and permanently damage the human body through inhalation, dietary consumption, and skin exposure [56]. By collecting TSP samples from the Wuda fire area, the particulate mercury (PHg) content in the near-surface air was determined to be 25–45 ng m^{-3}, with an average content of 33 ng m^{-3} [57]. This value was also rare and much higher than previously reported data such as Beijing (1.18 ng m^{-3}) [58], Shanghai (0.43 ng m^{-3}) [59], and Changchun (0.02–1.98 ng m^{-3}) [41]. PHg in Wuda coal fire area may exist as inorganic forms, such as $HgCl_2$, HgS, HgO, and $Hg(NO_3)_2 \cdot H_2O$, and predominantly adheres to fine particulate matter (≤ 2.5 μm) [57]. Hg^0 is known to convert to PHg under certain oxidative conditions [60, 61], but the formation process of PHg in coal fire zones has not been elucidated.

Hg contamination of surface sediment

In the Wuda coalfield, the Hg concentration was 289 (11–765) ng g^{-1} ($n = 11$) in the dustfall (upper ~ 1.5 mm of the ground surface) and 216 (15–970) ng g^{-1} in the surface soil (upper ~ 20 cm of the ground surface) [32]. Concentrations of Hg in topsoil and dust are lower than those of some mercury-contaminated metal mines, such as the Zarshuran gold mine in Iran (24,200 ng g^{-1}) [62], Almaden Hg mine, Spain (6000–8,889,000 ng g^{-1}) [63], Phichit gold mine, Thailand (210–20,960,000 ng g^{-1}) [64]. On the vertical profile of 0–30 cm, the soil Hg content decreased with the increase in depth, and was most enriched in the top layer [31]. This suggests that Hg deposits originating from surface flue gas vents and fissures are the source of Hg in the surface soil of the coal fire area. Meanwhile, the strong correlation between Hg and total carbon content observed in the soil also suggests that organic matter released from underground coal fires is pooled to the surface along with Hg. Soils in the downwind direction had higher levels of Hg, with concentrations around ten times higher than those in the windward direction.

Sulfur distributed near cracks and vents can have Hg levels of 900–3000 ng g^{-1}. Coal Fire Sponge (CFS) [65], a "cow dung-like" raised coal fire derivative distributed on the surface of the coal fire area, with strong acidity and strong corrosiveness. It appears to be present on the surface of coal fire zones in different countries. Under each sponge is a vent hole of an underground coal fire [4, 66], but it looks like a raised lump of soft clay. The CFS in the Wuda fire area was enriched in acid, sulfur, and F, with the average parameters of pH, HF, and SO_4^{2-} were 2.06 (0.61–3.84), 16.65 ppb (6–38 ppb), 188 mg g^{-1} (112–387 mg g^{-1}), respectively [67]. CFS samples from the #6 and the #8 fire zones were tested ($n = 73$), and the Hg content in CFS ranged from 2653 to 38,470 ng g^{-1}, with an average value of 13,967 ng g^{-1} [68]. The composition of CFS is not clear, but it exhibits a strong trapping and adsorption effect on mercury.

Migration of Hg from underground coal

Once it has entered the environment, Hg cycles between the air, land, and water, until it is eventually sequestered from the system through the processes, such as burial in deep ocean sediments or lake sediments [69]. Calculating the release of Hg from coal fires is a topic that needs to be

explored, which can then be incorporated into an assessment system for the global Hg cycle. Therefore, it is necessary to understand the whereabouts of coal fire mercury.

Hg in coal-fired flue gas exists mainly in elemental (Hg^0) and particulate (PHg) forms, and may also exist in small amounts in reactive gaseous mercury (RGM) [39, 70]. Among them, RGM is easily dissolved and transformed, and can be quickly removed from the atmosphere through wet and dry deposition processes. On the contrary. Hg^0 is insoluble and stable, and its atmospheric lifetime can reach 1 year [71]. The existence lifetime of PHg in the atmosphere is longer than that of RGM, but much lower than that of Hg^0. Due to the extremely low rainfall in the Wuda region, there are few opportunities for Hg to be wet deposited, and dry deposition is usually the main driving force for Hg accumulation in the soil (Fig. 2). Hg from underground coal fires, although CFS fixes a portion of it (probably PHg and RGM) in the form of HgO and $HgSO_4$ [67], is much lower than that released into the air. Subsequently, RGM and PHg are readily deposited in the mining area and surrounding areas under dry deposition. However, their migration did not end there. Based on the positive high Hg fluxes exhibited by the surface soils of the coal fire areas, it is suggested that the Hg deposited on the surface is re-emitted to the atmosphere. This process is carried out with the participation of photoreduction, which realizes the gasification of Hg in the soil in the form of Hg^0 [72]. Hg is released from the Wuda coal fires to surrounding areas by diffusion and advection, ultimately participating in the global Hg cycle.

It is worth mentioning that the urban area of Wuda is located 5 km downwind from the coal fire area. Therefore, this means that Hg from the coal fire area will be preferentially transported to the urban area and potentially threaten residents' health. Further speculation is that the Wuda urban area, with a population of ~ 130,000, may have suffered from coal fire Hg contamination for decades. The relationship between the Wuda coal fire and the adjacent urban area is just one case, and more extensive cases are distributed worldwide.

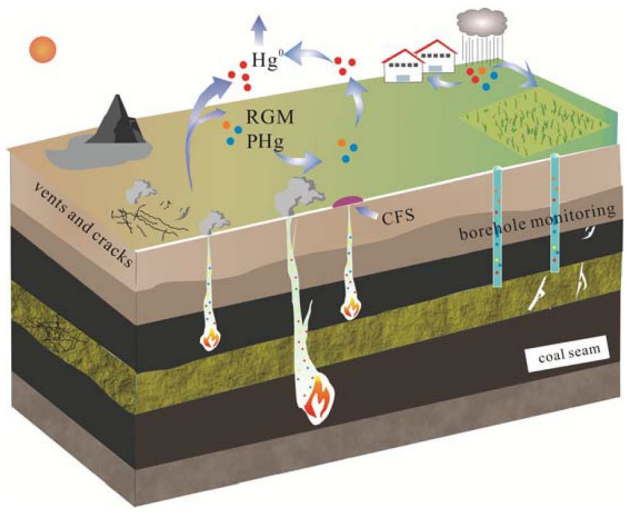

Fig. 2 Generalized diagram of the Hg migration process from the underground coal fires

As China's most deeply researched coal fire area, the data indicators obtained from the Wuda coalfield can provide a reference standard for evaluating the Hg emissions from underground coal fires in China. Due to the more vital migration ability of gaseous Hg, atmospheric Hg output from underground coal fires was primarily estimated based on gaseous Hg releases (formula 1). The values of M, C_a, C_b, C_0, P_a, and P_b were determined to be 2×10^7 [10, 25], 246, 14 [31], 290 [33], 257 [34], 33 [57], respectively. The results show that the annual Hg emission from underground coal fires in China is 4.85 tonnes, accounting for 2.4% of the conventional coal combustion emissions (202.3 tonnes) [73], which exceeds the annual Hg emissions from fuel combustion in Australia, New Zealand & Oceania Emissions (3.57 tonnes) [74].

$$Q = 10^{-9} * M * \frac{(C_a - C_b)}{C_a} * C_0 - \frac{P_a}{(P_a + P_b)}, \quad (1)$$

where Q is the annual emission of gaseous Hg (t); M is the annual burning loss of raw coal (t); C_a is the Hg content in the raw coal (mg kg^{-1}); C_b is the Hg content in the burned coal (mg kg^{-1}); C_0 is the Hg content in Chinese coal (mg kg^{-1}); P_a is the gaseous Hg concentration in the near-surface (ng m^{-3}); P_b is the PHg concentration in the near surface (ng m^{-3}).

Uncontrolled Hg release from underground coal fires

From 2006 to 2008, a surface block excavation method was adopted in the Wuda fire area to eliminate the coal fires. The original idea was to dig out the burning coal, backfill it with soil, and lay the surface with a hardener. However, in hindsight, it turned out to be an extremely unsuccessful project. This action led to the accelerated spread of coal fires and the destruction of almost all surface vegetation in the mining area, due to the failure to properly assess the difficulty of coal fire control [70]. Recently, governance measures have changed. The liquid nitrogen ejection project for the underground coal formation has been carried out several times in the Wuda coal fire area in an attempt to curb the further development of the coal fires. Figure 3 shows the variations of index parameters in boreholes 17-7-9 (Fig. 3a) and 17-7-9 (Fig. 3b) from the #10 fire zone before and after the liquid nitrogen ejection, including CO, Hg, and temperature (T). The execution dates of liquid nitrogen ejections were

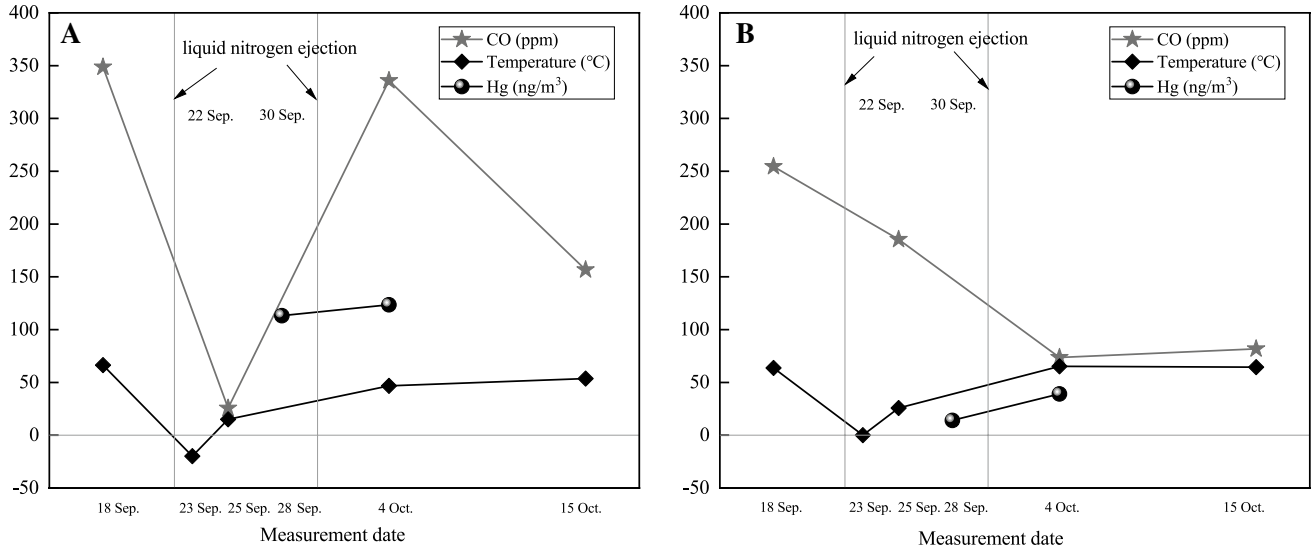

Fig. 3 Variations in borehole temperatures, CO, and Hg concentrations before and after the ejection of liquid nitrogen. Data were compiled from Shan et al. [26]

September 22 and September 30, 2018, and both injections reached 11.3 tonnes. It was observed that after the liquid nitrogen injection was performed on September 22, the CO concentration and *T* were significantly lower (based on the data on September 23 and 25) than those before ejection (the data on September 18), where the T even reached < 0 °C. However, after the ejection was performed again on September 30, although the changing trend of CO concentration in different boreholes diverged, the Hg concentration and *T* showed a rebounding trend. Judging from the evolution of temperature, implementing the liquid nitrogen ejection project can only briefly suppress the development of underground coal fires, but the rebound trend also followed. Since Hg vaporization is inherently highly controlled by temperature rather than atmospheric conditions, Hg release can proceed normally, even in an oxygen-free atmosphere. Therefore, even if no high-frequency and continuous Hg concentration monitoring data were acquired during the project implementation, the temperature-based rebound trend suggests that the coal-fired Hg release will continue.

Hg: a potential index gas of underground coal fires

Dynamic monitoring of underground coal fires is important in coal fire prevention and control. As a conventional high-precision geochemical detection method, indicator gas measurement plays an essential role in underground coal fire monitoring and early predicting potential coal fire disasters. According to the generation cause, coal fire indicator gases can be divided into three categories, including oxidizing gases (e.g., CO, CO_2), alkene gases (e.g., C_2H_2, C_2H_4, C_2H_6, C_3H_8), and special gases (radon) [24, 75–78]. In practice, CO is usually used as the first choice indicator gas due to its early generation. A large amount of production, fast production speed, and the indicator are economical and effective [79, 80].

Some potential coal fire indicator gases, such as C_2H4, C_2H_2, C_3H_6, and C_2H_6, cannot be applied in monitoring the Wuda underground coal fires. Because C_2H_4, C_2H_2, and C_3H_6 were not detected in the borehole gas, C_2H_6 was detected in only half of the boreholes and at lower levels (Table S1). The monitoring data of gas emitted in boreholes from areas with strong underground coal fire development (#2, #3, #4, #5, and #9 fire zones) show that the detected concentrations of Hg and CO were 34–62,513 ng m^{-3} and 0.006–0.594% [30]. The ratio of the highest detected concentration to the lowest detected concentration was 1838 times and 99 times, respectively, which indicates that the resolution of Hg is much greater than that of CO. Interestingly, field monitoring data from a wide zone range (#2, #3, #4, #5, #9, and #10 fire zones) showed that Hg release content from underground coal fires was significantly correlated with CO release content, with the R^2 of 0.78 (Fig. 4a). Similarly, the correlation between Hg and CO was reproduced in monitoring data from the Wyoming coal fires, USA (Fig. 4a). Furthermore, Hg release was more responsive to temperature than CO, with a higher correlation coefficient of $R^2 = 0.74$ (Fig. 4b). This is due to the fact that the Hg release process is highly controlled by its speciations and temperature. The Hg in Wuda coal mainly occurs in the speciation of HgS [81]. The central temperature of thermal decomposition for HgS is 310 °C [82]; that is, before reaching this temperature, the release concentration of Hg keeps rising. Even in an oxygen-free atmosphere (e.g., nitrogen and

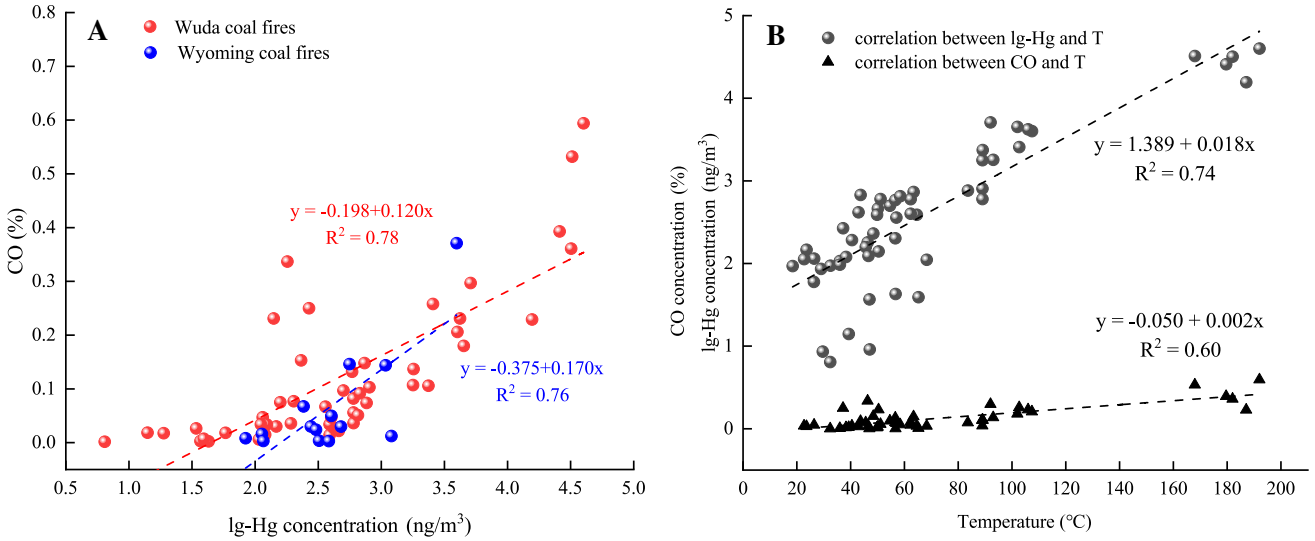

Fig. 4 Correlation analysis of Hg–CO, Hg–T, and CO–T The data of the Wuda coal fires were compiled from Shan et al. [26] and Hong et al. [30]. The data of the Wyoming coal fires were compiled from Engle et al. [36]

argon), Hg release can still occur. Due to the volatile nature of Hg, it is also applied in some important geological fields, such as earthquake prediction, detecting oil and gas, and searching for geothermal resources. In conclusion, Hg, as a common gas released from coal fires, has the potential to act as an indicator gas to monitor the development dynamics of underground coal fires due to its temperature sensitivity and high resolution.

Conclusions

This study aims to analyze the environmental impact, migration process, and release amount of Hg from underground coal fires, based on the integrated analysis of the observed data. Taking the Wuda underground coal fire area as an example, smoke with abnormally high Hg concentrations was released from surface vents and cracks, resulting in significant enrichment of Hg in the air and surface sediments. Particulate and reactive gaseous Hg tended to deposit near the fire zones. However, Hg would again escape from the soil-air interface, as indicated by the positive high Hg fluxes of the surface soils. Estimated 4.85 tonnes of gaseous Hg per year originate from underground coal fires in China and attempt to participate in the global Hg cycle. In addition, Hg, a common gas released by coal fires, has the potential as an indicator gas for monitoring the development and dynamics of underground coal fires due to its temperature sensitivity and high resolution.

Drawing on the governance experience of underground coal fires in Wuda, it is not easy to achieve the ideal effect by adopting the methods of isolating oxygen and cooling the coal seams intermittently. This is plagued by factors such as the complexity of the geological structure, the temperature rebound of the coal seams caused by the high-temperature surrounding rock, and the expensive engineering treatment cost. In the future, the control of underground coal fires should be regarded as one of the environmental protection causes that the world should jointly deal with. Given the widespread distribution of coal fire cases worldwide, Hg emissions from underground coal fires can be considered the essential strongholds for global Hg warehouses. Further assessment of global Hg emissions from underground coal fires is recommended.

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Data availability The data used in this article are available from the corresponding author upon reasonable request.

Declarations

Conflict of interest The authors declare that they have no conflict of interest.

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