SOME TRACE ELEMENTS IN COAL OF THE CZECH REPUBLIC, ENVIRONMENT AND HEALTH PROTECTION IMPLICATIONS

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SUMMARY

Mining for coal and its utilization have various impacts on the surrounding environment. Huge volumes of waste materials which are by-products of both the underground and open cast coal mining, pose one of the major environmental hazards in addition to air pollution caused by coal burning in power plants in the Czech Republic. Some of these risks could be reduced when having accurate and comprehensive data on coal quality. Statistical data processing of almost 35,000 coal samples from Late Paleozoic and Tertiary coal basins of the Czech Republic provided a unique information on the quality of lignite, sub-bituminous and bituminous coals and anthracites including the content of toxic trace elements (As, Be, Hg, Pb and Se). In this context related environment and health risks and protection implications are discussed.

Key words: Czech coal, arsenic, beryllium, lead, mercury, selenium

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INTRODUCTION

The negative impact of the energy production on the environment in the Czech Republic resulted from unfavorable structure of primary energy sources: 63 % of electric energy were generated in thermal power plants, mainly from coal, in 2003. As a matter of fact this is a certain progress since it was more than 90 % in early 1960s. About 34 % of electric energy were generated in 2003 in nuclear power plants, 2 % in hydro-energy plants, and 1% came from wind, solid biomass, industrial and municipal waste burning and biogas in 2003. Bituminous coal has been burnt in only two power plants because it was mostly used for coke production. On the other hand, lignite represents the major source for energy generation. Tertiary lignite deposits are distributed all over the territory of the Czech Republic. However, besides small deposits of local importance there exist three major areas of lignite mining: North Bohemian Basin, Sokolov Basin, and South Moravian coal field (Fig. 1). Bituminous coal and anthracite are confined to Late Paleozoic coal basins of various size. Their most important deposits occur in the Czech part of the Upper Silesian Basin, Central and West Bohemian basins, Intra Sudetic Basin, Blanice Graben, and Boskovice Graben (Table 1). The latter two areas are much less important. Lignite reserves of the Czech Republic will last for about following 35 years, in the case if our government cancel the regional and environmental limits for about the whole 21st century.

Following the year 1990, all large power plants in the Czech Republic were desulfurized, which resulted in reduction of SO_2 emissions by ca 90%. However, some other harmful substances are still escaping into the atmosphere during the coal firing in power plants even when applying the Clean Coal Technology.

For instance, during burning of lignite in fluidization furnace (350 $t.hr^{-1}$) about 80% of Hg, 5–10% of selenium and 70% of chlorine are released (1). But on the other hand 6% of arsenic, 20% of mercury and 50% of selenium were released into atmosphere during the burning of anthracite from the Donets Basin (2). The heavy metals in coal during its firing concentrate on particles of atmospheric aerosol forming stable compounds, which remain for long time in the atmosphere and can be transported at a long distance (3). The present paper is a survey of several major studies,

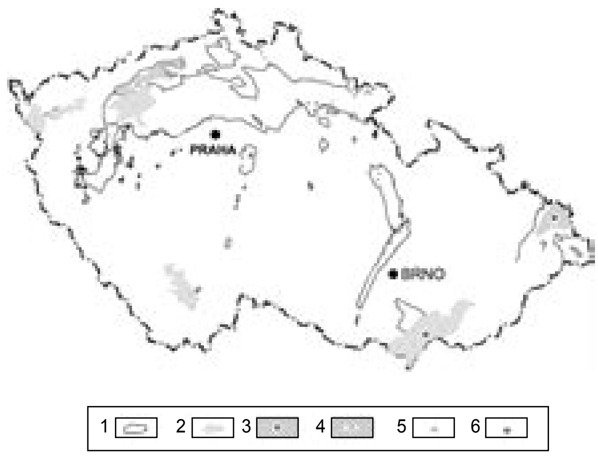

Fig. 1. *Distribution of bituminous and lignite deposits in the Czech Republic.*
1 = outline of Permo-Carboniferous bituminous coal basins, 2 = outline of Tertiary lignite deposits without or with unimportant mining activity, 3 = coalfield with several active bituminous deep coal mines, 4 = coalfield with several active lignite open-cast mines, 5 = coalfield with an active open cast mine, 6 = coalfield with an active deep mine.

Table 1. Rank and utilization of coal in the Czech basins

Basin	Rank	Utilization
North Bohemian Basin (NBB)	lignite to sub-bituminous coal	energy production, home heating
Sokolov Basin (SoB)	lignite to sub-bituminous coal	energy production, home heating
South Moravian coal field (SMCF)	lignite	energy production, home heating
Central Bohemian and West Bohemian basins (CWBB)	high volatile bituminous coal	mining was terminated
Czech part of the Upper Silesian Basin (CUSB)	bituminous coal to anthracite	coke production, energy production, home heating
Intra Sudetic Basin (ISB)	high to medium volatile bituminous coal	mining was terminated
Krkonoše Piedmont Basin (KPB)	high volatile bituminous coal	mining was terminated
Blanice Graben (BlG)	anthracite	mining was terminated
Boskovice Graben (BoG)	low volatile bituminous coal to anthracite	mining was terminated
Brandov relic (BrR)	anthracite	mining was terminated

Table 2. Arsenic content in the Czech coal (ppm)

Basin	Range	Average	Number of samples
NBB	0–1,101	40	9,172
SoB	0–2,020	333	13,339
SMCF	4–194	28	437
CUSB	0–1,220	133	441
CWBB	0–1,262	177	186
ISB	9–109	55	5
KPB	150–431	204	12
BlG	78–198	138	3
BoG	–	220	1
BrR	–	1,900	5

For explanation see Table 1

supplemented partially with unpublished data, and also providing results of the recent study of sulfur and selected trace elements in the Czech coals in the environment and health context.

SOME TOXIC TRACE ELEMENTS IN COAL

Coal contains most elements of the periodic table. Data on potentially dangerous elements are urgently needed when assessing environmental impacts of coal utilization. Trace elements in coals are proposed in five groups according to the degree of possible risks. In this study we focused on elements of the greatest concern, i.e., As, Be, Hg, Pb and Se in bituminous coals and lignites of the Czech Republic and their impact on the environment, particularly when burnt in local power plants. Cadmium was not considered because of insufficient number of analyses.

Arsenic

Arsenic belongs to elements of which higher concentrations in the burnt coal considerably adversely affect living organisms. Contents of this element vary from < 0.1 up to a few hundreds of ppm (4). The values exceeding ~80 ppm As are considered to be unusual in coal used for power production. However, reported maximum concentration of arsenic in coal ash is supposed to have been equal to 8,000 ppm (5). Its concentrations are usually low in low-sulfur coals consistent with the occurrence and amount of sulfides (6) and other authors report a significant affinity of As to sulfides such as pyrite and very rare arsenopyrite. In coal of various age rich arsenic besides pyrite and arsenopyrite also sulphates and clay minerals containing As were identified (7). They also found out that arsenic occurs in coal as As^{5+} and As^{3+}, which form organometallic compounds. Electron microprobe studies of the Czech coals high in sulfur and arsenic brought similar evidence of As being confined to framboidal pyrite/marcasite, aggregates and individual grains. Arsenopyrite was not identified in these coals in contrast with coal of the Nováky Basin in Slovakia.

The content of As was studied mostly in basins whose coal seams were extracted or are still being mined. With the exception of the Sokolov Basin (Table 2) majority of coals should be in the range 26–204 ppm. The highest concentration of the element was detected in coal ash of the Josef seam. For this reason the extraction of the coal seam was stopped in 1990s. Lignite with 900–1,500 ppm As was burnt in a power plant in the Slovak part of the former Czechoslovakia. In this particular case, despite the use of electrostatic eliminators, about half a ton of arsenic was emitted daily during the first decade of operation, according to rather conservative calculations. A considerable portion of the emissions was in the form of arsenic trioxide contained in fine fraction of the solid phase of the emissions, mostly condensed on the surface of fly ash particles.

Ecological aspects: Excessive contamination of the environment by arsenic has resulted in the extinction of honeybee colonies to the distance of up to 30 km in the direction of the prevailing winds from the Nováky power plant in central Slovakia. Increased concentrations of arsenic were detected in surface waters in the region, especially after the collapse of the dam containing waste ash from the power plant. The quantity of nitrogen in the soil was diminished with increasing level of arsenic. Soil samples containing approximately 165 $mg.kg^{-1}$ of arsenic also showed reduced quantities of soil bacteria as well as protozoans, and no rain worms were found in soil samples containing 150 ppm of arsenic. High levels of arsenic were detected in tissues of rabbits monitored from their birth to the age of one year, showing the same values in hairs as were detected in hair samples taken from local 10-year-old boys.

The effect of arsenic on reproductive functions of domestic animals was encountered in this area. A pig-breeding farm specialized in large-scale production of piglets was located in a village near the power plant. After the power plant went into operation, the incidence of abortions among sows started to increase, reaching the point that it was necessary to close the farm and move it away from the vicinity of the power plant. Arsenic is known to be a capillarotoxic poison. Pregnant sows received sufficient doses of arsenic in their forage to damage the placental blood capillaries to the extent that abortions started to occur. In fact, hog's placenta, compared to the human one, has a more complicated anatomic structure, which makes it more susceptible to capillarotoxic lesions. Abortion rates also increased among cattle in this area but did not reach such an epidemic proportions as in the pigs (8).

Human-health aspects: Workers engaged in stoking, boiler maintenance and boiler cleaning, by that time highly exposed, were found to show clinical manifestations of arsenic exposure such as nasal septum perforations (9).

Neurotoxicity: The first clinically recorded neurotoxic lesion liable to indemnity for an occupation-associated disease was found in the boiler operator of the power plant. This worker was found to possess a necrotic ulcus between his index finger and middle finger, on the place where he used to hold his cigarette. Due to impaired skin sensation, he failed to notice that the cigarette stub was so short as to burn and consequently necrotize the skin of the two fingers (9).

Another interesting event was the phenomenon of neurotoxicity among children of the most severely contaminated village. A detailed audiometric examination revealed hearing losses at high frequencies 4,000 Hz–8,000 Hz in both air- and bone-borne transmission (10). The exposed groups and the control groups were examined using the same apparatus, by the same person, and under identical time schedule in a noiseless audiologic chamber. The detected hearing damage was statistically proved on a group-diagnostic basis.

Immunotoxicity: In a study focused on immunological aspects of exposure, through inhaling emissions derived from burning coal with high arsenic content (11, 12), nine immunochemical parameters, immunoglobulins and proteins of acute phase were classified into normal and abnormal in the exposed workers (47 workers) and in the control group (workers of power plant burning coal of „normal" arsenic content). The prevalence of persons without and with one finding of abnormal values and the prevalence of persons with two and more abnormal values in the examined immunological parameters were determined. This approach is useful as it contributes to a more comprehensive view of the immunological status of the groups under examination.

The most interesting finding in this study was the high prevalence of abnormal values of coeruloplasmine (CPL) (43 %) in the exposed group compared to no abnormal values in the control group, which is considered to predict immunosuppressive effects of high CPL levels in the exposed group of workers.

This corresponds well with the observed increase in malignant tumours in this group compared to the mortality pattern of workers in power plants burning „normal" coal (13). The prevalence of persons with two and more abnormal parameters was 51 % in the exposed group compared to 3.7 % in the control group.

Carcinogenicity: Population-based transitional epidemiological study, which began in mid-1970s (14), covers the entire population of the Prievidza district, central Slovakia, with the primary goal of following up the incidence of all types of malignancies in this area. The study attempted to obtain a complete district register of malignant tumours within an administrative unit of the population of about 125,000. This project was feasible due to the former structure of the national health-care system, which operated in this country. Each cancer patient or any person suspected of any malignity was referred to the district oncologist who was responsible for the final diagnosis and for the therapy of the patient. Our originally intention was to perform a 10-year study. However, the data collection efforts and a comprehensive nature of the health-care system permitted to extend this study to 20 years. The study was initiated in 1976. The results of the first year were eliminated, as the system of data collection and trials of how our questionnaire was constructed and implemented was fine-tuned.

The district was split into two areas delimited by a 7-km circle around the power plant burning coal with high arsenic content. This circle was established using biological monitoring of human exposure within the particular locality. The exposure rates were established by the analyses of hair samples. The criterion of higher exposure was arsenic content exceeding, on the average, hair concentrations of $3 \mu g.g^{-1}$ of arsenic. About two-tenths of the district population under study live in a 7-km radius of the exposed region. Values up to $1 \mu g.g^{-1}$ are considered normal (15). For example, the population in Prague showed approximately $0.2 \mu g.g^{-1}$, which is less than one-tenth of the mean value, which predominated in this previously heavily emission-loaded area near Prievidza. Analysis of the database collected suggests a significant increase in the incidence of skin basalioma cancer in the most polluted part of the district compared with the data relevant for rest of the district and for the entire Slovak Republic (16). The incidence of skin basalioma is even markedly influenced by exposure to arsenic in occupational settings (14).

Considering the relatively long period of latency, so frequently described in arsenic-caused cancers, we may assume that the changed tumour mortality pattern resulted from arsenic exposures during the years characterized by much less favourable environmental conditions from the end of the 1950s to the last quarter-1970s.

The results obtained suggest that arsenic is probably a promotor rather than a true carcinogen. The non-threshold concept of arsenic carcinogenicity does not seem to be supported by the results of our database meta-analysis, which corresponds with the recently published data demonstrating its pertinence to endocrine disrupters (17).

Beryllium

It was reported that the beryllium content in the majority of coals varies from 0.1 up to 15 ppm (5). However, it has been found as much as 2,800 ppm Be in coal ash (4). It is generally considered that beryllium is confined to the organic matter of most coals, but Be may be contained in quartz and clay minerals in Indian coals. Various bonds of beryllium in coal (in organic matter, clay minerals, carbonates and pyrites) were established. This element is likely to be confined to very fine (ultra fine) mineral matter of which particles are dispersed in organic matter (18). Similar occurrence and bond of beryllium is believed to exist in coals of the Czech Republic (19) .

Mean contents of Be in coal ash in the Czech Republic attain mostly a few hundreds of ppm (Table 3). The lignites of the Josef Seam in the Sokolov Basin with about 100 ppm (maximum content 1,507 ppm) in ash were earlier burnt in a local power plant. Nevertheless, beryllium is one of the substances posing a high health risk, especially when inhaled in the form of oxide. Occupational diseases were induced even during slight and brief contact (20). Among healthy workers engaged in beryllium workshops, disturbances have been found in individual indicators of

Table 3. Beryllium content in ash of the Czech coal (ppm)

Basin	Range	Average	Number of samples
NBB	0–141	39	1,120
SoB	1–1,507	524	1,131
SMCF	–	–	–
CUSB	1–57	41	–
CWBB	7–364	50	25
ISB	7–41	24	4
KPB	–	25	1
BIG	6–8	7	2
BoG	–	20	1
BrR	–	40	5

For explanation see Table 1

natural immunity, i.e., elevated concentrations of IgG and IgA, weakening of some barrier functions and increased autoimmune reactions. The occurrence of "nonoccupational" berylliosis points to the danger to the population living near plants which pollute the atmosphere with beryllium. Different concentrations of beryllium, usually not exceeding MAC values, have been determined in the atmosphere of many large towns. The elevated levels of beryllium were related to the higher beryllium content in coal which was burned there. Some types of coal in the Czech Republic contained a higher amount of beryllium which gets into cinder and fly ash (21, 22). The use of such coal in power plants released emissions with relatively high amounts of beryllium compounds into the atmosphere (23).

Current evidence suggests that beryllium acts as a hapten with antigenic properties and is presented by antigen presenting cells to CD4+ T cells, which possess specific antigen receptors (24). With the objective of evaluating the effect of relatively low beryllium concentrations within the range of MAC values or not more than one order of magnitude, this study covered several aspects of the humoral immunity system in workers in two power plants using coal with a high beryllium content, and in the population of a town near one of the above mentioned plants, located in a zone of frequent inversion situations (23).

The level of antibodies in reactions of passive hemagglutination (RPHA) with the lung, heart, liver, spleen, thyroid gland, suprarenal tissues, and native DNA served as a characteristic of autoimmune activity. Physiological saline extracts of the organs of healthy people who had been killed in accidents were used as antigens.

Autoimmune reactions in the population, as compared with the plant workers, were substantially increased in the lungs, liver, thyroid gland, and suprarenals. The values of autoimmune and antihapten reactions did not differ in the male and female population. At a higher degree of beryllium exposure, substantially higher levels of antibodies against suprarenals were found in workers. Combustion products of coal containing beryllium obviously give rise to antigenic properties in nuclear and mitochondrial lung cell fractions. Circulating antinuclear and antimitochondrial antibodies were detected both in plant workers and in the population; in both cases the reaction in women was higher than in men (25).

The changes in humoral immunoreactivity cannot be regarded unequivocally as a sign of health damage in exposed persons (12). Beryllium also gives a positive reaction in cellular immunity. An inhibited migration of macrophages was found in exposed persons suffering from berylliosis. This demonstrates much better the intensity of the pathological process, that is, the degree of health damage of exposed persons. Nevertheless, an analysis of potential changes in cellular immunity could not be performed within this study for technical reasons. The present results, however, confirm the long-time experience that the present limits of 2 $\mu g.m^3$ of beryllium in occupational and 0.1 μg in environmental settings are not totally safe at all and are subject to discussion for quite a long time (26). As berylliosis developed in only a part of the persons either occupationally or environmentally exposed to beryllium, genetic aspects play undoubtedly an important role. Further research needs to include development of biomarkers of metal-induced immunologic diseases, detailed characterization of human exposure and – last but not least – examination of gene alleles that may confer risk, and association of exposure data with those on genetic susceptibility. It is essential that all persons with allergy be prophylactically excluded from work at risk of exposure to beryllium. The objective of the study was to study the possibility of using humoral immunity indicators for a potential assessing manifestations of exposure in people exposed to relatively low concentrations of this noxious agent in occupational and environmental settings in the context of current discussion concerning still not fully reliable MAC/TLV values for beryllium (23, 24).

Lead

Much attention was paid to lead in relation to health and the environment as it may cause some health problems in certain circumstance (27). But on the other hand there is no reason to consider coal a decive source of the occurrence of Pb in inorganic or organic matter of the coal (4). It is belived lead to be associated with galena, clausthalite (PbSe) and some Ba-minerals, in which Pb can substitute for Ba in sulfates, carbonates, phosphates and silicates. High concentration of lead in sulphides and increased content of Pb in clay minerals, particularly in illite, in addition to lead in pyrite were reported (28, 29). Besides major bond of lead in sulphides, carbonates and clay minerals, the element occurs in macromolecules of coal in connection with the character of sedimentary environment of Tertiary deposits. However, there also exist some other forms of lead in coal seams such as in crocoite ($PbCrO_4$), which was identified in Late Cretaceous bituminous Main seam in New Zealand. It seems reasonable to add pyrite and some organic compounds, probably in low-rank coal. The majority coals fall in a range of about 2–80 ppm Pb (4). The highest ever detected content of Pb in coal ash was 1,000 ppm (5).

Table 4 shows Pb concentration in ash of the Czech coals. Remarkably high mean and maximum concentrations are known from the Intra Sudetic Basin (9,550 ppm), Krkonoše Piedmont Basin (1,700 ppm) and Carboniferous relic near Brandov (1,500 ppm). High contents of Pb in these coals are mostly confined to sulphides, specifically to pyrite and galena. Different bonds of lead in lignite and bituminous coal were established (30). This author found out that Pb preferentially enters carbonates and also clay minerals and sulphides in bituminous coal of the Upper Silesian Basin. Lead in lignite of the North Bohemian Basin is confined to pyrite in particular.

Table 4. Lead content in ash of the Czech coal (ppm)

Basin	Range	Average	Number of samples
NBB	3–16	9	106
SoB	0.3–88	43	150
SMD	3–69	30	5
CUSB	28–295	151	133
CWBB	19–266	129	329
ISB	15–9,550	980	7
KPB	111–1,700	905	3
BlG	19–44	32	2
BoG	–	27	1
BrR	–	1,500	5

For explanation see Table 1

Table 5. Mercury content in ash of the Czech coal (ppm)

Basin	Range	Average	Number of samples
NBB	–	0.1	1
SoB	0.2–4.9	0.9	28
SMCF	0.01	0.01	2
CUSB	0.02–0.08	0.05	14
CWBB	0.001–0.37	0.008	10
ISB	–	–	–
KPB	–	0.01	1
BlG	0.005	0.005	2
BoG	–	0.01	1
BrR	–	–	–

For explanation see Table 1

Mercury

There is considerable concern about mercury effects on health in population and the proportion of anthropogenic Hg in the environment (31). Although, no unambiguous evidence exists so far of health problems caused by mercury released from coal burned potential risk of poisoning cannot be excluded. However, mercury still remains a problematic substance. Regardless of reduction of its emissions relatively high contents of mercury are stilll found locally in animal and human tissues (32). Mercury occurs in coal in three forms, namely in cinnabar (HgS), as metallic Hg, and in organometallic compounds. Mercury is closely associated with arsenic in coal of Northwestern Alabama (6). These elements are attributed to the occurrence of syngenetic pyrite originated during late stages of coalification. However, Hg can be confined to be to the organic matter but can be also found in sphalerite or associated with epigenetic pyrite. There exists a good correlation between Hg and pyrite. Most coal should be in the range of 0.02–1.0 ppm Hg (4). The maximum so far known content of Hg in coal ash is 50 ppm (5).

Contents of Hg in ash of the Czech coals are summarized in Table 5. The majority of coals should be in a range of 0.01–0.05 ppm Hg. The highest concentrations were detected in coal ash of the Josef seam in the Sokolov Basin ($S_p^d > 3.0$ %).

Table 6. Selenium content in ash of the Czech coals (ppm)

Basin	Range	Average	Number of samples
NBB	0.2–3.9	0.7	78
SoB	1.0–5.3	2.4	17
SMCF	2.2–3.0	2.6	3
CUSB	0.2–3.4	0.9	11
CWBB	0.3–2.7	1.2	4
ISB	–	1.9	1
KPB	–	–	–
BlG	–	–	–
BoG	–	–	–
BrR	–	–	–

For explanation see Table 1

Selenium

The influence of Se on the human health is a complex issue because traces of this element are essential for human beings as well as animals. The recommended quantity of selenium for adults is 0.5 ppm per day (33). Higher consumption of the element is obviously dangerous for living organism. Nearly 500 cases of human selenosis in southwest China were reported (34). The authors attributed this disease to selenium-rich carbonaceous shales having as much as 8,400 ppm Se of wich ash is used as an additive in fertilizing. It was discovered that Se confined to refractory clays organic matter and pyrite can easily be oxidized and transformed into water-soluble Se compounds. Such process allowed large amount of selenium to entre soil and subsequently the crops.

Average contents of Se in coal ash in the Czech Republic (Table 6) vary between 0.7 and 2.4 ppm. Slightly higher concentrations of this element were found in the Antonín Seam of the Sokolov Basin (3.9 ppm). Most Se is organically associated in Czech coal, and occurs in pyrite, galena as PbSe, at least in some coals and possibly in clays.

CONCLUSIONS

The database shows a broad spectrum of potentially harmful elements to occur in the Czech lignite, bituminous coal and anthracite. Age of sediments, places from where samples were taken, accuracy of methods, and number of analyses determined considerable differences in concentration of elements.

Most of ashes from lignite and bituminous coals from perspective deposits for future uses contain low concentration of potentially hazardous trace elements. In connection with human health, and environment protection is the presented knowledge of contents of elements (i.e., As, Be, Hg, Pb and Se) necessary at risk assessment of combustion process, stocking ashes and fly ashes, and their pertinent utilize to production of building materials.

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