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MODELLING OF MERCURY EMISSIONS FROM LARGE SOLID FUEL COMBUSTION AND BIOMONITORING IN CZ-PL BORDER REGION

MODELOWANIE I BIOMONITORING EMISJI RTĘCI Z MASOWEGO SPALANIA PALIWA STAŁEGO NA POGRANICZU POLSKO-CZESKIM

Abstract: Tightening of norms for air protection leads to a development of new and significantly more effective techniques for removing particulate matter, SO_x and NO_x from flue gas which originates from large solid fuel combustion. Recently, it has been found that combinations of these environmental technologies can also lead to the reduction of mercury emissions from coal power plants. Now the greatest attention is paid especially to the coal power plant in Opatovice nad Labem, close to Hradec Kralove. Its system for flue gas dedusting was replaced by a modern type of cloth fabric filter with the highest particle separation efficiency which belongs to the category of BAT. Using this technology, together with modernization of the desulphurisation device and increasing of nitrogen oxides removal efficiency, leads also to a reduction of mercury emissions from this power plant. The University of Hradec Kralove, the Opole University and EMPLA Hradec Kralove successfully cooperate in the field of toxic metals biomonitoring almost 20 years. In the Czech-Polish border region, comprehensive biomonitoring of mercury in bioindicators *Xerocomus badius* in 9 long-term monitored reference points is done. The values of mercury concentration measured in 2012 and 2016 were compared with values computed by a dispersion model SYMOS´97 (updated 2014). Thanks to modern methods of dedusting and desulphurisation, emissions of mercury from this large coal power plant are now smaller than before and that the downward trends continues. The results indicate that *Xerocomus badius* is a suitable bioindicator for a long-term monitoring of changes in mercury imissions in this forested border region. This finding is significant because it shows that this region is suitable for leisure, recreation, and rehabilitation.

Keywords: BAT flue gas dedusting, modelling of imissions, mercury biomonitoring

Introduction

Coal combustion is the worldwide largest source of mercury emissions to the atmosphere caused by human activity. Reserves of coal are relatively large compared to oil and natural gas deposits. Therefore, it is obvious that also in the future the coal will be used

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abundantly as a global source of energy. For this reason, it is important to pay increased attention to emissions of mercury from coal combustion [1].

The United Nations Environment Programme (UNEP) provided a review of global mercury emissions to the atmosphere and indicated that coal combustion caused around 46% of total emissions. Emissions from coal combustion could continue to rise in the following years, especially in rapidly developing areas. Considering the importance of coal combustion to global emissions, reducing and monitoring of mercury emissions, especially from coal power plants, will be an important task [2].

Mercury is the most volatile of all trace elements. During the combustion of coal at temperatures of approx. 700-800°C the thermal decomposition of mercury compounds occurs, which causes a production of gaseous elemental mercury (Hg^0) flowing out of the combustion chamber in the flue gas. Estimated amount of mercury captured in the bottom ash is less than 2% of the total amount of mercury present in coal combustion [3]. During the combustion of municipal solid waste, about 4% from the entire incoming mercury remains in the bottom ash. Mercury in emissions can be present also as an oxidized form Hg^{2+} ($HgCl_2$, Hg_2Cl_2 or HgO). Mercury (Hg^0 and Hg^{2+}) concentration in the flue gas ranges from 5 to 20 μg/Nm^3. The oxidized form of Hg^{2+} can be removed simultaneously with capturing SO_3 (during wet or dry flue gas desulfurization). In contrast, the elemental mercury Hg^0 is very difficult to remove [4].

Mitigation of mercury emission

In bigger boilers which are equipped with separators, a larger portion of mercury usually condenses on fly ash particles and is collected in the separators; a smaller portion of mercury is transported into the atmosphere where the mercury or its volatile compounds get into aerosol [5]. Mercury in the environment causes a serious health threat, it is a very powerful neurotoxin that accumulates in food chains. It penetrates into the human body particularly by inhalation of mercury vapors which persist a relatively long time in the atmosphere. Mercury occurs in the elementary form (Hg^0), oxidized form (Hg^{2+}) or is bound to very fine-grained aerosols particles [5, 6].

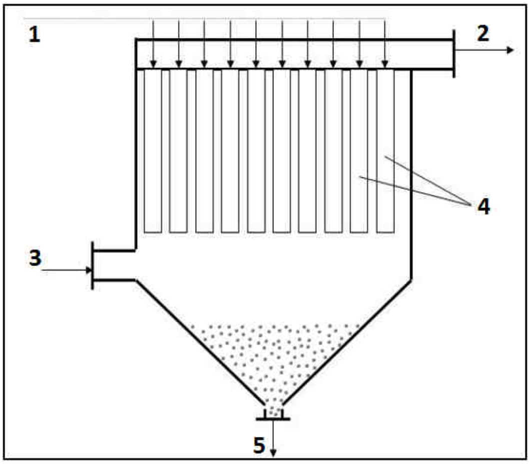

Fig. 1. A cloth filter scheme: 1 - compressed air supply for backwashing, 2 - gas outflow, 3 - gas inflow, 4 - textile filter tubes, 5 - dumping hopper for collected dust [7]

In coal power plants, the system for separation of solid pollutants which is based on mechanical and electro-static separators can be replaced by modern flue gas dedusting technology. This system called cloth fabric filter (FF) represents a modern type of separator with the highest particle separation efficiency which belongs to the category of BAT (*Best Available Techniques*). BAT means the most advanced technologies and methods of operation designed to prevent or to reduce emissions and their impact on the environment.

These hose filters are made of textil fibers combined with teflon, which is suitable for filtration of gases at high temperatures. The gas flow velocity through the filter septum is at temperatures of approx. 700-800°C, velocity in the filter area must therefore decrease considerably in comparison with the speed in the duct. That is why the filter chambers occupy a large space and are also thermally insulated. The frames and cabinets for the cloth filters as well as dumping hoppers are made of stainless steel sheets, the filter septa are stretched to baskets made of steel wires. Dedusted gas flows from the outer side of the filter fabric, while a filter cake of the collected dust remains on the inner side. The filter material has a shape of long tubes [7].

Regeneration of the filter is carried out by backwashing. A part of the filter (about ¼) is shut down and the filter area is connected with the atmosphere, so the surrounding air is supplied into the filter and it flows through the filter material in the opposite direction than the gas flows during the actual filtration. Separation efficiency of the cloth filters is 99.99%.

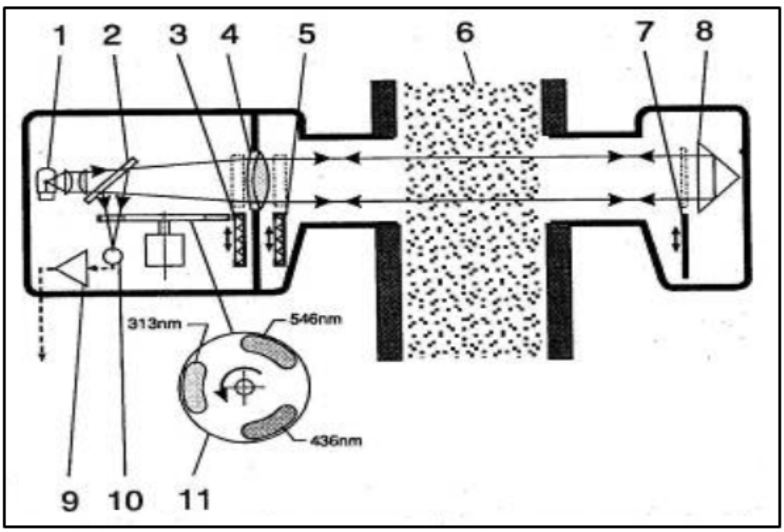

Fig. 2. DUSTHUNTER C200 - a device for operational measurements of dust particles concentration: 1 - source of radiation (mercury lamp), 2 - semi-transparent mirror, 3 - check-reflector (clean side), 4 - lens, 5 - check-reflector (smoke duct side), 6 - smoke duct, 7 - check-iris, 8 - main reflector, 9 - amplifier, 10 - radiation detector, 11 - rotational iris with interference filters [8]

For determining the content of most pollutants in the flue gas, an infrared absorption spectroscopy is the most commonly used method. It is based on the measurement and interpretation of changes in molecules which occur during their interaction with electromagnetic radiation in the wavelength range from 1 to 1000 μm (wavenumbers from

10 to 10 000 cm^{-1}). The internal energy of the molecule can be increased by absorption of electromagnetic radiation only if there is a simultaneous change of the electric dipole moment vector of this molecule. The intensities of obtained infrared absorption lines are proportional to the magnitudes of these dipole moment changes.

Absorption of infrared radiation can also be caused by increasing the translational speed of solid particles with which the radiation interacts. In this case, the absorption does not proceed in quanta, but continuously, which creates a continuous component in the spectrum.

During a measurement of dust in the flue gas, an attenuation of the light beam passing through the channel is examined. The relationship between absorption of electromagnetic radiation (*i.e.* light as well) and a concentration of dust particles is given by a known Lambert-Beer law. However, dust particles have various sizes and therefore it is necessary to perform a gravimetric measurement before each equipment startup. A new analyzer must be also calibrated prior to use [8].

The next generation of air quality monitoring of particulate matter is described in [9]. A new progressive method of using ^{210}Pb isotope as a pollutant emission indicator is described in [10].

Calculation and modelling of air pollution by SYMOS´97

To assess the contribution to imissions, it is needed to calculate the dispersion of the source smoke plume. For the calculation of air pollution, the “Methodical directions of the department of air protection of the Ministry of the Environment for the calculation of air pollution from point and mobile sources SYMOS 97” is established since 1998. The abbreviation “SYMOS” means “System for Modelling of Stationary Sources” [11].

The method SYMOS 97 is based on the Gaussian model of pollutant scattering in the atmosphere:

$$\bar{c}\,(x, y, z) = \frac{Q}{2\pi\sigma_y\sigma_z\bar{u}} \exp\left(-\frac{y^2}{2\sigma_y^2}\right)\left[\exp\left(-\frac{(z-h)^2}{2\sigma_z^2}\right) + \exp\left(-\frac{(z+h)^2}{2\sigma_z^2}\right)\right] \tag{1}$$

where: $\bar{c}$ - averaged pollutant concentration in the position [x, y, z], Q - mass flow rate of pollutants emitted by a continuous source [$mg\cdot s^{-1}$], $\bar{u}$ - averaged flow velocity along the x axis [$m\cdot s^{-1}$], σ_y, σ_z - transverse scattering parameters, y - perpendicular distance of the reference point (*i.e.* the point at which the concentration is calculated) from the plume axis.

Thus $y > 0$ if the direction vector source - reference point differs from the entered direction of flow. z - height difference between the reference point position and the effective source height, h - effective height of the source (the physical height of the source plus the thermal plume rise), x - the plume axis length. If the direction vector source - reference point is identical with the direction of flow, x is equal to the distance source - reference point.

Lower reflection modelling

The formula above corresponds to the lower estimate - a situation when a reflection of emissions from the ground surface ($z + h$) occurs.

Upper reflection modelling

It is obvious that this assumption is valid only for a flat terrain or for situations when the reference point is located on an elevated place. If the terrain raises too close to the source, the upper estimate is used - we omit the last expression in the equation, thus we obtain:

$$\bar{c}\,(x, y, z) = \frac{Q}{2\pi\sigma_y\sigma_z\bar{u}}\exp\left(-\frac{y^2}{2\sigma_y^2}\right)\exp\left(-\frac{(z-h)^2}{2\sigma_z^2}\right) \qquad (2)$$

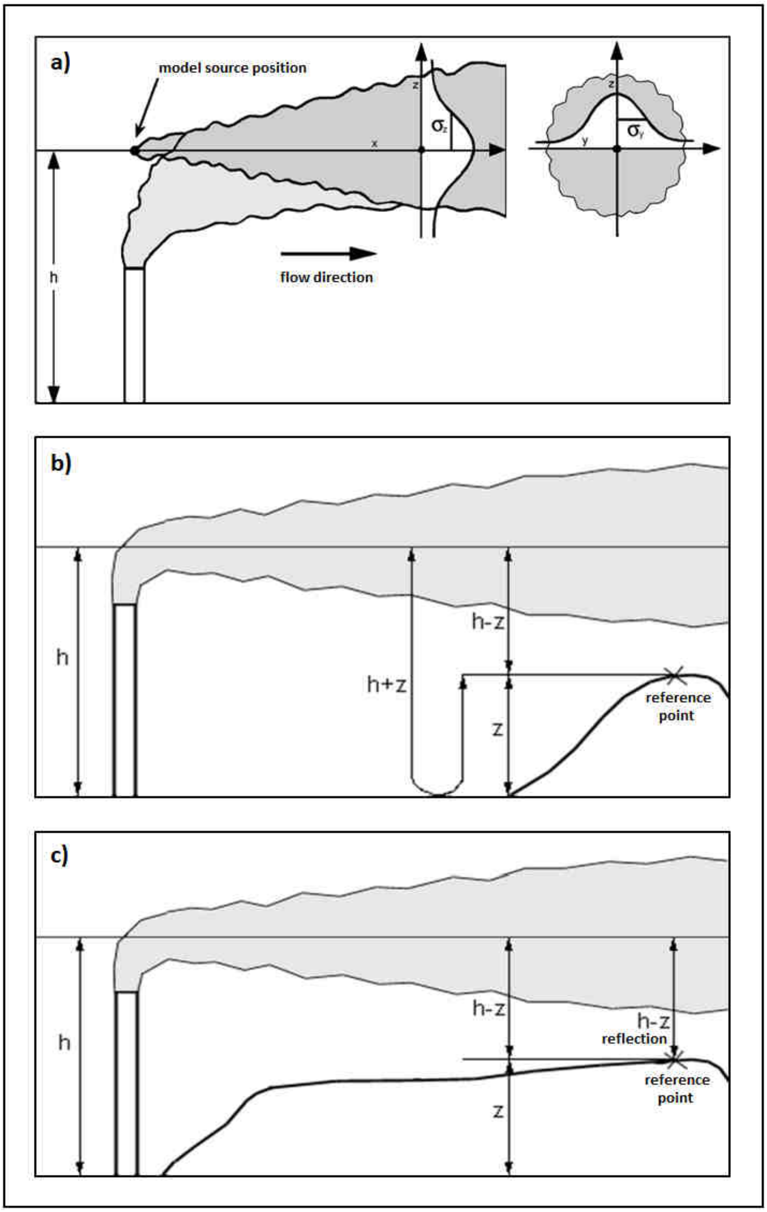

Fig. 3. Gaussian model of emissions scattering in the atmosphere (a), a scheme of the lower estimate (b) and a scheme of the upper estimate (c) [12]

According to the SYMOS 97 methodology, the dispersion model is then adapted to the form:

$$c = \frac{M_e}{2\pi\sigma_y\sigma_z u + V_s} \exp\left(\frac{-y^2}{2\sigma_y^2}\right) \exp\left(-k_u \frac{x_L}{u}\right) \cdot \quad (3)$$

$$\cdot K_h \left[(1+\vartheta)\exp\left(\frac{(z-h)^2}{2\sigma_z^2}\right) + (1-\vartheta)\exp\left(\frac{(|z|+h)^2}{2\sigma_z^2}\right)\right] \quad (4)$$

where: M_e - mass flow rate of emissions (in the previous case denoted as Q), V_s - volume flow rate of emissions, k_u - a parameter which reflects a transformation and fallout of pollutants during a transmission to the reference point, K_h - a parameter which reflects a reduction of impact of low resources in a mountainous terrain.

The expressions (1 + ϑ) and (1 – ϑ) include the upper and lower estimates to this methodology. The coefficient ϑ is determined by a ratio of the area of the terrain vertical section plane and the area of a rectangle between the source and reference point. All terrain height extremes beyond this rectangle are ignored. The value of ϑ then lies in the interval (0, 1).

In Figure 3a is a scheme of Gaussian model of emissions scattering in the atmosphere, in Figure 3b is a scheme of the lower estimate, in Figure 3c is a scheme of the upper estimate, adjusted according [12].

Biomonitoring of mercury imissions changes

Mercury imissions and its accumulation in the surface layers in the Giant Mountains is examined in publication [13].

Boletus badius is used especially for biomonitoring of ^{137}Cs radionuclide activity. This radionuclide got into the soil by a fallout after the Chernobyl accident in 1986. To assess the contamination of bioindicators by toxic metals, so called concentration ratios are used in some publications. The concentration ratio is expressed as the ratio of a particular pollutant concentration in a certain part of the mushroom to the concentration of this pollutant in the soil. Regarding soil, the most important for contamination by both the radioactive cesium and toxic metals is the organic soil layer. Therefore, only concentrations and activity measured in the organic layer are used for the calculations [14-24].

Results

In the coal power plant in Opatovice nad Labem, a system for separation of solid pollutants based on mechanical and electro-static separators had been originally used. This system was replaced by a modern type of separator with the highest particle separation efficiency which belongs to the category of BAT. To achieve the required emission limits of solid pollutants (100 mg/m^3 before application of the wet desulfurization method, 20 mg/m^3 at chimney exit) a cloth filter was installed. The filter is made of NOMEX fibers combined with teflon, which is suitable for filtration of gases with temperature of about 250°C. The gas flow velocity through the filter septum is 1-3 cm·s^{-1}, velocity in the filter area must therefore decrease considerably in comparison with the speed in the duct. That is why the filter chambers occupy a large space and are also thermally insulated. The scheme of this dedusting system is in Figure 1. After the reconstruction of the power plant in Opatovice nad Labem, a new modern continuous analyzer of dust particles in the flue gas

DUSTHUNTER C200 (manufacturer SICK) was installed there. The scheme of this analyzer is in Figure 2.

A demonstration of data measured by the continuous analyzer DUSTHUNTER C200 in the power plant in Opatovice nad Labem after the reconstruction is in Figure 4.

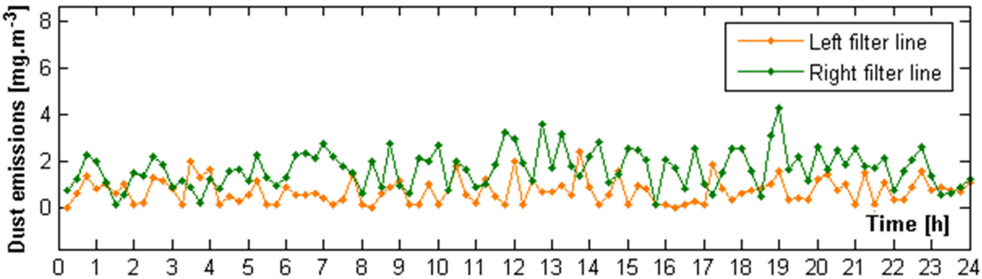

Fig. 4. A demonstration of a continuous measurement in the left and the right section of the cloth filter by the continuous analyzer DUSTHUNTER C200

In Figure 5a is a statistical evaluation of data measured by a continuous analyzer GM 30 (manufacturer SICK) before the reconstruction of the dedusting section. Figure 5b shows a statistical evaluation of data measured by a continuous analyzer DUSTHUNTER C200 after the reconstruction.

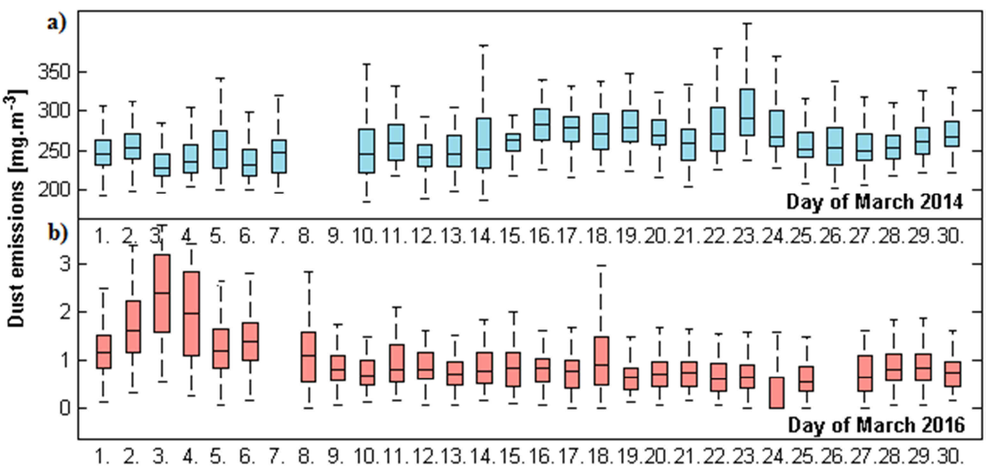

Fig. 5. Statistical evaluation of dust emissions in 2014 (a) and in 2016 (b)

For performing the calculations, the following data is entered into SYMOS 97 (updated 2014): exact location of the source, mass flow rate of emissions, height of the chimney, heat spreading rate or flue gas temperature (to calculate the flue gas uplift), operating hours per year and volume flow rate of flue gas. Data for a wind rose for Opatovice nad Labem was provided by the Czech Hydrometeorological Institute.

Table 1 shows locations of the reference points which were used for calculation and modelling of air pollution by SYMOS´97 and for biomonitoring of mercury emissions originating from coal power plants. These reference points, as well as the power plants, are shown on a map of the Czech-Polish border region in Figure 6.

Table 1

Location of reference points for modelling and biomonitoring of mercury emissions from large coal power plants in the Czech-Polish border region

Reference Point Number	X	Y	Z	GPS - North latitude	GPS - East longitude
3001	–600071	–1036653	1115	50°18'05"	16°23'52"
3002	–581919	–1066826	995	50°03'14"	16°41'14"
3003	–569116	–1057714	1224	50°48'30"	16°18'2"
3004	–569135	–1050600	1423	50°12'18"	16°50'58"
3005	–587648	–1059587	765	50°06'31"	16°37'23"
3006	–594841	–1045185	992	50°13'55"	16°29'06"
3007	–601984	–1030860	1084	50°21'01"	16°20'59"
3008	–612729	–1023881	624	50°23'59"	16°12'59"
3009	–603464	–1012629	773	50°31'03"	16°19'00"

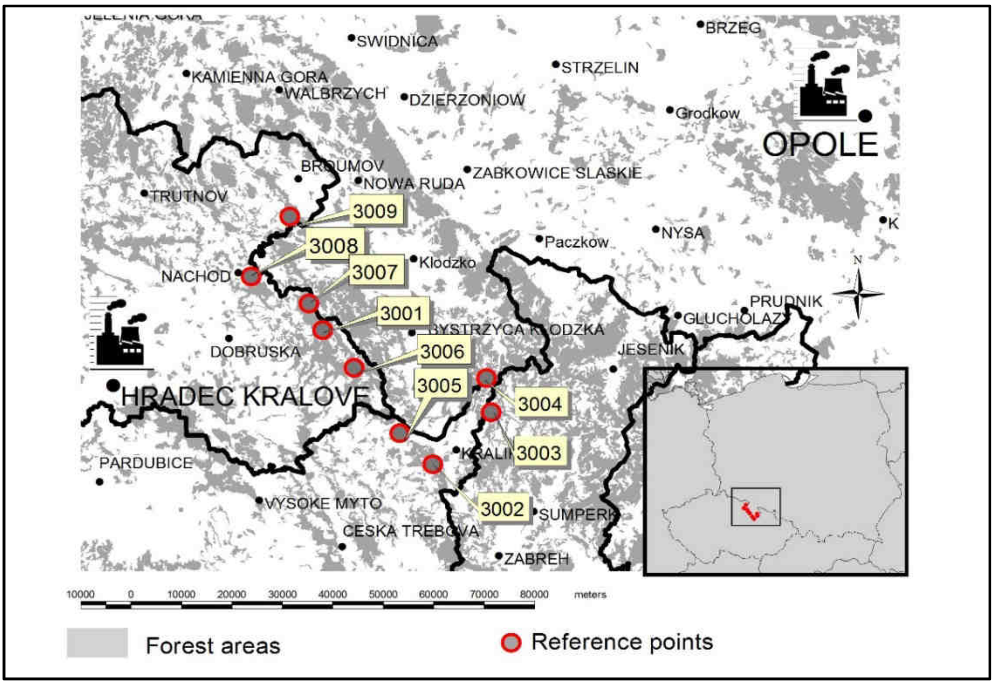

Fig. 6. A map showing the location of the coal power plants and the reference points for modelling and biomonitoring of mercury emissions in the Czech-Polish border region

Samples of *Boletus badius* were taken in a forest, at the distance of at least 100 meters from the edge of the forest and 200 meters from buildings and roads accessible by cars. The distance between each mushroom and a trunk of a tree was at least 2 meters. The sampling was done if possible in six locations close together, the total weight of samples was about 200 g. Above ground parts of the stipes and the whole caps were collected and impurities were removed from them. The caps were separated from the stipes and all the mass was cut into slices using a plastic knife and dried at the temperature of 298 K to a constant weight. Then this material was homogenised in an agate grinding mortar, sieved (sieve mesh diameter was 0.4 mm) and placed in closable plastic containers made of polyethylene (PE) or polypropylene (PP).

Furthermore, an organic soil horizon was sampled from a depth up to 10 cm. In all cases, the samples were taken from five points in a small area (the sampling points were about 50 cm from each other), the middle sampling point was at the place where the *Boletus badius* was collected. The dug soil, whose weight was about 500 g, was poured together and mixed thoroughly. Then it was sieved with a plastic sieve (mesh diameter 2 mm) and dried at the temperature of 373 K. The entire processing of the samples was done in such a way that their contamination from the environment was prevented.

Mercury was determined in the homogenised dried samples of mushrooms 0.1-0.2 g and of organic layer of soil 0.3-0.4 g using a cold-vapour AAS analyser (AMA254, Altec Prague, Czech Republic) at wavelength of 296 nm. Drying time was 300 s, decomposition time was 250 s, waiting time (necessary for quantitative trapping of the mercury in a gold amalgamator) was 45 s. A running analytical control and assurance quality (AC/AQ) was performed through the analysis of blank samples and certified reference materials Epiphytic lichen IAEA-336. Differences between experimentally determined and certified contents were up to 3%. Mean differences between duplicates were up to 5%. Blank background levels were below the detection limits.

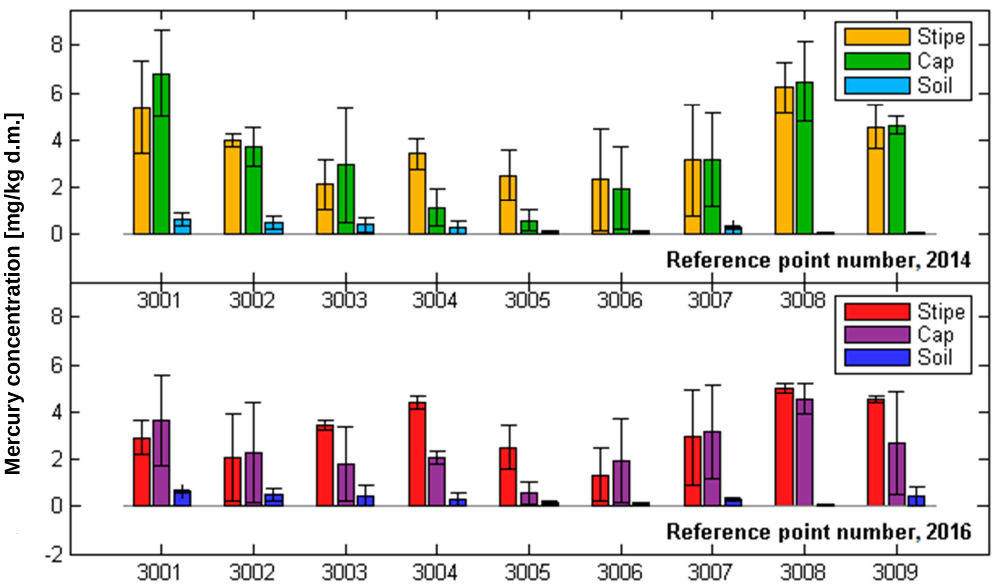

Fig. 7. The results of monitoring of mercury concentration in dry matter of *Boletus badius* in 2014 and 2016

Figure 7 shows the results of mercury concentration monitoring in dry matter of *Boletus badius* in 2014 and 2016. Table 2 shows the mean mercury concentration values in *Boletus badius* stipes and caps and in the soil sampled in the vicinity of the collected mushrooms. It also shows the results of modelling the emissions of mercury and solid pollutants PM10 and PM2.5 in the reference points. Data for modelling in Symos'97 were taken from International Power Opatovice, a.s. 2012 (internal company documents).

Table 2

The results of modelling the emissions of mercury and solid pollutants PM10 and PM2.5 and the results of mercury biomonitoring using *Boletus badius*

Modelling in reference points on CZ-PL border region				**Biomonitoring in reference points on CZ-PL border region**		
Year	**Modelling Symos'97**			**Biomonitoring *Xerocomus badius***		
	Max. values of PM and Hg emissions arithmetic average / year [$\mu g \cdot m^{-3}$]			**Hg concentrations mean values [mg/kg d.m.]**		
	PM10	**PM2.5**	**Hg**	**Stipes**	**Caps**	**Soil**
2012	0.0039	0.0023	$0.0012 \cdot 10^{-3}$	3.72±1.93	3.44±2.44	0.32±0.23
2016	0.0023	0.0017	$0.001 \cdot 10^{-3}$	3.53±1.58	3.41±1.95	0.30±0.24

Results and discussion

In the past, emissions from large coal power plants located in the Polish and Czech border region caused very heavy burden on the environment. Trees in this area were drying out and had to be replaced by new outplanting. Therefore special attention to the measurements of emissions and modelling of pollution dispersion directions was required. The University of Hradec Kralove, Opole University, EMPLA Hradec Kralove and EMITOR Opole successfully cooperate in the field of toxic metals biomonitoring almost 20 years. In the studies, samples of periodically collected lichens, mosses, birch leaves, needles of coniferous trees and surface soil layer (hummus) were used.

In our research, *Boletus badius* was used as a bioindicator particularly for biomonitoring the activity of ^{137}Cs radionuclide which had fallen into the soil in the Czech-Polish border region after the Chernobyl accident in 1986.

The aim of this part of our research was to find out if also the sporocarps of *Boletus badius* are suitable as a bioindicator for a long-term monitoring of mercury imissions changes in this region.

It was proven that *Boletus badius* is a suitable bioindicator for monitoring of mercury imissions changes in this area, thanks to the fact that these mushrooms grow every year abundantly in the vicinity of all the nine long-term monitored reference points and the distribution of mercury concentration in the soil is uniform.

The biomonitoring results were compared with values computed by a dispersion model SYMOS in the nine reference points in the Czech-Polish border region.

Evaluation of the acquired data and creating graphs was done in MATLAB, mathematical modelling of mercury emissions was performed in SYMOS’97.

Both of these indicate that, thanks to modern methods of dedusting, emissions from large coal power plants on both sides of the Czech-Polish border are now much smaller than before and that the downward trend continues. There is a gradual reduction of the concentrations of mercury emissions accumulated in *Boletus badius*. This finding is significant because it shows that this region is suitable for leisure, recreation, and rehabilitation.

Conclusions

Development of new, effective techniques for removing particulate matter, SO_x and NO_x from flue gas from large solid fuel combustion also reduces mercury emissions from coal power plants. This effect was observed also in the coal power plant in Opatovice nad Labem, where a modern type of cloth fabric filter with the highest particle separation efficiency was installed.

The University of Hradec Kralove, the University of Opole and EMPLA Hradec Kralove successfully cooperate in the field of toxic metals biomonitoring almost 20 years. Among common surveys, a long-term biomonitoring of mercury in bioindicators *Xerocomus badius* in the Czech-Polish border region was carried out. The values of mercury concentration measured in 2012 and 2016 were compared with values computed by a dispersion model SYMOS´97. Conformance of the results indicate that *Xerocomus badius* is a suitable bioindicator for a long-term monitoring of changes in mercury imissions in this forested border region. This finding is significant because this region is suitable for leisure, recreation, and rehabilitation.

Acknowledgements

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