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APPROACH FOR COMPREHENSIVE SUPERVISION OF THE WORKPLACE EXPOSURE TO LEAD AND ITS COMPOUNDS

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Abstract

This paper presents an interdisciplinary methodology for handling the exposure of workers to lead and its compounds in a systematic way by connecting different disciplines, and thus enabling the comprehensive supervision of the workplace exposure.
The exposure of workers to lead and its compounds was studied in the Republic of Slovenia. The research comprised 1079 employers being exposed to the lead concentrations between 0.01 and 0.9 mg/m^3, which exceeded the limit value by 9 times. Workers in the production of lead-acid batteries were exposed to the highest concentrations; however, the employers have already introduced the technological measures for the concentration reduction. The glazier's trade has already decreased the concentrations by even 90 % in the grinding process of crystal glass. According to the biological monitoring the highest lead values amounting up to 226 μg/L were present in the blood of workers who recycle lead-acid batteries, followed by the production of lead-acid batteries and glass production with 165 μg/L. The average values have not been exceeded; however, some individuals in these branches (above all smokers) have the exceeded values above 400 μg/L. Moreover, the research showed that workers are exposed to lead compounds which are authorized. The proposed methodology provides algorithmic flow charts of the implementation procedures, and can serve as a guide for analysing the exposure to other dangerous substances.

Key words: lead, methodological approach, risk assessment, safety, workplaces

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1. Introduction

In the economy, there are practically no branches where workers would not be exposed to any dangerous substances. According to International labour organization (ILO) data (ILO, 2014) more than 313 million accidents at work happened all over the world in 2010, considering only accidents with the worker's absence over 4 days. Due to accidents at work and work-related diseases more than 2.3 million people die every year, whereas, over 350,000 people die due to accidents at work (15 %) and almost 2 million due to work-related diseases. Almost 900,000 people die due to handling with dangerous chemical substances which represents 38 % of all fatal accidents (evaluation for 2011). This represents the loss of 4 % of global Gross Domestic Product (GDP) (ILO, 2013). The costs of accidents at work and work-related diseases in the European Union are estimated between 2.6 % and 3.8 % of GDP (ILO, 2009).

In 2014 there were 217 million workers in the EU (EC, 2014). In the same year 3.2 million accidents at work with the worker's absence over 4 days were registered, whereas 3739 workers died due to accidents at work (Eurostat, 2014). According to ILO estimations 167,000 workers die due to their work every year in the EU-27. 159,500 fatal victims die due to work-related diseases, whereas 74,000 die due to the exposure to dangerous substances (EU-OSHA, 2009). According to estimations 2.7 million of new cancer cases were diagnosed in the EU countries in 2012 (OECD, 2014). In 2012, 79,700 workers died of

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professional cancer in the EU (Jongeneel et al., 2016). As reported by European Agency for Safety and Health at Work 15 % of workers have to handle dangerous substances at work, the next 19% inhale smoke, vapor, dust in their workplace (Parent-Thirion et al., 2007). With regard to the seriousness of this issue the European Commission included dangerous chemical substances into the EU Strategic Framework on Safety and Health at Work for the period between 2014 and 2020 (EC, 2014) where further special attention should be paid to professional types of cancer, asbestos-related diseases, lung and skin diseases, asthma and other chronical diseases. The conditions of safe working standards in the workplace are defined by the legislator, therefore, the best way of implementing the regulation in the work environment should be established to avoid differences between the regulated and actual circumstances in the workplace (Caraballo-Arias, 2015), and the efficient internal and external supervision can surely make a contribution (Arntz-Gray, 2016; Darie et al., 2019; Swuste and Reniers, 2016).

The employer can reduce the exposure to dangerous chemical substances by replacing dangerous substances with less dangerous (Greco et al., 2016), by introducing technological solutions for emission reduction in the work environment (Gurit Group, 2017), with an appropriate work organisation, whereas, accordance does not only depend on knowledge, understanding and skills of individuals (Gray and Silbey, 2011) but also on the motivation of individuals (Tyler, 2011) and by using suitable personal protection equipment (Gherardi et al., 2011). Furthermore, exposure can be reduced by introducing suitable procedures of control and approval of dangerous and very dangerous substances (Brückner et al., 2016; Cozma et al., 2018; Darabont et al., 2018), by considering the interaction between noise and ototoxic chemicals (Estill et al., 2017), appropriate training of workers (Breslin and Smith, 2006; Evanoff et al., 2016), appropriate measurements of dangerous substances concentrations (Vaquero et al., 2016), and a suitable biological monitoring (Ayélo et al., 2016).

The problems of exposure are often tackled partially, although it is a complex field requiring interdisciplinary approach including the fields of safety and health at work, general chemistry, analytical chemistry, chemical process techniques, medicine and law. A general systematic methodology is needed that would guide the practitioners towards the effective supervision of the workplace exposure. In our previous work (Korat et al., 2014), health and safety aspects of exposure to styrene were investigated. The present article represents an upgrade of our past work with the development of a methodological approach for the comprehensive assessment of exposure to lead and its compounds being illustrated with an example of exposure of workers in the Republic of Slovenia. This methodology represents a novelty in the sense of integration between different fields, procedures and measures within a systematic approach providing the algorithmic flow charts of the implementation procedures. The methodology can be used as guidance for the supervision of the workplace exposure to different dangerous substances.

2. Development of the methodological approach for the exposure supervision to lead and its compounds

The main problem for the efficient management and supervision of exposure to lead and its compounds is the lack of knowledge about this demanding problem, the extensive legislation and the complexity of the field which has not been dealt with in a comprehensive way. Risk at work has been evaluated by using different methodologies recommended in the literature (AUVA, 2006, 2009; EC Practical guidelines, 2006 - harmonization with the requirements of the EC Regulation 1272 (2008) of the European Parliament and of the Council of 16 December 2008 on classification, labelling and packaging of substances and mixtures (CLP Regulation) (ILO, 2001; ISO 31000, 2009; Kinney and Wiruth, 1976; OHSAS 18001, 2007; OHSAS 18002, 2008). A methodological approach which connects or in some cases upgrades the above mentioned methodologies is necessary. Therefore, a systematic approach for the comprehensive supervision of the exposure to lead and its compounds in workplace has been elaborated as shown on Fig. 1, which consists of five major steps as described below.

2.1. Collection of data on lead and its compounds to which workers are exposed

Experts for safety and health at work collect data from employers' technical documents, safety data sheets with or without enclosure of exposure scenario, safe work manuals, packaging labels and different data bases. Beside educational institutions various institutes and associations as well as Labor Inspectorate of the Republic of Slovenia and Chemicals Office support the branch of safety and health at work, which do not act exclusively as supervisory authorities but also often play an advising role. The following international and European authorities and organisations cover this field and support recognizing lead and its compounds at work: ILO (International Labour Organization, http://www.ilo.org/global/lang--en/index.htm), IALI (International Association of Labour Inspectors, http://www.iali-aiit.org/), WHO (World Health Organization, http://www.who.int/en/), ACGIH (American Conference of Governmental Industrial Hygienists, http://www.acgih.org/), IARC (International Agency for Research on Cancer, http://www.iarc.fr/), NIOSH (National Institute for Occupational Safety and Health, http://www.cdc.gov/niosh/), DHHS (U.S. Department of Health & Human Services, U.S. National Toxicology Program (NTP), http://ntp.niehs.nih.gov), OSHA (European Agency for Safety and Health at Work, https://osha.europa.eu/en/), ECHA (European Chemicals Agency, http://echa.europa.eu/).

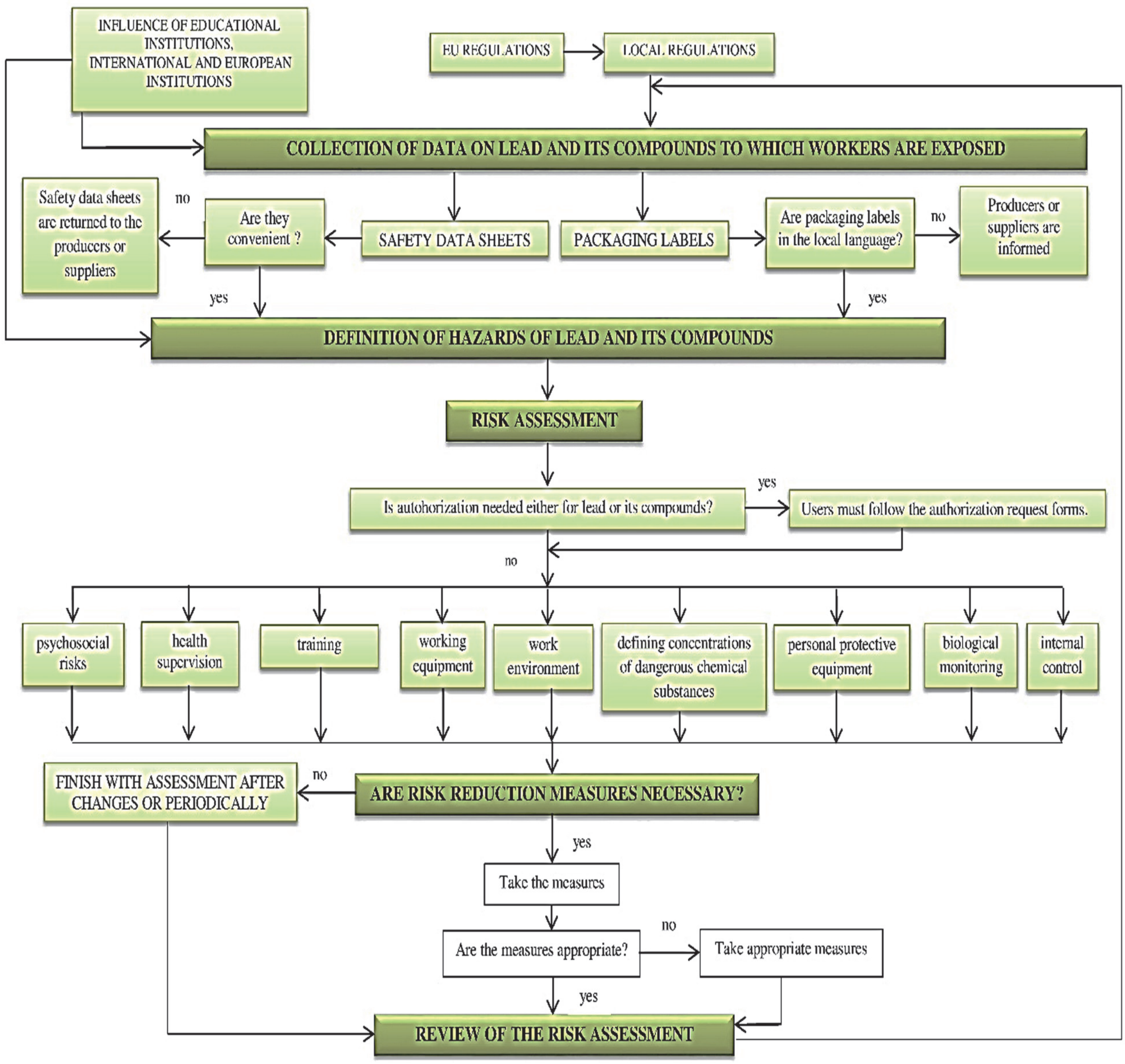

Fig. 1. Methodological approach for the exposure supervision

2.2. Definition of hazards of lead and its compounds

Lead affects the nervous system and the cardiovascular system, and above all it causes higher blood pressure (Vaziri and Sica, 2004) and kidney disorders. It has been known for a long time that lead changes the hematologic system with the inhibition of activities of more enzymes which are included into the heme biosynthesis. Delta-aminolevulinic acid dehydratase (ALAD) is especially sensitive to lead toxicity. Blocking the ALAD appears in a wide spectrum of lead in blood and it starts at the concentration level under 10 µg/dL (Ahamed et al., 2005; Jin et al., 2006).

Anaemia caused by lead has resulted from hindering the heme synthesis and shortening the life period of Erythrocytes, moreover, lead can cause unsuitable synthesis of erythropoietin hormone leading to insufficient maturing of red blood cells and finally to anaemia. The changes in the circulation of the thyroid hormones level, namely thyroxine (T4) and thyrotropin (TSH) - the thyroid-stimulating hormone, occurred in general with workers who had the middle value of lead in blood PbB ≥ 40-60 µg/dL (Singh et al., 2000). However, the changed serum levels of reproductive hormones, above all Follicle stimulating hormone (FSH), Luteinizing hormone (LH) and Testosterone, were perceived with workers with the concentration level of lead in blood PbB ≥ 30-40 µg/dL (Ng et al., 1991). The changed immune parameters in some T cellc subgroups (Sata et al., 1998) and the changed polymorphonuclear leukocyte (PMN) functions (Valentino et al., 1991) were discovered with workers with blood lead concentration between 30 and 70 µg/dL. The exposure to high lead concentrations ranging between 100 and 120 µg/dL for adults or 70 and 100 µg/dL for children causes encephalopathy. Symptoms develop after a longer exposure and include apathy, irritability, poor concentration, stomach ache, constipation, vomiting, cramps, coma and death (Kumar et al., 1987). Bad state of health, forgetfulness, irritability, apathy, headache, tiredness, impotence, decreased libido, dizziness, frailty, vertigo and paresthesia are symptoms of workers with the blood lead level between 40 and 80 µg/dL (Rosenman et al., 2003).

According to researches there is a connection between the lead exposure and hearing disorders (inner ear, cochlea) (Chuang et al., 2007; Counter and Buchanan, 2002; Hwang et al., 2009; Park et al., 2010). The connection between low levels of lead in blood (approx. 7 µg/dL or more) and occupational noise-induced hearing loss was ascertained (Hwang et al., 2009).

2.3. Risk assessment

Annex VI to the CLP Regulation classifies substances according to hazard classes and categories. It has been determined that lead compounds have been classified in category 1 as toxic to reproduction, some of them in category 2 as carcinogenic. Lead compounds meet the criteria to be included in the REACH Annex XIV. Substances which would like to be placed on the market or put into service and are included in Annex XIV of REACH Regulation need to be authorized irrespective of the quantity. 173 substances are included in the authorization list, 31 of them represent different lead compounds (http://echa.europa.eu/sL/candidate-list-table). The seriousness of consequences of occupational diseases are beyond the control, however, the probability of developing occupational diseases can be decreased in a way that important influence factors are handled accurately, e.g. psychosocial risks, an appropriate health supervision, training, working equipment, a suitable working environment, measurements of chemical hazard, appropriate personal protective equipment, biological monitoring, and appropriate internal control.

Labour Inspectorate of the Republic of Slovenia with its repressive as well as advising role plays an important role in a risk reduction and issues injunction decisions on the basis of inspection supervisions at the employers' including deadlines for employers to abolish irregularities.

2.4. Risk reduction measures

Step four comprises a definition of risk reduction measures, including technical, organizational and individual safety measures (Directive 89/391/EEC European Council; EC Directive 24, (1998)). A technical measure is a safety measure with the highest advantage. The implementation of organizational safety measures usually does not require huge capital investments; however, it can contribute essentially to the risk reduction. In case risk of workers due to exposure to dangerous chemical substances cannot be reduced by using technical and organizational measures, the third type of measures – individual safety measures – should be applied.

2.5. Risk review

According to Law on Safety and Health at Work the safety statements have to be completed with every new danger and change of risk level. This means the whole content of a safety statement including the risk assessment has to be corrected and completed in each case and not only in specific time periods and it always has to be adjusted to the current situation.

3. Materials and experimental methods

The approach presented in Section 2 was applied to assess the exposure of workers in Slovenia to lead and its compounds, and to establish a monitoring and control system for managing the exposure.

3.1. Collection of data on employers and employees

Basic information was gained in the national central register and data bases of Chemicals Office of the Republic of Slovenia which maintains the national register on chemicals in the production or import in the Republic of Slovenia. Based on data on possible exposure to lead 29 employers with 6,788 employees were included into the supervision where it was determined that 1,078 employees, among them 901 male (83.6 % of the exposed) and 177 female (14.6 %), aged from 19 to 64 years, with the average age of 43 years (standard deviation SD 10.7), were exposed to lead and lead compounds.

3.2. Measurement procedures of lead and lead compounds airborne concentrations

An appropriate measurement plan according European Standard EN 689:1995 (CEN, 1995) had been prepared by experts from different fields, such as safety engineers, technologists, production managers, performers of measurements of chemical hazards at workplace, trade union representatives, workers' representatives, etc.

The following was considered while preparing the sampling plan:

- work technology and technological procedures,
- work in one or more shifts,
- type of work, the quantity of substances that workers are exposed to,
- number of exposed workers.

Moreover, the measuring time was defined according to exposure time (short-term exposure, 8-hour exposure, etc.), intervals between individual measurements, sampling types (stationary, personal). The possibility of homogeneous group forming was studied and in case homogeneous groups have already existed the sampling of at least 10 % of exposed workers was performed.

3.2.1. Method

The NIOSH method No. 7105 (NIOSH, 1994) for measuring elemental lead and lead compounds except alkyl lead in dust samples was used for analysing samples. This method can be used for the working range from 0.002 mg/m^3 up to 1 mg/m^3 in the case of 200 L of sampling air passing the filter. The method is applicable to elemental lead, including Pb fume, and all other aerosols containing lead. High concentrations of calcium, sulphate, carbonate, phosphate, iodide, fluoride, or acetate can be offset by an additional sample treatment step.

3.2.2. Equipment

Sampling of lead and its compounds in the workplaces air was performed by using hand pump (Model 224-PCXR8; SKC Inc, Eighty Four, PA, ZDA) with adjustable flow from 1 to 4 L/min. The dust was collected in cellulose ester membrane filter (pore size 0.8 μm, diameter 37 mm). Sample was analysed with the Varian SpectrAA-800 - electrothermal atomic absorption spectrometer (Varian Australia Pty Ltd Mulgrave, Victoria, Australia) with an atomization unit with a graphite cuvette with the platform of Agilent Technologies (GTA 120 - graphite tube atomizer) and with a magnet unit enabling the Zeeman correction of background as well as with PSD 120 programmable sample dispenser.

3.2.3. Sampling

The flow rate of the sampling pump can be adjustable from 1 to 4 L/min. Before sampling the sampling pumps were calibrated with the attached sampling filters. The sampling flow rate was set to 2 L/min. The worker had the pump hooked on his belt during the whole measuring period. The maximum sampling period was 8 hours. During the sampling time the total air volume passing through the filter was 960 L. The sampling time was chosen in such a way that the average of a working procedure according to the sampling plan, was included. Regarding the time results were adjusted for 8 hours. Measurements were carried out during the week during the realistic exposure of workers. The total dust of sample loading on filter did not exceed 2 mg. In the sample preparation we used HNO_3 and solution of HNO_3 with H_2O_2 (NIOSH, 1994).

3.2.4. Accuracy

As mistakes are not defined in the framework of the NIOSH method No. 15 they need to be defined individually by performers and included in the report when presenting results.

3.2.5. Calibration and quality control

Six working standards covering the range from 0.002 to 0.1 μg/mL Pb (0.02 to 1.0 μg Pb per sample) were prepared for calibration. In the literature (BDR, 1990) the LOD (limit of detection) was determined at 0.02 μg per sample, with a LOQ (limit of quantitation) of 0.05 μg per sample (Taylor et al., 1977). The analytical procedure was evaluated by DataChem Laboratories in 1990 (BDR, 1990).

3.3. Biological monitoring procedure and blood analyses

The inspection was aimed at the performance of biological monitoring at different workplaces in different industrial sectors. The performance plan of biological monitoring, including blood collection, analysis and evaluation of results, was written before the inspection supervision, and it was carried out according to the organizational chart on Fig. 2. The biological monitoring was performed with 735 workers exposed to lead and lead compounds (among them 117 female and 618 male, aged from 19 to 64 years, with the average age of 42 years) (standard deviation SD 11.2). Blood samples were taken from Tuesday to Thursday in the employer's premises, in medical centers which were the closest to workers or at occupational medicine practitioners'. Blood was taken into test tubes for trace elements with additive of Heparin-Natrium and transported to the lab in Ljubljana University Medical Centre.

Workers were asked to fill in a questionnaire regarding their workplace, their whole working life, the working life at this workplace exposed to lead, smoker/non-smoker, medical problems and number of children or children wish, by occupational medicine practitioners before taking blood. Moreover, workers were asked about their most common troubles which appear when they have been exposed to lead for a long time, e.g. headache, dizziness, visual disturbances, kidney disorders, increased blood pressure, pins and needles in their arms and legs, etc.

Laboratories for analyzing lead in blood, delta-aminolevulinic acid dehydratase and Protoporphyrin in Erythrocytes have been specified.

However, lead has been analyzed by using electro thermal atomic absorption spectrometry (ETAAS-Zeeman) on the SpectrAA-800 Varian machine in the room with filtered air. Pb Trisol Standard Merck has been used for calibration and Seronorm Whole Blood (SERONOM, L1-21μg/L and L2-396μg/L) for the control. The delta-aminolevulinic acid dehydratase was measured by UV-1601 Shimadzu spectrophotometer by using the colorimetric method (perchloric acid, triton x-100, phosphate buffer, δ-ALA substrate, Ehrlich's reagent, trichloroacetic acid). The Protoporphyrin analysis in Erythrocytes has been carried out by using hematofluorometer.

4. Results and discussions

The supervision of the exposure to lead and its compounds was carried out according to the provisions of Health and Safety at Work Act (GRS, 2011) and other implementing regulations on health and safety at work.

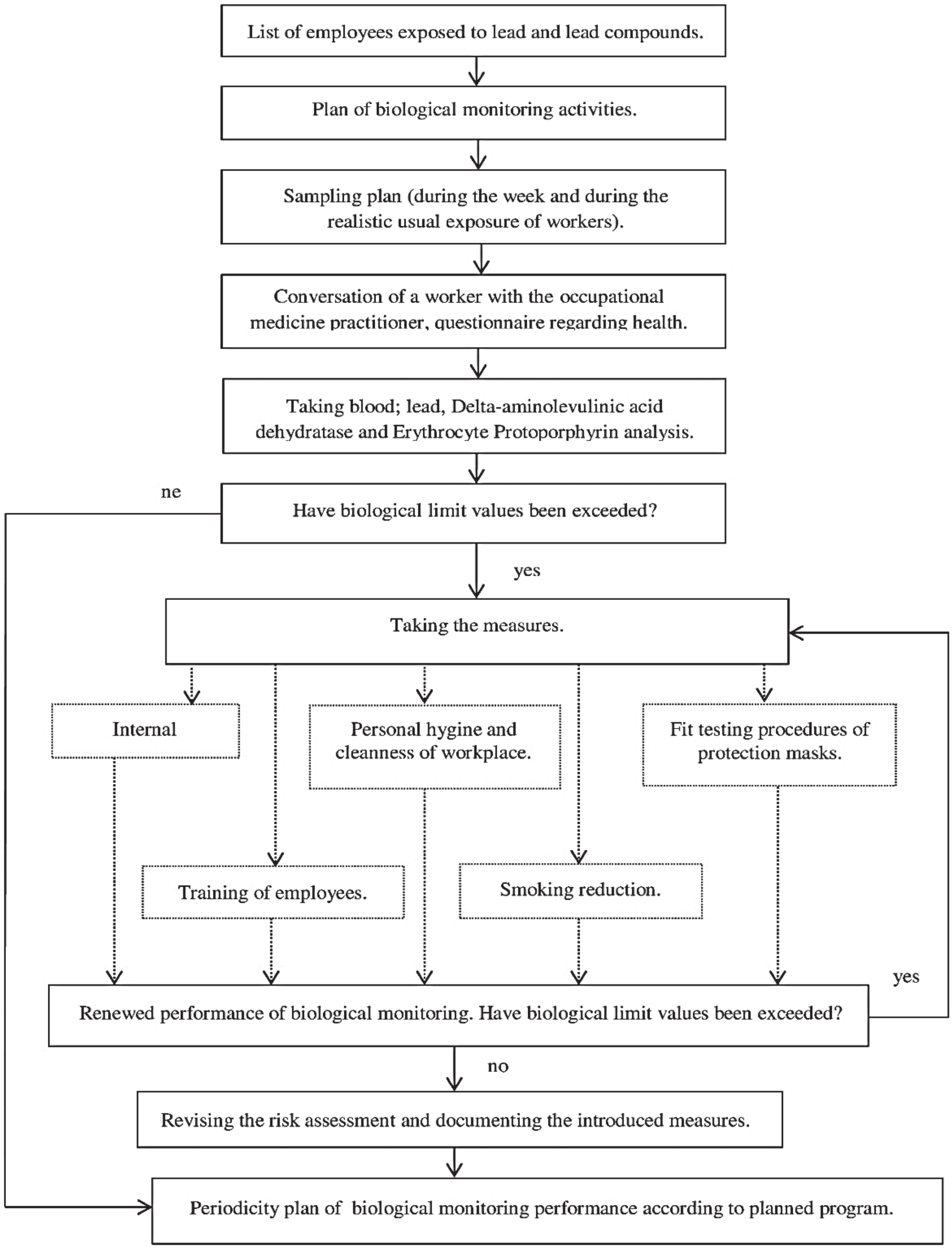

Fig. 2. Flowchart for carrying out a biological monitoring

4.1. Identifying workplaces where workers are exposed to lead and lead compounds

During the supervision it was ascertained that workers were exposed to lead and lead compounds in the following processes: lead-acid batteries production; ore melting, refining, compound production and casting; glass production (including cutting and etching); lead-acid battery recycling; work with metallic lead and lead contained in mixtures; pigment and paint production; other industries (including pipes, lights and cables); waste disposal industry; pottery and glazing; inorganic and organic compounds production; badges, jewellery decoration and glassware decoration.

With regard to the type of lead compound it was established that:

- In all above mentioned activities workers were exposed to lead or lead (II) oxide, except in the pigment and paint production and inorganic and organic mixtures production.

- Workers in the pigment and paint production were exposed to lead chromate, lead chromate molybdate sulfate red (C.I.Pigment Red 104), lead sulfochromate yellow (C.I.Pigment Yellow 34) and fluid lead octoate.
- Workers in the inorganic and organic mixtures production were exposed to lead nitrate, lead carbonate and lead hydrogen carbonate when producing some fluid galvanic preparations.
- Workers handling with metallic lead and lead contained in compounds when soldering with soldering wires were exposed to the S-Sn60Pb38Cu2 compound, named fluitin and the so called tin wire consisting of Si-Sn6Pb36Ag2 and Pb_3O_4 (red lead, orange lead).
- The following workers were exposed to red lead: in the production of lead-acid batteries, when gluing elastomers, those included in other industrial sectors as well as in the production of rubber products, being classified as a business activity with metallic lead and lead contained in compounds.
- Workers in the PVC pipe production were exposed to tetra lead trioxide sulphate which is the PVC stabiliser.

4.2. Risk assessment

4.2.1. Hazards of exposure to lead and lead compounds

Slovenian workers were exposed to the substances on the authorization list as presented in Table 1. Besides, they were exposed to lead (CAS No. 7439-92-1), lead carbonate (CAS No. 598-63-0), lead hydrogen carbonate (CAS No. 13468-91-2) and lead octoate (CAS No. 7139-86-0).

Lead carbonate and lead hydrogen carbonate used in the production of some fluid galvanic preparations are not allowed for the use as substances and compounds for preparations of colours, except restoration of works of art and historical buildings and their interior (REACH Regulation). The substances stated above as well as lead and lead octoate have not been included in the authorisation list yet, however, special attention should be paid to handling with these substances.

As shown in Table 1 the Slovenian workers are exposed to lead substances of category 1A, which are known as toxic to the reproduction of humans. Some lead substances, such as lead chromate, lead chromate molybdate sulfate red and lead sulfochromate yellow, are not only toxic to reproduction but also carcinogenic (category 1B). These substances have already been authorized (https://echa.europa.eu/addressing-chemicals-of-concern/authorisation/recommendation-for-inclusion-in-the-authorisation-list/authorisation-list). The decision on authorisation has been made public for lead chromate molybdate sulfate red and lead sulfochromate yellow (https://echa.europa.eu/addressing-chemicals-of concern/authorisation/applications-for-authorisation-previous-consultations).

It follows from the decision that the allowed usage depends on the usage type from 21 May 2019 to 21 May 2022. EC Directive 37, (2004) on the protection of workers from the risks related to exposure to carcinogens or mutagens at work has not been adjusted to new findings on lead compounds, as these substances are not included. They are included in EC Directive 24, (1998) which, however, does not include carcinogens. Taking into account the permanent findings regarding characteristics of substances or mixtures it would make sense to introduce a unified Directive on the protection of workers from the risks related to exposure to dangerous substances including carcinogens and mutagens as well as substances toxic to reproduction.

4.2.2. Measurements of lead and lead compounds concentrations in the air

During the supervision, limit values of lead and some lead compounds in different countries or regulations were checked (Table 2) indicating substantial differences among countries. Supervision results are presented in Table 3 including measured airborne concentrations in the last column.

Table 1. The substances in the authorization list to which Slovenian workers are exposed

CAS No.	*Lead or lead compound*	*The substance was included in Annex XIV of REACH Regulation due to this classification*
1317-36-8	lead monoxide	category 1A
1314-41-6	lead tetroxide	category 1A
7758-97-6	lead chromate	category 1B category 1A
12656-85-8	lead chromate molybdate sulfate red (C.I.Pigment Red 104)	category 1B category 1A
1344-37-2	lead sulfochromate yellow (C.I.Pigment Yellow 34)	category 1B category 1A
301-04-2	lead acetate	category 1A
10099-74-8	lead nitrate	category 1A
12202-17-4	tetralead trioxide sulphate	category 1A

Category 1A – toxic to reproduction, category 1B – carcinogenic

Table 2. Limit values (LV) for lead or leads compounds

Substance	***Lead or inorganic compounds***		***Tetraethyl lead***		***Tetramethyl lead***		***Reference***
CAS No.	**7439-92-1**		**78-00-2**		**75-74-1**		
Remarks	As Pb		As Pb		As Pb		
	LV – 8 hours (mg/m^3)	LV – short term (mg/m^3)	LV – 8 hours (mg/m^3)	LV– short term (mg/m^3)	LV – 8 hours (mg/m^3)	LV – short term (mg/m^3)	
EC Directive 24, (1998)	0.15						EC Directive 24, (1998)
SCOEL	0.10						SCOEL, 2002
Slovenia	0.10[1]	0.40[1]	0.05	0.2	0.05	0.2	MDDSZ, 2015
Austria	0.10[1]	0.40[1]	0.05	0.2	0.05	0.2	IFA, 2017
Belgium	0.15		0.1		0.15		IFA, 2017
Denmark	0.05[1]	0.10[1]	0.05	0.1	0.05	0.1	IFA, 2017
Finland	0.10		0.075	0.23	0.075	0.23	IFA, 2017
France	0.10[1]		0.1		0.1		IFA, 2017
Germany	0.15[1,2], 0.10[3]		0.05	0.1	0.05	0.1	IFA, 2017
Hungary	0.15[1], 0.05[4]	0.60[1], 0.20[4]	0.05	0.2	0.05	0.2	IFA, 2017
Ireland	0.15		0.1		0.15		IFA, 2017
Italy	0.15[1]						IFA, 2017
Latvia	0.005	0.01	0.005				IFA, 2017
Poland	0.05		0.05				IFA, 2017
Spain	0.15[1]		0.1		0.15		IFA, 2017
United Kingdom	0.15		0.1		0.1		IFA, 2017

LV = limit value, [1] inhalable aerosol, [2] binding occupational exposure, [3] reference value, [4] respirable aerosol

The findings by the inspection supervision indicated that workers in the production of lead-acid batteries were exposed to the highest concentrations of lead in inhalable dust. The highest concentrations were measured in the lead plate cleaning process where the lead concentration amounted up to 0.9 mg/m^3 despite the local extraction. The increased concentrations to the amount of 0.5 mg/m^3 were measured in the mechanical pilling up process of lead plates. In addition, limit values were exceeded in the plate forming process - welding with the concentration of 0.2 mg/m^3, and in the wet filling process of small nets with 0.17 mg/m^3.

The glass production concentrations were higher in the dry glassware cleaning process with 0.6 mg/m^3. In the process of preparation compounds and potsherds the concentrations amounted up to 0.07 mg/m^3.

Lead concentrations have been also exceeded in the lead-acid battery recycling at drum furnaces (0.43 mg/m^3) and in the refining (0.25 mg/m^3 and 0.33 mg/m^3). Workers in other technological processes were not exposed to exceeded limit values of lead or lead compounds.

4.2.3. Biological monitoring

Biological monitoring was aimed at defining the dimensions of workers' exposure to dangerous chemical substances and their potential health impairment. Table 4 shows biological limit values (BLV) for lead in some countries.

Supervisions revealed that the highest lead values amounting up to 226 µg/L in average were present in the blood of workers who recycled lead-acid batteries, followed by the production of lead-acid batteries and glass production with the average value of 165 µg/L as well as ore melting, refining, compound production and casting with 120 µg/L of blood (Table 5). Biological monitoring was carried out by these employers every year. Some workers in the production of lead-acid batteries had exceeded biological limit values; therefore, they have been relocated temporarily to another workplace. These employers set goals to achieve values of Pb under 300 µg/L of blood for male workers and under 200 µg /L for female workers.

The supervision revealed that biological monitoring was carried out in an ununified way. In the lead-acid battery production, ore melting, refining, compound production and casting as well as lead-acid battery recycling only lead in blood was checked but no Delta-aminolevulinic acid dehydratase and no Erythrocyte Protoporphyrin in a liter of erythrocytes. When comparing Delta-aminolevulinic acid dehydratase in a liter of erythrocytes during different types of technological processes we came to the conclusion that results showed its decreased activity when values of lead in blood were higher, e.g. in case of glass production we can notice the value of Delta-aminolevulinic acid dehydratase in a liter of erythrocytes amounting in average 29.6 U/LE, on the other hand in the pigment and paint production it amounted to 50.5 U/LE. Erythrocyte Protoporphyrin in a liter of erythrocytes was just the opposite. The last phase of heme-synthesizing activity is the introduction of Fe^{2+} into Protoporphyrin XI (PP). This step is disturbing for lead. Hindering the Fe^{3+} reduction when being exposed to lead causes the decrease of Fe^{2+} transfer into mitochondria (Taketani et al., 1985) resulting into PP accumulation in erythrocytes.

Table 3. Supervision results of employers in 12 groups of branches

Type of technological processes	***Total number of employees***	***Total number of workers exposed to lead and lead compounds***	***Male workers exposed to lead and lead compounds***		***Female workers exposed to lead and lead compounds***		***% of employers who treated exposure as a special risk***	***% of employers who taught their employees about dangerous characteristics of lead and lead compounds***	***% of personal protective equipment usage***	***8-h lead concentrations (mg/m³)***
			N	%	*N*	%				*Average (range)*
Production of lead-acid batteries	498	418	403	96.4	15	3.6	100	100	100	0.09 (0.001-0.9)
Ore melting, refining, compound production and casting	365	217	205	94.5	12	5.5	25	25	50	0.02 (0.01-0.06)
Glass production	754	151	77	51.0	74	49	100	100	100	0.06 (0.01-0.6)
Lead-acid battery recycling	85	82	81	98.8	1	1.2	100	100	100	0.09 (0.01-0.43)
Work with metallic lead and its compounds	1664	67	12	17.9	55	82.1	33.3	20	75	0.02 (0.01-0.04)
Pigment and paint production	892	47	45	95.7	2	4.3	40.0	20	67	0.02 (0.01-0.03)
Other industries (including pipes, lights and cables)	1775	30	29	96.7	1	3.3	33.3	0	33.3	0.01 (0.005-0.015)
Waste disposal Industry	98	26	26	100	0	0	0	0	not defined	-
Pottery and glazing	17	14	8	57.1	6	42.9	0	0	not defined	0.001
Inorganic and organic compounds production	45	12	7	58.3	5	41.7	0	100	100	0.001
Other processes	23	10	4	40.0	6	60.0	0	0	not defined	0.001
Badges, jewellery decoration and glassware decoration	572	5	5	100	0	0	100	0	0	0.005
Total	**6788**	**1079**	**901**	**83.6**	**177**	**16.4**				

Table 4. Biological limit values for lead in different regulations or countries

Country or regulation	***Pb in blood (µg/L)***					***ALAD (U/lE)***	***EP (mg/lE)***	***Reference***
	Universal BLV	***BLV for female***	***BLV³ for female in reproductive period***	***BLV³ for female under the age of 45***	***BLV³ for young workers***			
EC Directive 24 (1998)	700¹							EC Directive 24 (1998)
SCOEL	300²							SCOEL (2002)
Slovenia	400¹	300¹				<15¹	1.5¹	MDDSZ (2015)
Germany	400¹			300¹				TRGS 903 (2006)
United Kingdom	500³		250³		400³			Health and Safety (2002)
	600⁴		300⁴		500⁴			

ALAD = Delta-aminolaevulinic acid dehydratase, EP = Erythrocyte Protoporphyrin, BLV = biological limit value; ¹ binding biological limit value, ² recommended biological limit value, ³ action level, ⁴ suspension level

Table 5. Biological monitoring results regarding the types of technological processes

Types of technological processes	*Biological monitoring*								
	No. of checked workers N	*Pb in blood (μg/L)*		*Delta-aminolevulinic acid dehydratase in a liter of erythrocytes (U/LE)*		*Erythrocyte Protoporphyrin in a liter of erythrocytes (mg/LE)*		*8-hour TWA Pb concentration (mg/m³)*	
		Average (field)	*SD*	*Average (field)*	*SD*	*Average (field)*	*SD*	*Average (field)*	*SD*
Production of lead-acid batteries	403	165 (25-577)	100					**0.09 (0.001-0.9)**	0.22
Ore melting, refining, compound production and casting	65	120 (11-345)	79					0.02 (0.01-0.06)	0.01
Glass production	135	165 (38-340)	61	29.6 (15.1-50.9)		0.97 (0.44-2.85)	0.44	0.06 (0.01-0.6)	0.15
Lead-acid battery recycling	82	226 (23-397)	87					0.09 (0.01-0.33)	0.08
Work with metallic lead and its compounds	12	21 (7-38)	11	59.4 (24.0-81.3)	13,6	0.45 (0.24-0.76)	0.15	0.02 (0.01-0.04)	0.01
Pigment and paint production	19	32 (6-110)	22.6	50.5 (31.1-74.1)	12.0	0.47 (0.25-1.16)	0.21	0.02 (0.01-0.03)	0.01
Other industries (including pipes, lights and cables)	15	56 (9-205)	45.9	47.6 (21.2-55.2)	12.0	0.43 (0.27-0.69)	0.11	0.01 (0.005-0.015)	0.004
Badges, jewelery decoration and glassware decoration	4	71 (35-134)	37.7	48.2 (36.2 – 71.4)	14.0	0.49 (0.3-0.6)	0.11	0.005 (0.004-0.006)	0.001
Total	**735**	**6-577**							

When comparing Tables 3-5 it can be seen that biological monitoring was not carried out in all types of technological processes. The reason is the dilemma whether biological monitoring is necessary if workers are exposed to low lead concentrations. The answer was given in the Occupational Safety and Health Standards (OSHA, 29 CFR 1910.1025) defining the biological monitoring in case of exposure to the concentration amounting to 0.03 mg/m^3 for more than 30 days in a year. As the Republic of Slovenia does not have such a regulation this should be included when changing the legislation.

4.2.4. Inspection on organization of health and safety at work

Supervision identified several facts that reflected an inappropriate organization of safety and health, for example, high fluctuations in the biological monitoring values within an individual technological process, high values of lead and its compounds at workplaces, unsuitable training, the failure to use personal protective equipment etc. Workers used personal protection equipment unscrupulously as only 33.3 % were used in some sectors. The share of employers who taught their employees about lead and lead compounds risk ranged between 0 % and 100 %. The vacuum and ventilation systems were often insufficient in the case of increased production, therefore, increased lead and lead compounds concentrations appeared at some workplaces, as shown in previous sections. The appropriate noise supervision is important as an interaction may occur due to noise and lead exposure. Special attention should be paid to lead-acid batteries recycling as it comes to high temperatures in the smeltery, and the measures for physiological heat stress monitoring have to be defined.

4.3. Measures for exposure reduction

4.3.1. Technical safety measures

Those employers with the highest exposure of workers to lead and its compounds started to introduce various technical measures for reducing the concentrations. In the lead-acid battery production technological changes were introduced so that a storage desk with extraction from the bottom and lateral side was placed in the workplace, however, an exhaust duct for fresh air ventilation was placed above the workplace. Renewed measurements were carried out after these changes, yet concentrations did not decrease essentially. The automatic plate cleaning was then ensured by the employer and this is how they decreased the concentrations below the limit value. In the mechanical pilling up process of lead plates the employer introduced the local extraction, however, this measure did not lower the lead concentration in the work environment below the limit value; therefore the employer ensured additional mobile vacuum cleaners in this process phase. In the plate forming process – welding, the employer still needs to find technological solutions for the permanent emission reduction, and similarly in the wet filling process of small nets.

In the dry glassware cleaning process work was performed in a robot cell so workers were only temporary exposed to high concentrations. In the process of preparation compounds and potsherds the limit values have not been exceeded, nevertheless, the employer searched technological solutions for the reduction of lead emission by introducing the crystal glass production and substituting a dusty lead compound with the granulated one. Grinding crystal glass workers were exposed to lead oxide for the whole working time. In 2011 the employer renewed and changed the ventilation system in the premises for wet grinding. However, it was found out that the new ventilation system was not sufficient as the lead concentration in the stationary sampling procedure amounted up to 0.18 mg/m^3 (in the middle of the room), whereas in the personal sampling procedure of the workers' respiratory organs at the machine the concentrations ranged between 0.20 mg/m^3 and 0.61 mg/m^3.

Labour Inspectorate of the Republic of Slovenia required the ensurance of local extraction at all 64 workplaces where wet grinding was performed and the employer performed it in 2012. The concentrations decreased to 0.009 mg/m^3 in the stationary sampling procedure in the middle of the place, whereas in the personal sampling procedure of the workers' respiratory organs it amounted between 0.017 mg/m^3 and 0.018 mg/m^3. Due to the efficient local extraction the lead concentration in the work environment has decreased by more than 90 %.

4.3.2. Organizational safety measures

The implementation of the following organizational safety measures was advised to the employers:

- to divide technological premises by walls to avoid unnecessary workers' exposure.
- to reduce the duration and intensity of exposure with appropriate work and production scheduling.
- to implement sufficient hygienic measures including cleaning the dust.
- to inform workers about dangerous properties of lead as they would use the necessary personal protective equipment more carefully.

4.3.3. Individual safety measures

The reason for exceeded biological limit values was also the failure to use personal protective equipment for respiratory organs or its unsuitable usage, insufficient cleanness regarding the workwear change and insufficient hand washing as it was established that workers smokers have mostly exceeded limit values. At some workplaces workers used tight fitting masks for the respiratory tract protection (the full face mask FFP3, a half face mask or a respirator), however, employers did not ensure a quantities and quality mask or respirator fit testing. In case the personal protective equipment for the respiratory protection with tight fitting masks is chosen it is necessary to carry out a FIT testing (CEN, 2005; HSG 53, 2013; OSHA, 29 CFR 1910.134, 1998).

No violation of personal protective equipment use for body and hands was noticed during the supervision. However, it is necessary to check permanently if workers use the personal protective equipment which is directed according to their risk. Workers exposed to lead dust are obliged to wear overalls with hood which are impermeable to hard particles and protective gloves for mechanical risks. Workers exposed to lead during the lead melting process are obliged to wear heat and flame protective clothing and heat protective gloves. Due to potential contact with sulphuric acid workers in the production and recycling of lead-acid batteries are obliged to wear protective clothing against liquid chemicals and chemical resistant neoprene gloves.

The following additional cleanness measures were advised to the employers regarding lead and lead compounds: the white-black principle in the dressing and dining rooms, as well as in the resting rooms, daily change of personal protective equipment, washing the personal protective equipment, wearing long sleeves for body protection, washing working surfaces and transport pathways including the outdoor by using cleaning machines, insurance of regular internal control concerning the implementation of individual safety measures.

5. Conclusions

This article presented the development of the methodological approach for the comprehensive assessment and supervision of the workplace exposure to lead and its compounds. This is a general interdisciplinary approach comprising safety and health at work, analytical and general chemistry, chemical engineering, medicine and law.

Within the research it was established that workers in the lead-acid battery production were exposed to the highest lead concentrations which exceeded the limit values by 9 times; followed by the glass production. In average the highest lead values were in the blood of workers who recycle lead-acid batteries, within the production of lead-acid batteries and glass production. The research revealed that biological monitoring was carried out in an ununified way as in some activities only lead in blood was checked but no Delta-aminolevulinic acid dehydratase and no Erythrocyte Protoporphyrin in a liter of erythrocytes although required by by-laws.

The application of the proposed methodology provided, for the first time, a comprehensive evaluation of workers' exposure to lead within the entire Republic of Slovenia. It resulted in several improvements at the employers, and gave some most important instructions regarding individual regulations from this field.

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