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Original article

Assessment of exposure to metals in lead processing industries

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ABSTRACT

Inhalation of particulate matter in industrial environments has been associated with respiratory symptoms and lung diseases, which continues to lead to long- and short-term hazardous health effects on exposed subjects. The main objectives of this study were (a) to determine the dust exposure of workers from the lead industry in different operations and (b) to evaluate if the Exhaled Breath Condensate (EBC) can be used as a non-invasive tool to evaluate this exposure. Therefore, this cross-sectional study not only measured the exposure to Airborne Particulate Matter (APM) and to the associated elements but also analysed the EBC elemental composition. APM was collected in Ind1, Ind2, Offices and outdoor with Gent samplers, which delivers two size fractions: fine particulate matter (<2.5 μm), and coarse particulate matter (between 2.5 and 10 μm). EBC samples were collected from the workers and from a non-exposed group working in Offices. The techniques INAA and PIXE were used for the APM element characterization and ICP-MS for EBC elemental content. The $PM_{2.5}$ and $PM_{2.5-10}$ mass concentrations were significantly higher in the industries studied than in Offices and in the environment. At the industrial sites surveyed the coarse fraction dominated and both factories had different fingerprints: APM elements with higher expression were Pb, Sb, Na, Cl and Fe in Ind1 and Pb, Si, Br, Ca, Al, Cl and Na in Ind2. Most of these elements revealed a gradient of concentration where Ind.1 > Ind.2 > Offices and EBC revealed a clear translation of this exposure, suggesting the latter to be a potential good indicator of exposure to metals in occupational settings. Pb in EBC presented the most representative results. Even though EBC was found to reflect predominantly the inhaled coarser fraction it is more related to concentration levels of exposure than to the predominance of APM fraction.

The present study demonstrated not only the ability of EBC to reflect environmental exposure to metals but also the importance of measuring and characterizing different fractions of APM for a correct assessment.

Introduction

Epidemiological studies have consistently shown an association between Airborne Particulate Matter (APM) pollution and the number of deaths from cancer, cardiovascular and respiratory diseases (Brook et al., 2010; Pope, 2007). There is also evidence linking particulate air pollution, especially the fine particle fraction ($PM_{2.5}$, denoting PM < 2.5 μm diameter size), and increases in hospital admissions for respiratory and cardiovascular diseases (Zanobetti et al., 2009; Pope et al., 2008). Several studies demonstrated that particles can induce alveolar inflammation, with release of mediators capable of causing exacerbations of lung diseases and increasing blood coagulability in susceptible individuals, thus, also explaining the observed increases in cardiovascular deaths associated with urban pollution episodes (Brook et al., 2010). Epidemiological data from the USA suggested that rises of 10 μg m^{-3} in $PM_{2.5}$ are accompanied by an increase in relative mortality risk of about 4%, including elevated risks from both cardiopulmonary mortality (6%) and lung cancer mortality (8%) (Pope et al., 2002).

In industrial scenarios, particles are a major concern. Firstly, dust concentrations inside industries are very high comparing with the environment. Almeida et al. (2010) showed that PM_{10} concentrations in a foundry industry reached values 50 times higher than in the outdoor air. Secondly, there are more toxic compounds in the composition of the particles sampled inside the industries. In the same study, Almeida et al. (2010) measured concentrations of lead 35,000 times higher in PM_{10} sampled in the foundry industry than in the atmosphere. Thirdly, the majority of the people spend more time in the workplace (33% of the day) than in the outdoor (10% of the day) (USEPA, 1989; Oliveira Fernandes et al., 2009), therefore, exposure to pollutants in the workplace is more relevant than outdoor pollution.

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In industrial environments, particle size appears to be dependent on the source type. Among metal processing industries, the foundry/smelting and welding processes contribute mostly to the particles with lower granulometries (Wake et al., 2002), whereas cleaning processes, handwork and cut emit mainly coarse particles (e.g. Hlavay et al., 1992; Cheng et al., 2008). Moreover, the type of processes and the raw material used are associated with characteristic emission regarding the combination of metals and chemical compounds in each particle size fraction. Karlsen et al. (1992) showed the differences between the chemical composition and the morphology of particles provided from welding fumes and gridding dust.

Despite the technological requirements, imposed by safety regulations, that guarantee an improvement in indoor industrial air quality, workers continue to be excessively exposed to APM (e.g. Goldoni et al., 2004; Nawrot et al., 2008; Almeida et al., 2010; Félix et al., 2012). This occurs, not only due to the ineffectiveness or inexistence of equipment that promotes the extraction of particles or protects the workers individually, but also to the negligence of the subject himself.

Exhaled Breath Condensate (EBC) has been progressively considered a potential bioindicator of exposure especially fitted to occupational assessments due to several features: (1) it is non-invasive; (2) quickly collected; (3) representative of the organ of direct contact with the toxicant (the lungs); (4) it is not aggressive for the subject, allowing the possibility of repeated collections; (5) and easily analysed, without complex protocols of sample preparation (Hunt, 2002; Goldoni et al., 2004; Caglieri et al., 2006; Cao and Duan, 2006; Almeida et al., 2010; Gube et al., 2010; Hoffmeyer et al., 2011; Félix et al., 2012).

The aim of the present study was to determine the exposure level of workers in their workplace in two lead processing factories and evaluate the potential of EBC as bioindicator. For that, metal concentrations were measured both in the APM collected in different sites at the workplace and in the EBC of workers.

Methodology

Industry and study group

The present study was performed with the collaboration of two industries that process lead: Industry 1 (Ind1) that recycles batteries and Industry 2 (Ind2) that produces batteries. Although dealing with the same contaminant, both factories have distinct features, such as, physical processes and fabrication, number of employees, air extraction requirements and architectonics. The foundry industry (Ind1) has two contiguous areas in an open space, one with two furnaces for lead meltdown, and a second where workers re-melt lead with additives for refinement. The area of the factory is not a closed area due to material transfer requirements. The facility has fumes and dust extractors running, according to national regulations. There are no fixed work-posts. The second industry (Ind2) is a battery assembling facility where workplaces are divided into two categories: lead plates cut (automatic and manual); and assembling lines. Each workplace has air extractors for particle removal originated from the work activities. Workers alternate frequently in their workplace between automatic and manual cuts and within the several posts in the assembling line, but not between the two main categories of manufacturing. In both factories, air quality and appropriate personal safety measures are implemented as imposed by national regulations and all personnel is required to wear protective cloths, glasses and masks.

Ind1 labours 24 h a day, on three 8 h shifts and Ind2 operates only on two 8 h shifts. Workers from both factories labour five days a week with two intercalary resting days.

In Ind1, 17 workers participated in the study whereas in Ind2 83 workers were enrolled, all labouring for more than five years in the factory. For reference purposes, a group of 54 volunteers working in Offices and not exposed to gases, dusts or fumes in their working activity was also constituted. All subjects gave their informed consent to participate in the study and filled a questionnaire reporting on age, smoking habits, gender and past respiratory diseases (the selected did not report any respiratory pathologic history).

APM

Sampling

APM was collected with low volume Gent samplers (Maenhaut, 1992). Gent samplers were equipped with a PM_{10} pre-impactor stage and with a Stacked Filter Unit (SFU) of two stages, carrying 47 mm Nuclepore^{TM} polycarbonate filters. Air was sampled at 15–16 L min^{-1}, which allowed the collection of particles with aerodynamic diameter (AD) between 2.5 and 10 μm in the first stage and particles with AD < 2.5 μm in the second stage.

Sampling was carried out in factories using several Gent samples working in parallel. Samplers were operated during labouring period at 1.6 m high, which corresponds to the breathing height of workers, thereby ensuring the best representativeness of working conditions.

In Ind1 four samplers were used in parallel distributed throughout the factory, in representation of the whole labouring area. In Ind2, where activities are divided in different rooms and/or areas, four samplers were distributed along each manufacturing line (manual and automatic plates cut and assembly).

For reference purposes, sampling was also carried out in Offices placed in the same geographical area of the studied factories (30 km maximum distance) and in outdoor environment. In Offices one Gent sampler was used per flat. Also, in the outdoor environment only one Gent sampler was used (Almeida et al., 2005, 2006a).

Chemical analysis

The filter loads were measured by gravimetric means in a clean room (ISO7). Nuclepore filters were weighted using a 0.1 μg sensitivity balance (Mettler Toledo UMT5). Filter mass before and after sampling was obtained as the average of at least three measurements, assuring that the variation coefficient <5%.

Each filter was divided providing portions suited for elemental analysis by Particle Induced X-Ray Emission (PIXE) and by Instrumental Neutron Activation Analysis (INAA). The techniques are multi-elemental and complementary in terms of detected elements (Almeida et al., 2006b). The elements As, Br, Cd, Cr, Na, Sb, Se and Sn were determined by INAA and Al, Ca, Cl, Cu, Mn, Ni, Pb, Si, Ti and V by PIXE. K, Fe and Zn concentrations were measured by both techniques with a good agreement and therefore an average of both techniques was calculated for each sample.

Due logistic reasons, INAA analysis (Cornelis et al., 1976) of air sampling filters collected at Ind1 was performed in the Portuguese Research Reactor of ITN; and air sampling filters from Ind2 were analysed at the Higher Education Reactor of TU Delft, The Netherlands.

At ITN, the portion of the filter to be analysed by INAA was rolled up and put into a thin foil of aluminium and irradiated for 5-h at a thermal neutron flux of 1.03×10^{13} cm^{-2} s^{-1}. After irradiation the sample was removed from the aluminium foil and transferred to a polyethylene container. For each irradiated sample, two gamma spectra were measured with a hyperpure germanium detector: one spectrum 3 days after the irradiation and the other one after 4 weeks. The k_0-INAA method (De Corte, 1987) was used and 0.1% Au–Al discs were co-irradiated as comparators. The software k_0-IAEA was used to interpret the spectra and to calculate the concentrations.