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Urinary beryllium – a suitable tool for assessing occupational and environmental beryllium exposure?

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Abstract Objectives: The reasons for the slow progress and lack of new knowledge in the biological monitoring of beryllium (Be) are to be found in the presumed small number of working activities involving exposure to the metal, and the lack of adequate analytical methods. The reference values for urinary Be reported earlier in the literature appear to be too high, due to the poor specificity and sensitivity of the adopted methods. The aim of this study was to correlate Be air concentrations and Be urinary levels to ascertain whether the biological indicator was suitable for assessing occupational exposure to the metal. Methods: To investigate the relationship between the Be concentrations in air and those excreted in urine, we examined 65 metallurgical workers exposed to very low levels of the metal, and 30 control subjects. The exposed workers were employed in two electric steel plants and two copper alloy foundries. The alloys were produced in electric furnaces, starting with scrap containing Be as an impurity. The Be concentrations in the air were monitored by area samplers and the levels of Be in the urine of the workers were determined in samples taken at the end of the shift. Both determinations were carried out by ICP-MS. Results: The median airborne Be concentrations in the copper alloy plants were 0.27 μg/m³ in the furnace area and 0.31 μg/m³ in the casting area. Median values of 0.03 to 0.12 μg/m³ were determined in the steel plants, the relatively wide range probably due to differing amounts of Be in the scrap. Regression analysis was performed on the median values from four work areas and the corresponding urinary samples. A significant correlation was found for the relationship between external and internal exposure. The urinary Be levels were in the range between 0.12 and 0.15 μg/l with observation of the recommended TLV-TWA for inhalable dust of 0.2 μg/m³ (0.2 μg/l at the upper 95th percentile). Conclusions: Sufficient data are not currently available to be able to propose a BEI for urinary Be. Our results show that new investigations are necessary to improve the evaluation of dose indicators and the relationship between external and internal exposure to Be.

Key words Beryllium · Ambient and biological monitoring · Electric steel plant · Copper alloy foundries

Introduction

Ten years ago the monograph on Beryllium (Be) for the CEC series on “Biological Indicators for the Assessment of Human Exposure to Industrial Chemicals” [6], stated that the urinary excretion of Be was higher in exposed subjects than in the general population, although occupational exposure to the metal was low and the urinary Be levels were higher at the end of the work shift. The relationships between internal and external exposure and between internal exposure and (early) effects were, however, not known, and the need for more research was emphasized.

Advancements in the biological monitoring of Be have been quite modest over the past decade, as demonstrated by the small number of papers published on the subject. When, for example, the articles quoted in the period 1989–1999 in the MED LINE data bank are examined, one finds few references regarding Be and human health [1, 8, 9, 14, 18–20, 25]. Some of these articles deal with ambient pollution or Be diseases, but without using any biological indicator of the metal.

Two papers mention the biological monitoring of Be: the first in the context of an investigation about
the urinary reference values for 13 metals in the general population of the USA [24], the second in the context of the theoretical significance of biomonitoring for carcinogenic metals [25]. In addition we found a recent article on gemstone cutters exposed to beryls in which urinary Be is specifically investigated as a biological indicator in occupationally exposed subjects [30].

In our opinion the range of industrial processes in which occupational exposure to Be occurs has, on the contrary, increased over the past two decades, and it is very likely that unknown or not easily identifiable sources of exposure to the metal exist. This is confirmed by the recent evaluation carried out by Kauppinen et al. [17] of the number of EU workers exposed to carcinogens: in this review the number of individuals occupationally exposed to Be is given as 67,000 and Be is classed as a carcinogen of the same potency as ceramic fibres and styrene-7,8-oxide.

The environmental sources of Be may also be of interest for the general population, in particular the emissions from plants directly or indirectly processing Be, from incinerators and from electric power plants using coal or mineral oil. There are, however, conflicting data about the presence and amount of Be in the environment. Another important non-occupational source of the element is tobacco smoke [1, 11, 26].

The effects of Be on human health have long been a concern of occupational medicine. Of the diseases related to Be, only the acute Be pulmonary syndrome is clearly dose-related, being caused by short-term exposure to high concentrations of Be [12]. Pulmonary chronic Be disease (CBD), on the other hand, is an immunologically mediated syndrome, defined as the occurrence of lymphocyte proliferation coupled with the presence of alveolar granulomas. This proliferation can be detected using the beryllium lymphocyte proliferation test (BLPT) and the granulomas are harvested by transbronchial biopsy [22, 23].

As suggested some years ago [13], it has not been possible to diagnose many cases of disease induced by exposure to Be. Particularly with cases of pulmonary granulomatous diseases, it is advisable to investigate the possible existence of occupational exposure to Be. At a symposium organized by the ACGIH entitled “Beryllium: Effect on worker health”, held on September 23rd, 1999, in Washington, D.C., it was suggested that positive results in the BLPT is the most appropriate effect indicator (critical endpoint) upon which to base the occupational limit for Be in air [2].

The other well-known effect of occupational exposure to Be is lung cancer. For this reason Be is classified as an A1 human carcinogen by the ACGIH, and a carcinogen of group 1 by the IARC. When we consider CBD and cancer, exposure to low and very low concentrations may be of great interest, and the ACGIH is going to propose a “notice of intended changes” for Be, with a TLV-TWA of 0.2 µg/m³ as inhalable particulate, instead of the current TLV-TWA of 2 µg/m³ [2].

Crucial for the assessment of low exposures on the basis of ambient and biological monitoring, is the availability of adequate analytical methods. For the biological monitoring of subjects with low exposure to Be, the sensitivity and specificity of the analytical method determines the accuracy with which the amount of Be in biological matrices is measured.

The determination of Be has generally been performed using graphite furnace atomic absorption spectrometry (GFAAS) and more recently by inductively coupled plasma atomic emission or mass spectrometry (ICP-OES/MS).

Direct analysis by GFAAS is, however, the technique most frequently used for biological samples, and when correctly performed allows detection limits to be reached adequate to determine Be concentrations in urine, for example in the range 0.1–1 µg/l [4, 7, 15, 24].

Therefore, analytical performance and the identification of possible exposures are critical for Be biological monitoring, and would appear mandatory for assessing the new threshold limits for Be in air and (possibly) related biological limit values.

To assess the feasibility of the biological monitoring of occupational exposure to Be it would therefore be useful to take into consideration some of the investigations carried out in working environments with low and very low exposure to the metal as a starting point.

Materials and methods

Groups and workplaces

We investigated four groups of metallurgical workers, and a control group of mechanical workers employed in activities without exposure to metals (in particular Be). The exposed workers (n = 65) were employed in two steel electric plants and in two copper alloy foundries. Steel and non-copper alloys were produced in electric furnaces starting with scrap. In each plant we examined the more highly polluted working areas: furnace charging, melting and casting.

The furnace workers were exposed to metals during the following process phases: selection and preparation of raw materials, scrap charging, refining and melting.

The casters were employed in pouring of molten metal into the tundishes or the ingots. Both groups were therefore exposed to a mixture of metals, including Be. Possible sources of Be is the presence of the element in dust and smoke from raw materials or from steel and copper alloys during and after the melting process.

The exposure to dust and smoke containing the metals was not constant during the work shift and all workers used respiratory protective devices irregularly.

Our control group was composed of workers employed in mechanical activities (assembling, finishing trucks) known not to be exposed to metals.

The general characteristics of the groups investigated are reported in Table 1.

Ambient monitoring

The very low Be concentrations in the air meant that great volumes had to be sampled. Consequently, personal samplers could not be used. Stationary samplers with a flow rate of 10–15 l/min were used for periods of 5–6 h.