Beryllium Decontamination with Different Solvents on Different Structures

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Received 28 February 2009; in final form 24 June 2009; published online 15 July 2009

The objective of the present work was to estimate the efficiency of moistened wipes in removing beryllium with different solutions including Citranox™, Alconox™, NaCl 5%, Resolve™, and Ledizolv™ on various types of surfaces such as unpainted metal, wood frames, painted metal, concrete, painted concrete, and Plexiglas™ from three different occupational settings. Of the three plants that were investigated, only surfaces in the aluminium smelter were decontaminated down to the clearance reference level of 0.2 μg 100 cm⁻², with all the solvents used. In the machine tooling and milling department, the clearance level of 0.2 μg 100 cm⁻² was reached after the three decontaminations, with all the solvents. In the machine plant for the military, aerospace, and telecommunications industries, the beryllium concentrations on the concrete wall, before decontamination with the high-pressure gun, were usually >3 μg 100 cm⁻², and concentrations as high as 31 μg 100 cm⁻² were measured. After the high-pressure cleanup, the beryllium concentrations were sometimes reduced by a factor of 10, but never reached the clearance level. Beryllium compounds that had adhered to most types of structures that we attempted to decontaminate were reduced to below the clearance reference value except on concrete floors. There did not seem to be any difference between the decontamination actions for all the solvents used in this study.

Keywords: Alconox™; beryllium; Citranox™; Ghost Wipe™; Ledizolv™; NaCl 5%; Resolve™; surfaces

INTRODUCTION

Chavalitnitikul and Levin (1984) suggested that, in some cases, dermal contact with contaminated surfaces may be the most significant pathway for hazardous substances to enter the body. These surface contaminants may enter the body directly by percutaneous absorption and/or by ingestion (Caplan, 1993; Klingner and McCorkle, 1994). Surface decontamination is an important aspect in preventing worker exposure to dangerous contaminants. Characterizing the surfaces that are potentially available for contact through dust or residue surface loadings (μg cm⁻²) or contaminant concentrations (μg g⁻¹) provides inferential information that could be related to exposure and ultimately to dose (Rodes et al., 2001).

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A few articles have explored the decontamination of beryllium-contaminated surfaces. Mitchell and Eutsler (1964) tested the efficacy of two decontamination methods on beryllium-contaminated wood floors. These authors showed that contamination near the surface of the wood could be removed. They also determined that beryllium had impregnated into the wood, even though the concentration near the surface was acceptable. Others reported that a combination of sandblasting, manual scraping, hydrobrushing, and manual scrubbing resulted in residual surface contamination <1 μg Be 100 cm⁻² in a large building significantly contaminated with beryllium (Gronka et al., 1971). Other researchers have investigated the decontamination of reactors, ships, and fusion chambers, but decontamination procedures have focused specifically on these types of equipment (Haigh and Pick, 1997; Lumia et al., 2003; Raffestin and Teillerie, 2005).
The Occupational Health and Safety Administration technical manual (OSHA, 2008) recommends the wipe sampling technique as an important tool for anticipating exposure, for evaluating personal protective equipment, and for evaluating the efficacy of cleaning and decontamination programmes. However, wipe sampling procedures do not have the degree of precision generally accepted in industrial hygiene practice (Lichtenwalner, 1992). A single wipe sample seems inefficient in collecting the entire surface contamination (Chavalitnitikul and Levin, 1984). Nevertheless, the wipe remains a valuable tool for estimating surface contamination (Farfel et al., 1994; Lamphere et al., 1995; Wheeler and Stancliffe, 1998).

Rosyter and Fish (1967) showed that the collection efficacy of the wipe method was affected by the type of surface and could vary from 23.5 to 74.5% for radioactive particulates. This efficiency and variability of manual wipe sampling was found to be affected by a number of factors (Wheeler and Stancliffe, 1998) such as (i) concentration and particle size distribution of the surface contaminant, (ii) surface roughness and porosity, (iii) area of surface sampled and wiping pressure applied, and (iv) the surveyor. Particle size not only affects the ability of a particle to dislodge but also can influence the probability of retention of the particle on the skin surface after its transfer (Rodes et al., 2001).

Regulatory surface contamination limits at US Department of Energy sites were established by the Chronic Beryllium Disease Prevention Programme rule (DOE, 1999). These limits are 0.2 µg 100 cm⁻² for release of beryllium-contaminated equipment to a non-beryllium area and 3 µg 100 cm⁻² as a housekeeping limit inside areas where beryllium work is performed. In a recent paper (Dufresne et al., 2009), we estimated the efficiency of moistened wipes with different solutions including Citranox™, Alconox™, Luminox™, Fanstastik™, Z-99™, water, and NaCl 5% in the recovery, decontamination, and frequency of surface rubbing of beryllium compounds present on smooth and porous surfaces of moulds or plates made of beryllium–copper (Be–Cu) alloy. Citranox, an acidic solution, was the most efficient cleansing solution and removed the highest concentration of beryllium on the surfaces of moulds and plates. We concluded that a layer of beryllium oxide was continuously formed on materials made of Be–Cu alloy and that decontamination of these surfaces was not achievable to concentrations <0.2 µg 100 cm⁻².

The objective of the present work was to estimate the efficiency of moistened wipes for removing beryllium with different solutions including Citranox, Alconox, NaCl 5%, Resolve, and Ledizolv on various types of surfaces such as unpainted metal, wood frames, painted metal, concrete, painted concrete, and Plexiglas™ in three different industrial settings.

MATERIAL AND METHODS

The decontamination was performed on surfaces of structures in three occupational settings (see Fig. 1 showing some of the structures):

(i) An aluminium smelter, in which a painted concrete wall, a painted metal wall, and a wood frame were cleaned. Particles collected from these surfaces appeared to have settled out. There did not appear to be any disturbance due to compression, mechanical equipment, or chemical reactions of the particles that deposited on the structures. It was felt that they may have been re-suspended by an air draft.

(ii) The machine tooling and milling department of a plant producing extruded plastic bottles, which was undergoing a decontamination process. A contractor had the mandate to vacuum the structures with a system equipped with an high efficiency particulate air (HEPA) filter and to wash them with a solvent called Resolve. A table covered with a sheet of Plexiglas, a metal table, and a ‘dirty’ concrete floor were decontaminated in this setting. A cutting oil residue was visible on the surfaces that were sampled or decontaminated with a wipe. The cutting oil residue was very prominent on the concrete cement floor.

(iii) A large machine shop undergoing decontamination previously used to produce metal parts for the military, aerospace, and telecommunications industries. The painted metal frames (horizontal and vertical) of an overhead crane, a ‘damp and greasy’ cement floor before and after decontamination with Ledizolv solution and a high-pressure gun, a painted concrete wall, and an unpainted cement wall.

The decontamination protocol included the following water-based cleaning solutions:

(i) Citranox (2%) (Alconox, White Plains, NY, USA): citric acid in solution (Alconox, 2008), pH = 2.6;
(ii) Alconox (1%) (Alconox): solution of sodium decylbenzenesulfonate, sodium carbonate, tetrasodium pyrophosphate, and sodium phosphate (Alconox, 2009), pH = 9.2;
(iii) NaCl 5% solution; pH = 6.57;
(iv) Tap water (Montréal);
(v) Resolve (1%): (Dustbane Products, Ottawa, ON, USA): solution of propylene glycol n-butyl ether, dipropylene glycol methylether, trisodium phosphate, and ethoxylated alcohols C₁₂–C₁₅ (Dunlop, 2007), pH = 10.2; and
(vi) Ledizolv (LSZ, White Plains, NY, USA), an anionic liquid detergent (LSZ, 2001).

GhostWipes™ were selected for sampling surfaces. The sampling technique was from Institut de Recherche Robert-Sauvé en Santé et Sécurité du Travail Protocol I-MAT-012 (Dion and Perrault, 2005), which is based on the National Institute for...
Occupational Safety and Health method 9102 (NIOSH, 1994) and on the American Society for Testing and Materials method (ASTM D6966) (ASTM International, 2005). The three operators applied a steady pressure and made an effort to maintain the same pressure through all the successive samples. Surfaces were wiped according to patterns made of two ‘S’ curves, in a vertical and horizontal fashion with each movement exposing a fresh surface of the wipe by folding it and finally completing the sampling at the periphery of the surface to be sampled (Dion and Perrault, 2005). Results were expressed in units of micrograms of total beryllium per 100 square centimetres (µg 100 cm\(^{-2}\)).

The sequence of wipe sampling was as follows: (i) a wipe sample to measure the background concentration and to test the homogeneity of the chemical on equivalent surfaces (W1), (ii) sequence of cleaning (or decontamination) with three consecutive wipes dipped in the studied solution (D1–D3), and (iii) a wipe sample after the decontamination (W2).

RESULTS

Table 1 presents the results for the beryllium concentration of surfaces sampled in an aluminium smelter. Most of the beryllium deposited on the concrete wall was collected with the first wipe sample (W1). For the painted metal wall and particularly for the wood frame, with all the solvents, significant amounts of beryllium were still extracted during the decontamination (D1–D3). However, no beryllium was collected (<0.05 µg 100 cm\(^{-2}\)) with the final wipe sample (W2) after the three decontaminations with all the solvents.

Table 2 presents the results for the beryllium concentration of surfaces in the machining department of the plastics industry. While the beryllium concentration decreased to <0.2 µg 100 cm\(^{-2}\) (W2) for the Plexiglas and the metal table, beryllium still remained and the concentrations were above the clearance value with all the solvents that were used on the concrete floor.

Table 3 presents the results for the beryllium concentration on surfaces in the machining plant for the military, aerospace, and telecommunications industries. As expected, for the horizontal and vertical frames of the overhead crane, prior to cleaning, the horizontal surface contained more beryllium than the vertical surface. All solvents were efficient in bringing the beryllium concentration down to 0.2 µg 100 cm\(^{-2}\) after the workers had vacuumed the structures with a system equipped with an HEPA filter and before they had cleaned the frame with a high-pressure gun supplied with the Ledizolv solution. It should be noted, however, that beryllium was
not detected for most of the samples collected with any of the solvents used. High beryllium concentrations were sampled or were present in the wipes used to decontaminate the concrete floor before it was cleaned with the high-pressure gun. For example, concentrations as high as 20 μg 100 cm⁻² with Citranox and 26 μg 100 cm⁻² with Ledizolv™ solvents were collected with the wipes dipped in the solution (D1). Even after the thorough decontamination with the high-pressure gun, beryllium concentrations remained >0.2 μg 100 cm⁻², the highest one having a residual contamination of 2.30 μg 100 cm⁻² (W2 with Alconox). Decontamination of the painted concrete wall and the unpainted wall allowed beryllium concentrations of 0.2 μg 100 cm⁻² to be reached except for the W2 wipe sample which was 0.31 μg 100 cm⁻² for the Citranox solution.

DISCUSSION

Of the three plants investigated, only surfaces in the aluminium smelter (Table 1) seemed to have been decontaminated down to 0.2 μg 100 cm⁻², with all the solvents used: Citranox, Alconox, the solution of NaCl 5%, and tap water. One explanation of the decontamination efficiency may be that the deposited aerosol was loose, with minimum adherence to the surfaces. Interestingly, the aerosol that adhered to the wood frame showed more resistance in the removal process. This observation is demonstrated by the presence of beryllium in the three decontamination steps. Nevertheless, most beryllium concentrations in the second or third decontamination were <0.2 μg 100 cm⁻². Even if contamination near the surface was absent, there was still a possibility that beryllium compound had impregnated the wood as shown in a previous study (Mitchell and Eutsler, 1964).

In the machine tooling and milling department (Table 2), surfaces that were decontaminated appeared ‘greasy’, and deposited aerosols seemed to adhere strongly to the surfaces. The level of 0.2 μg 100 cm⁻² was attained after the three decontaminations, with all solvents, i.e. Citranox, Alconox, and Resolve, only for the smooth surfaces, the table covered with a Plexiglas sheet, as well as the metal table. The greasy appearance, most likely produced by layers of cutting oil deposited over time, made the release of the beryllium particles adhering to these surfaces more difficult. For the concrete floor, the level of 0.2 μg 100 cm⁻² was not achieved after the sequence of three decontaminations with any of the solvents. The highest beryllium concentration...
was measured during the first decontamination with the Resolve solvent (D1). At the present time, we cannot speculate which solvent was the best for removing beryllium. However, it is clear that more than three decontaminations were needed to achieve the clearance level of 0.2 μg 100 cm⁻² for the concrete floor.

In the machine plant for the military, aerospace, and telecommunications industries (Table 3), there was an opportunity to measure the level of beryllium contamination on a concrete floor before and after the decontamination of surfaces with a high-pressure gun fed with the Ledizolv solvent. The beryllium concentrations on the concrete wall, before decontamination with the high-pressure gun, were usually >3 μg 100 cm⁻² on the wipe used before decontamination with all the solvents (W1). However, after the high-pressure cleanup, beryllium concentrations were reduced but never reached 0.2 μg 100 cm⁻² as shown with all the wipes sampled after the third decontamination (W2).

<table>
<thead>
<tr>
<th>Solvents</th>
<th>Step</th>
<th>Concrete floor before high-pressure decontamination</th>
<th>Concrete floor after high-pressure decontamination</th>
<th>Horizontal painted metal frame of overhead crane after dust</th>
<th>Vertical painted metal frame of overhead crane after vacuumed dust</th>
<th>Concrete painted wall before high-pressure decontamination</th>
<th>Concrete wall after high-pressure decontamination</th>
</tr>
</thead>
<tbody>
<tr>
<td>Citranox</td>
<td>W1</td>
<td>17</td>
<td>0.40</td>
<td>3.7</td>
<td>&lt;0.05</td>
<td>3.1</td>
<td>&lt;0.05</td>
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<td></td>
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<td>&lt;0.05</td>
<td>0.17</td>
<td>&lt;0.05</td>
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<tr>
<td></td>
<td>D2</td>
<td>12</td>
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<td>0.27</td>
<td>&lt;0.05</td>
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<td>D3</td>
<td>6.2</td>
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<td>&lt;0.05</td>
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<td>Alconox</td>
<td>W1</td>
<td>18</td>
<td>0.65</td>
<td>5.0</td>
<td>0.07</td>
<td>3.3</td>
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<td>0.87</td>
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<td>0.33</td>
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<td>2.7</td>
<td>0.35</td>
<td>&lt;0.05</td>
<td>0.12</td>
<td>&lt;0.05</td>
</tr>
<tr>
<td></td>
<td>W2</td>
<td>3.7</td>
<td>2.3</td>
<td>0.07</td>
<td>&lt;0.05</td>
<td>0.08</td>
<td>&lt;0.05</td>
</tr>
<tr>
<td>Tap water</td>
<td>W1</td>
<td>22</td>
<td>0.79</td>
<td>2.8</td>
<td>0.07</td>
<td>3.5</td>
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<tr>
<td></td>
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<td>0.32</td>
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<td>0.07</td>
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<tr>
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<td>7.8</td>
<td>1.3</td>
<td>0.16</td>
<td>&lt;0.05</td>
<td>0.08</td>
<td>&lt;0.05</td>
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<td>Ledizolv</td>
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<td>&lt;0.05</td>
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<td>0.31</td>
<td>&lt;0.05</td>
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<tr>
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<td>0.29</td>
<td>&lt;0.05</td>
<td>0.12</td>
<td>&lt;0.05</td>
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<tr>
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<td>D3</td>
<td>16</td>
<td>0.40</td>
<td>0.20</td>
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<tr>
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<td>0.41</td>
<td>TD</td>
<td>&lt;0.05</td>
<td>0.17</td>
<td>&lt;0.05</td>
</tr>
</tbody>
</table>

W1 = wipe sample to measure the background concentration and to test the homogeneity of beryllium on equivalent surfaces; D2, D3, D4 = decontamination (or cleaning) with three consecutive wipes dipped in the studied solution; W2 = final wipe sample after decontamination; TD = technical difficulties; <0.05 = below the limit of detection.

Table 3. Beryllium recovery in the machining plant for the military, aerospace and telecommunications industries

From the concrete floor after high-pressure decontamination. At the present time, we do not have a clear explanation for this observation.

Conversely, on less porous surfaces such as the painted frames of the overhead crane, the painted concrete wall, or on less damaged surfaces such as the unpainted cement wall, the beryllium concentrations were more easily reduced to 0.2 μg 100 cm⁻².

In the present study, the type of surface was found to be a significant factor in beryllium removal, similar to that shown for lead removal (Lewis et al., 2006). Our study did not allow the effectiveness of decontamination before and after vacuuming to be observed in the machine shop. However, we could study the effect of the high-pressure gun procedure on decontamination of the concrete floor (in the plant producing metal parts) and it was seen that the effectiveness of the procedure was not demonstrated on this rough surface. Vacuum pre-treatment has been shown to be efficient for lead decontamination in houses (Yiin et al., 2004). However, we believe that in an industrial environment such as a machine shop, this process may have limited effect owing to the residue of cutting oil aerosol, which seems to increase the adherence of particulates on surfaces. Nevertheless, vacuuming reduces dust loading and as a secondary effect increases the effectiveness of sampling with a wipe.
It should be emphasized that particle and surface interactions are governed by forces such as molecular interactions, electrostatic interactions, liquid bridges, double-layer repulsion, and chemical bonds such as polar and metallic bonds (Ranade, 1987). These factors may directly influence increased particle adherence and may indirectly make it difficult to estimate the real concentration of contaminants on surfaces.

CONCLUSION

For the three companies with beryllium abatement, almost all post-cleaning beryllium loading of surfaces made of unpainted metal, wood frames, painted metal, concrete, painted concrete, and Plexiglas were <0.2 μg 100 cm⁻² using the cleaning solvents such as Citranox, Alconox, Ledizolv, Resolve, and 5% NaCl. However, beryllium concentrations were over the reference value for the concrete or cement floor with oil residue and with all solvents used on this type of structure. The combined effect of settled sticky cutting oil aerosol and the porosity of the concrete floor were most likely determinants that contributed to keep hold of beryllium compounds.

There did not seem to be any difference in decontamination for all the solvents used in this study. At the present time, no evidence has been found to support the use of a specific solvent.

The wipe method is a common method for sampling surfaces for beryllium contamination and is an effective method for non-porous, smooth surfaces.

Although workers risk assessment was not addressed in this paper, training and proper use of personal protective equipment are important determinants of exposure for workers who perform maintenance and cleanup operations in large-scale settings. A variety of hygiene measures are required to implement the transfer of contaminated materials from the work zone. For example, the entire work zone must be kept under negative pressure, access to the site must be restricted, and workers must shower before leaving the contaminated work zone.

FUNDING

Institut de recherche Robert-Sauvé en santé et sécurité du travail (G206745 IRSST 099-303); Évaluation des paramètres de surveillance environnementale des travailleurs exposés au beryllium (G213800 IRSST 099-302); Guide de nettoyage et de décontamination des lieux de travail où il y a présence de beryllium.

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