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# **Beryllium Surface Levels in a Military Ammunition Plant**

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This study evaluated the presence of beryllium surface contamination in a U.S. conventional munitions plant as an indicator of possible past beryllium airborne and skin exposure and used these measurements to classify job categories by potential level of exposure. Surface samples were collected from production and nonproduction areas of the plant and at regional industrial reference sites with no known history of beryllium use. Surface samples of premoistened wiping material were analyzed for beryllium mass content using inductively coupled plasma-atomic emission spectroscopy (ICP-AES) and results expressed as micrograms of beryllium per 100 square centimeters ( $\mu g/100 \text{ cm}^2$ ). Beryllium was detected in 87% of samples collected at the munitions plant and in 72% of the samples collected at regional reference sites. Two munitions plant samples from areas near sanders and grinders were above 3.0  $\mu g/100 \text{ cm}^2$  (U.S. Department of Energy surface contamination limit). The highest surface level found at the reference sites was 0.44  $\mu g/100 \text{ cm}^2$ . Workers in areas where beryllium-containing alloy tools were sanded or ground, but not other work areas, may have been exposed to airborne beryllium concentrations above levels encountered in other industries where metal work is conducted. Surface sampling provided information useful for categorizing munitions plant jobs by level of past beryllium airborne and skin exposure and, subsequently, for identifying employees within exposure strata to be screened for beryllium sensitization.

Keywords airborne particles, munitions workers, sanding and grinding, surface sampling

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### INTRODUCTION

C onventional (non-nuclear) munitions workers are exposed to a variety of toxicants, including organonitrogen compounds, solvents, metals, and depleted uranium. The metal beryllium typically has not been a component of conventional munitions, but munitions workers often use nonsparking tools made from beryllium-copper alloys to reduce the risk of explosion. The addition of 2% to 3% beryllium to copper

makes a nonmagnetic alloy six times stronger than pure copper; beryllium-copper alloys are used to make nonsparking tools for use in situations that constitute a fire or explosion hazard.<sup>(1)</sup> Chronic beryllium disease (CBD), a debilitating lung disease characterized by granulomas in the lungs and mediastinal lymph nodes, is caused by inhaling airborne beryllium particles.<sup>(2)</sup> In the early stages, CBD may be asymptomatic with only radiographic abnormalities or immunologic responses.<sup>(3)</sup> CBD generally has been associated with industrial exposures; however, CBD cases have occurred among family members of beryllium workers and residents living near beryllium refineries.<sup>(4)</sup> Ambient atmospheric concentrations of beryllium have not been reported to cause CBD.

Sensitization to beryllium is a precursor to CBD, and beryllium sensitization may progress to CBD.<sup>(5)</sup> Skin exposure also may lead to beryllium sensitization.<sup>(6,7)</sup> Sensitization to beryllium is measured by testing the increased proliferation of lymphocytes from venous blood samples—beryllium lymphocyte proliferation test (BeLPT).<sup>(8)</sup>

Henneberger et al.<sup>(9)</sup> estimated that as many as 134,000 workers in government and private industry are potentially exposed to beryllium in the United States. These workers include approximately 1500 in the primary beryllium industry and 26,500 Department of Energy (DOE) or Department of Defense (DOD) workers. Although munitions workers were not mentioned specifically by these investigators, machinists, grinders, and tool and die makers in a variety of other industries were included in their estimates of workers potentially exposed to airborne beryllium.

The plant in which this study was conducted has been manufacturing conventional munitions at its present location in the Midwest since 1941. Conventional missile warheads and a variety of large caliber tank ammunitions, mines, mortars, artillery shells, demolition charges, and weapons component parts are produced at the plant. In addition, this plant is a demilitarization site where obsolete ordnance is destroyed.

Beryllium was not used as a component in the production of conventional munitions, but beryllium-containing, nonsparking tools such as hammers, punches, and chisels were used for many years, possibly since the earliest days of production. The tools were used throughout the plant for a variety of functions where spark control was critical. As the tools deformed with routine use, they were taken to the main machine shop or several smaller maintenance shops on production lines to be dressed, i.e., sanded or ground, to re-create a point, edge, or smooth surface on the tools. Between 1999 and 2004, nonsparking beryllium alloy tools were phased out of use at the plant and replaced with aluminum-bronze alloy tools. No other industrial sources of beryllium exposure have been documented in the conventional weapons areas of the plant, but beryllium was present in components of the nuclear weapons assembled in one production line of the plant. Nuclear weapons were assembled on this production line from 1947 until 1975. This production line was overseen by the Atomic Energy Commission (AEC) and was separate from the conventional weapons production lines of the plant.

This article describes the results of an industrial hygiene survey for potential past beryllium exposure, with a focus on the use and maintenance of nonsparking tools. This survey was conducted as part of a larger health outcomes study of conventional munitions workers at the plant. The objective of the survey was to use surface sampling to investigate the presence and level of beryllium on surfaces in work areas throughout the plant. This data was used to identify work areas and, thereby, job categories that historically had potential beryllium exposure. The sampling results were used to index past and current job categories into beryllium exposure strata. Current and former workers were selected subsequently from exposure strata to assess sensitization to beryllium using the Be-LPT. These surface samples were compared with samples at reference sites that were not known to have used berylliumcontaining materials. The surface sample results were also compared with contamination housekeeping levels established by the U.S. DOE for beryllium on surfaces.<sup>(10)</sup>

#### **METHODS**

#### **Description of Sampling Locations**

Surface wipe samples were collected from 12 munitions plant buildings that housed maintenance, production, support, and administration activities. Targeted areas included the main machine shop where beryllium alloy tools reportedly were dressed most frequently; support maintenance shops for which primary activities did not involve machining (automotive, crafts, railroad); a sample of production lines including their respective small maintenance shops and break rooms; and nonproduction areas including cafeterias, a change house, a laundry facility, and an administration (civilian personnel) building. Sample locations included buildings still in use and others that had not been used for several years.

Wipe samples also were collected at six reference companies to determine background beryllium concentrations to which munitions plant results could be compared. Reference sites were located within a 120-km radius of the plant and represented the following industries: machining, metalworking, automotive work, and paper processing. Two of the sites have been operating in the same location for more than 40 years. To qualify as a comparison site, businesses were screened for prior work with beryllium or beryllium-alloy materials. Management representatives at all six reference sites stated that to their knowledge, beryllium was never used in tools or components at their facilities. Management at each site was familiar with the metal as well as the occupational hazard it presented.

At each location, wipe samples were collected from surfaces with visible dust accumulation judged to have been undisturbed by housekeeping, production, or maintenance activities for relatively long periods of time. In most cases, employees and management representatives estimated surface dust probably represented years to decades of accumulation. These sampling locations included, but were not limited to, suspended light fixtures, tops of cabinets or equipment, door frames or window ledges, and supply air diffusers. When possible, wipe samples also were collected from employee break rooms located near work areas. Dust on most of the sampled areas was from elevated surfaces approximately 180 to 250 cm above the floor and 80 to 150 cm above machine surfaces. Therefore, the accumulated dust likely occurred by airborne deposition, representing particulate matter that at some time was suspended in air and could have been inhaled.

#### Sampling and Analytical Methods

The methods used to collect surface samples for beryllium were the same as those used by Sanderson in a previous study.<sup>(11)</sup> The methods used in that study were cited by the DOE in *Chronic Beryllium Disease Prevention Program; Final Rule* as the recommended method to conduct surface wipe samples for beryllium contamination.<sup>(10)</sup>

Surface wipe samples were collected with individually packaged nonwoven fabric sheets (Ghost Wipes) manufactured by Environmental Express (Mount Pleasant, S.C.). These wipes are 15-cm  $\times$  15-cm sheets premoistened with deionized water that meet ASTM Standard E1792 specifications for sampling materials for lead and have been used successfully on other metals, including beryllium, as specified in Occupational Safety and Health Administration (OSHA) Method ID-125G, Addendum B.<sup>(12,13)</sup> All sampler sheets were from the same lot.

Using a clean nitrile glove for each sample to prevent crosscontamination, a single wet wipe folded in half was firmly drawn across the sample surface using horizontal strokes. The sample was then folded inward upon itself, and the same surface was wiped using equal pressure as before using vertical strokes. Finally, the wipe was folded upon itself into a quarter, and the surface was wiped using diagonal strokes. The sample was then folded again into an eighth so that the collected particles were inward to minimize material loss to the container wall, and the sample was placed into a prelabeled container. Only two individuals collected the samples (Sanderson and Ott) using the same sampling technique.

The sample area was measured with a flexible tape and recorded, along with a description of the sample location, elevation, and distance to any known potential

TABLE I.	Beryllium Surface	<b>Contamination</b>	Criteria Used by	y U.S. De	partment of Energy
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Limit	Reference				
$0.2 \ \mu g/100 \ cm^2$	10 CFR 850.31 release criteria level				
	DOE surface concentration limit to determine items acceptable for release to public.				
	Surface contamination must not exceed 0.2 $\mu$ g/100 cm <sup>2</sup> when released to the public for nonberyllium use.				
$3.0 \ \mu g/100 \ cm^2$	10 CFR 850.30 surface contamination limit				
-	Surfaces contaminated with beryllium dusts and waste must not exceed a removable contamination level of 3 $\mu g/100 \text{ cm}^2$ during non-operational periods.				
	Employers must provide protective clothing and equipment where surface levels exceed 3.0 $\mu$ g/100 cm <sup>2</sup> .				
	Housekeeping efforts must keep surface contamination at or below this level during nonoperational hours.				

beryllium-generating sources such as sanders and grinders. Sample areas ranged from 24 to 744 cm<sup>2</sup>. On each sampling day, wipe samples that contacted no contaminated surfaces were submitted as field blanks and handled in the same manner as field samples. Field blanks were submitted at approximately 10% of the field sampling rate. Submitted samples and chainof-custody forms were shipped overnight to the analytical laboratory.

An AIHA-accredited laboratory analyzed field samples and blanks per National Institute for Occupational Safety and Health (NIOSH) Method 7300 (inductively coupled plasmaatomic emission spectroscopy).<sup>(14)</sup> The analytical limit of quantitation (LOQ) was 0.02 micrograms ( $\mu$ g) beryllium per sample unless stated otherwise. Wipe sample results were provided as micrograms of beryllium per sample and converted to surface concentrations as micrograms beryllium per 100 square centimeters ( $\mu$ g/100 cm<sup>2</sup>).

Measurable surface beryllium concentrations were divided into three categories based on two surface criteria cited by the DOE standard: Chronic Beryllium Disease Prevention (Table I).<sup>(10)</sup> These criteria were selected as reference points because they are the only criteria available in the United States for beryllium surface contamination. The DOE criteria are housekeeping standards to determine cleaning effectiveness for work surfaces where beryllium is present. These surface standards are not based on health effects. Differences in the frequencies of samples in each concentration category by location were analyzed by Chi-square analysis.

#### RESULTS

A total of 95 samples were collected from 12 munitions plant buildings, and 46 samples were collected from six reference sites. Beryllium analyses of all field blank samples were below the reporting limit, indicating no significant precontamination of the wipes with beryllium or contamination during handling and shipping. Beryllium was detected in 83 samples (87%) collected at the munitions plant and 33 samples (72%) collected at reference sites (Table II). Two munitions plant sample concentrations were above 3.0  $\mu$ g/100 cm<sup>2</sup>. None of the reference site concentrations exceeded 3.0  $\mu$ g/100 cm<sup>2</sup>. Twenty-six surface samples (27%) from the munitions plant exceeded 0.2  $\mu$ g/100 cm<sup>2</sup>, whereas only three samples (7%) from the reference sites exceeded this level. A greater proportion of the surface sample measurements from the munitions plant were above the LOQ and greater than 0.2  $\mu$ g/100 cm<sup>2</sup> than from the reference sites (Chi-square = 11.1, p = 0.011).

The highest surface concentrations from the munitions plant were all associated with surfaces at or near sanders and grinders in maintenance shops where beryllium alloy tools reportedly were dressed (Table II). The two surface concentrations exceeding  $3.0 \,\mu g/100 \,\mathrm{cm}^2$  were collected from the room of the main machine shop building where the beryllium alloy tools were usually brought to be reshaped by machinists and tool and die workers. These two samples were collected from horizontal surfaces over 250 cm above the floor and along exhaust ventilation ductwork within 6.5 m of a large belt sander reported as the primary machine used to dress tools. Three other surface concentrations exceeding  $1.0 \,\mu g/100 \,\mathrm{cm}^2$  were collected from small maintenance shops serving production lines, on or very near the surface of flooror bench-mounted grinders in these shops. The average height of all samples collected in the munitions plant was 216 cm. The average height of the surface samples collected near the machines used to dress the beryllium-containing tools was 222 cm, while the height of the machines was about 105 cm.

The areas with the greatest proportion of samples that exceeded 0.2  $\mu$ g/100 cm<sup>2</sup> were the main machine shop (57%) and the production line maintenance shops (45%), while only 15% and 18% of the samples collected on the production lines and in the change house and laundry, respectively, exceeded 0.2  $\mu$ g/100 cm<sup>2</sup>. No samples from nonproduction or administrative areas exceeded 0.2  $\mu$ g/100 cm<sup>2</sup>. Chi-square analysis of sample frequencies in the concentration categories across munitions plant areas showed that the frequencies in the machine shop, line maintenance shops, and laundry and change areas were not significantly different from each other (the p-values ranged from 0.19 to 0.3). Sample frequencies from the production lines and laundry and change areas also were not significantly different from each other (p = 0.34); sample frequencies in nonproduction and administrative areas were significantly lower than in all other munitions plant areas

# TABLE II. Beryllium Surface Concentrations by Munitions Plant Locations and Reference Sites

			Above LOQ			
	No. Samples	< LOQ	$LOQ \le x \le 0.2$	$0.2 < \times \leq 3.0$	> 3.0	
Munitions Plant						
Main machine shop <sup>A</sup>	14	7	36	43	14	
Production line maintenance shops <sup>A</sup>	22	5	50	45		
Production lines <sup><math>B</math></sup>	39	5	80	15		
Change house and laundry <sup>A,B</sup>	11	18	64	18		
Nonproduction and administrative areas <sup>C</sup>	9	67	33	_		
All munitions plant samples <sup><math>D</math></sup>	95	13	60	25	2	
Reference Sites						
Precision machine-welding	5	100	_	_		
Sheet metal fabrication	4	75	25	_		
Automotive machining	10	20	60	20		
Custom automotive fabrication	4	75	25	_		
Paper processing	16	_	94	6		
Automotive machining	7	_	100	_		
All reference site samples <sup><math>D</math></sup>	46	28	65	7	0	

Percentage of Samples in Concentration Categories ( $\mu$ g/cm<sup>2</sup>)

 $^{A,B,C}$  Munitions plant areas with significant (p > 0.05) frequency differences in their sample concentration categories according to Chi-square analysis.  $^{D}$  Chi-square test comparing sample concentration category frequencies between all munitions plant samples and all reference plant samples = 11.1; p = 0.011.

(p < 0.05). Sample frequencies in all nonmaintenance shop areas were similar to those of the reference sites (p = 0.157).

Three samples from the reference plants were found to exceed 0.2  $\mu$ g/100 cm<sup>2</sup>. Two of these samples were collected from an automotive machining company, and one of the samples was from a paper processing company. Beryllium and beryllium alloys were not known to be used in these plants; however, beryllium alloys are present in some automotive parts and machining, and manipulation of these parts might be the source of the surface beryllium concentrations.

This data was used in conjunction with historical job descriptions and interviews with current and former workers regarding job tasks and locations to assign all munitions plant jobs into one of the three beryllium exposure categories (Table III). Tool and die workers and machinists who worked in the main machine shop, and who were assigned the responsibility of grinding and sanding beryllium alloy tools on an ongoing basis, were assigned to Category 2: Airborne or skin exposures may have occurred that involved direct contact or work with beryllium alloy tools, or close proximity to areas where these tools were reshaped. Production line workers, explosives operators, component operators, inspectors, quality control, plant maintenance, construction, and craft and trade workers who typically worked or spent time in the production areas where beryllium alloy tools were used were assigned to Category 1: Airborne or skin exposures may have occurred as a result of bystander exposure, not as a result of dressing beryllium alloy tools. Laundry and change house workers-who did not work in the production areas or visit any of the machine shops-were also assigned to *Category 1* because they handled all workers' clothing, and 18% of the surface samples in these areas had concentrations above  $0.2 \ \mu g/100 \ \text{cm}^2$ . Workers in all other jobs were assigned to exposure *Category 0: Airborne and skin exposures were not likely. Category 0* included administration positions, data and computer operators, grounds workers, medical staff, engineers, and security workers.

#### DISCUSSION

A pproximately half the beryllium surface concentrations in the main maintenance shop and production line shops exceeded  $0.2 \mu g/100 \text{ cm}^2$  with greater frequency than what might be found in other industries where beryllium or berylliumcontaining alloys were not known to be used. This data suggests that workers in these areas, particularly associated with sanders and grinders where beryllium-containing tools were dressed, could have encountered beryllium concentrations above what might be found in other working environments. The results also suggest that workers in the production areas and the laundry and change house occasionally may have been exposed to beryllium concentrations. Workers in other areas of the plant probably were not exposed to beryllium at levels any greater than the general population of industrial workers.

Sampling results from the reference sites indicate that settled airborne beryllium particles may not be uncommon in general industry, particularly where metal work is conducted. Beryllium may have been present in many items used at these sites. Beryllium and beryllium alloys are used in a variety of

# TABLE III. Beryllium Exposure Categories

Category	ry Description					
2	Airborne or skin exposures may have occurred that involved direct contact or work with beryllium alloy tools, or close proximity to areas where these tools were reshaped.					
	Workers assigned primarily to (or spent most of time in) main machine shop where beryllium alloy tools were designated to be reshaped					
	Examples:					
	machinists					
	tool and die makers					
	tool inspector					
	template repair					
	millwrights					
1		have occurred in areas where beryllium alloy tools were used, but as a result of result of dressing beryllium alloy tools.				
		Workers who worked primarily in areas where beryllium alloy tools were used; Workers who were likely to use				
	beryllium alloy tools in their tasks; Workers who regularly spent time on production lines in support activities;					
	Workers who did not work on production lines but handled contaminated clothing of those who worked in					
	maintenance shops or production areas					
	Examples:					
	explosives operators	electricians				
	components operators	melt operators				
	production workers	fire inspectors				
	change house attendants	plumbers and pipefitters				
	laundry workers	ironworkers				
	inspectors	maintenance workers				
	custodians	electricians				
0	Airborne and skin exposures were not likely.					
	Examples:					
	administration and administrative support					
	data entry and computer operators					
	engineers	scientists				
	grounds workers	security workers				
	medical staff	X-ray technicians				
	inventory checkers					

products, including automotive parts, precision instruments, computers, and electrical equipment.<sup>(15)</sup> These measurements indicate that beryllium may be present in manufacturing facilities but do not indicate the potential airborne concentrations.

The results of the surface sampling at the munitions plant show that beryllium concentrations on some surfaces were above hygiene levels used by DOE facilities. One possible conclusion is that the higher surface levels in the machine shop areas reflect the past grinding of beryllium alloy tools and potentially higher inhalational exposures to beryllium. However, the major limitation of these surface samples is that they cannot be used to directly estimate the inhalational exposure levels workers would have encountered. The higher surface concentrations in certain areas may reflect the deposition of particles with large aerodynamic diameters over unknown periods of time and, thereby, have little relevance to inhalational exposures. But the authors believe that the results indicate that, historically, machinists and tool and die workers who reshaped these beryllium-containing tools had the greatest potential for inhalation and skin beryllium exposures at the munitions plant.

Fewer than 100 surface wipe samples were collected throughout the munitions plant. This is a modest number of samples and may not fully characterize the range of surface concentrations present. Locations were selected following several walk-through tours to represent a broad range of potential for beryllium exposure: activities ranged from dressing beryllium alloy tools (main machine shop and small production line maintenance shops) to several production lines, service areas (laundry, change house), and areas where no production activities or beryllium alloy tool use had ever occurred. Current and past workers and plant administration personnel helped identify the areas where beryllium-containing tools were used and dressed.

Additional sampling would need to be conducted in the many buildings and work areas that were not sampled to fully characterize this large industrial site. However, the measurements collected suggest that with the exception of shop areas, surface beryllium concentrations likely would be below the DOE surface contamination housekeeping level of  $3 \mu g/100 \text{ cm}^2$ . Surface wipe samples are useful for monitoring potential contamination and the effectiveness of cleaning efforts, but the relationship between surface concentrations, inhalation exposures, and disease risk is not known, and no health-based standards for beryllium surface concentrations have been established.

One of the many limitations of surface sampling is that it collects particles of any size, including large particles that are unlikely to be inhaled. However, the average height of the surface samples collected near the machines used to dress the beryllium-containing tools was 222 cm, while the height of the machines was about 105 cm. Large berylliumcontaining particles would have largely been discharged from the grinders down and away from the elevated surfaces that were sampled. The large vertical belt sander, which was the primary machine used to reshape tools in the main machine shop, was equipped with local exhaust to collect particles.

Particles with an aerodynamic diameter of 100  $\mu$ m released at 105 cm would have a terminal settling velocity of approximately 25 cm/sec.<sup>(16)</sup> In stirred settling, less than 1% of these particles would remain suspended in air after 20 sec. Therefore, it is safe to assume that most of the beryllium collected on these elevated surfaces was from particles less than 100  $\mu$ m in diameter. According to the American Conference of Governmental Industrial Hygienists (ACGIH<sup>®</sup>), particles with aerodynamic diameters of 100  $\mu$ m are considered to be 50% inhalable.<sup>(17)</sup>

Surface contamination generally is not distributed evenly and can vary widely across surfaces. Therefore, sampling a small area may not accurately represent the contaminant deposition at a selected work site. Surface samples also collect particles that may have been deposited over the course of many years. The debris accumulated on these surfaces likely represents particle deposition over many years and may not be used to estimate current work exposures but could serve as an index of historical exposures. There may also be some variance in the pressure applied from sample to sample and between sample collectors, which could result in the collection of greater or lesser amounts of debris on some samples than others.

This study was not able to confirm that the source of the beryllium surface concentrations at the munitions plant was the beryllium alloy nonsparking tools. It is possible that using and machining other materials that contained trace or unknown beryllium amounts led to the accumulation of surface beryllium particles over long periods of time.

The risk of employees to become sensitized to beryllium through their work at the munitions plant is not known. However, it has been documented that workers exposed to very low concentrations of airborne beryllium for even short periods of time became sensitized and developed CBD.<sup>(18–22)</sup>

The sampling results suggest a range of potential historical beryllium exposure via inhalation and skin exposures, with highest potential exposure among those workers who would have dressed tools.

The sample results were used for categorizing jobs and, thereby, munitions plant workers into exposure strata for assessing the prevalence of beryllium sensitization. A sample of workers from each exposure strata were selected to participate in a beryllium sensitization prevalence study. Little information currently exists on the beryllium sensitization of workers in job categories with no expected beryllium exposure or exposure similar to that of workers in general industry. The findings of this study, in addition to the findings of a future prevalence study of beryllium sensitization among plant workers, may have important health implications for other munitions facilities and work sites where beryllium alloy tools were sanded or ground.

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