

Aerosols Generated During Beryllium Machining

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Some beryllium processes, especially machining, are associated with an increased risk of beryllium sensitization and disease. Little is known about exposure characteristics contributing to risk, such as particle size. This study examined the characteristics of beryllium machining exposures under actual working conditions. Stationary samples, using eight-stage Lovelace Multijet Cascade Impactors, were taken at the process point of operation and at the closest point that the worker would routinely approach. Paired samples were collected at the operator's breathing zone by using a Marple Personal Cascade Impactor and a 35-mm closed-faced cassette. More than 50% of the beryllium machining particles in the breathing zone were less than 10 μm in aerodynamic diameter. This small particle size may result in beryllium deposition into the deepest portion of the lung and may explain elevated rates of sensitization among beryllium machinists.

In late 1949, the US Atomic Energy Commission recommended the current beryllium occupational exposure levels for use by Atomic Energy Commission contractors.¹ The adoption and use of these levels reduced the occurrence of acute beryllium disease and initially appeared to reduce the occurrence of chronic beryllium disease (CBD).^{1,2} Some researchers suggested that the few reported cases of CBD observed during the 1960s and 1970s were likely related to higher previous exposures or to exposure excursions beyond the recommended levels,^{3,4} although low-level exposures had been linked to chronic disease in other investigations.^{5,6}

Beginning in the 1980s, researchers began to report CBD in individuals with low average exposures. Cullen et al⁷ reported five cases of CBD in a precious metal refinery, four of which developed in an area where exposures were believed to be consistently below the 2- $\mu\text{g}/\text{m}^3$ permissible exposure limit according to the Occupational Safety and Health Administration (OSHA). Kreiss et al⁸ also reported beryllium sensitization and CBD in a modern beryllium ceramics facility where median measured beryllium exposures were below the standard.

In the 1980s, the refinement and workplace application of a beryllium-specific lymphocyte proliferation blood test facilitated the identification of individuals sensitized to beryllium and the early diagnosis of CBD.⁹ Multiple epidemiologic investigations using this medical surveillance tool have revealed that both

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sensitization and disease continue to occur. In a study of nuclear weapons plant beryllium machinists, we observed a sensitization rate of 11.8% (6 of 51) with 7.8% (4 of 51) having documented CBD.¹⁰ Further study at the same workplace of another 895 workers revealed a plant-wide beryllium sensitization rate of 2.0% among workers with beryllium exposure and with 12 of the 18 having a diagnosis of CBD.¹¹ A beryllium sensitization rate of 9.4% was reported in a group of 646 workers studied at a beryllium manufacturing plant¹² as well as a rate of 3.5% among 227 self-reported beryllium exposed individuals working with beryllium ceramics.¹³ In each epidemiologic study, although high exposure work tasks were associated with elevated disease risk, cases of sensitization and disease were also detected among individuals with seemingly trivial bystander exposures, such as security guards, managers, secretaries, and spouses.¹⁴ In addition, exposure to beryllium copper mixtures with low beryllium percentages have been shown to result in CBD.^{12, 15, 16} These studies show consistently that beryllium sensitization and CBD continue to occur in industry even when measured exposures are believed to be below the current OSHA standard. The incorrect conclusion that CBD was disappearing under the Atomic Energy Commission standard^{1, 2} may have been contributed to, in part, by missed diagnoses, decline in reporting of new cases to the US Beryllium Disease Case Registry, and the long latency between exposure and observable disease in some individuals.

Researchers have found that the rates of beryllium sensitization and CBD do not seem to be evenly distributed among the employees exposed to beryllium. Although some studies have linked duration and amount of beryllium exposure to sensitization or disease,¹³ other researchers have suggested that sensitization and disease rates may be influenced by other factors such as

chemical form or particle size. Kreiss et al¹² conducted a study at a beryllium manufacturing facility and reported higher than expected rates of CBD in individuals working with beryllium-containing ceramics and among individuals producing beryllium metal. Other studies have found that employees involved in machining beryllium have higher sensitization and disease rates.^{8, 11} In a recent study from Japan, beryllium sensitization was associated with exposures as low as 0.01 $\mu\text{g}/\text{m}^3$.¹⁶

Cullen et al⁷ found that workers exposed to beryllium fume in high temperature beryllium operations had higher rates of CBD than did workers exposed to higher concentrations of beryllium in other areas of the facilities. Fume aerosols could differ in both particle size and chemical form compared with aerosols produced from mechanical processes. For example, beryllium oxide that has been fired at or below 500°C is reported to be more toxic than beryllium oxide fired at higher temperatures.¹⁷ Other researchers have suggested that the solubility and beryllium oxide layer thickness may alter the toxicity of a beryllium aerosol.¹⁸

In addition to influencing the total amount of beryllium that will be deposited in the respiratory tract, the aerodynamic size of the particles in a beryllium aerosol may also affect the site of deposition of the particles and thus alter the ability of the particles to cause sensitization and disease.^{19, 20} If the aerodynamic size of an insoluble aerosol is such that a portion of the aerosol is deposited below the ciliated region of the conducting airways, then that portion of the aerosol will clear more slowly than larger particles deposited in the upper, ciliated airways.²⁰ Insoluble aerosols deposited in the nasopharyngeal or the tracheobronchial region of the lung may be cleared much more rapidly than particles deposited in the distal airways and alveoli.²⁰ Slower clearance rates may result in greater beryllium retention and greater opportunity for contact be-

tween beryllium and cells of the immune system, potentially resulting in an increased sensitivity and disease rate. Unfortunately, there are few published data regarding the particle size and other physiochemical properties of beryllium as generated by beryllium machining in industry. Currently, the American Conference of Governmental Industrial Hygienists and other researchers are reexamining the Threshold Limit Values of many aerosols with the objective of determining the particle size most linked to the observed health effects and the concentration within that size range that should represent the threshold limit values.²⁰⁻²²

The purpose of this project was to determine the aerodynamic particle sizes of aerosols generated by specific beryllium machining operations as well as the aerodynamic particle size of the aerosols to which the workers involved with those machining operations were exposed. We hypothesized that typical beryllium machining operations generate a substantial amount of respirable-size beryllium particulate. The investigation was conducted at a precision beryllium machining facility that uses a number of different machining processes as well as several different beryllium materials to produce a wide range of beryllium components for both industry and government. The facility has been in operation since 1969, and, to date, 8.5% of the workforce has developed beryllium sensitization or disease.²³ Cases of beryllium sensitization and CBD continued to occur during the time that our sample efforts were performed.

Methods

Sample Collection

A large number of industrial processes are used at this precision machining facility, including receipt and inspection of materials, mechanical machining and polishing, electrical discharge machining, acid etching, and final inspection and quality

assurance. We limited our study to five mechanical processes typically used to machine beryllium: milling, deburring, lapping, lathe operations, and grinding. The facility also uses a number of different beryllium materials, including beryllium metal, beryllium-aluminum alloy (alubemet), and a beryllium metal/beryllium oxide matrix called E-metal.

We were interested in both the aerosol produced by a specific process and the characteristics of that aerosol as it reached the breathing zone of the worker directly involved with the process. For that reason, four different types of samples were obtained at each process. Two of the samples were stationary samples mounted to the machine and designed to characterize the aerosol generated by that process. Both of these samples were taken using Lovelace Multijet Cascade Impactors (LMJ) (InTox Products, Albuquerque, NM) sampling at a rate of approximately 24 L per minute for 4 to 8 hours using an electric vacuum pump. The aerodynamic cut points on the eight stages of the LMJs were 10 μm for stage 1, 6.3 μm for stage 2, 4.0 μm for stage 3, 2.5 μm for stage 4, 1.6 μm for stage 5, 1.0 μm for stage 6, and 0.6 μm for stage 7, with all particles smaller than stage 7 being collected on the final filter for stage 8. For comparison purposes, stages 2, 3, 4, and 5 were combined to obtain results comparable with the five stages used in the personal cascade impactors described below. The impactor collection substrates and the final filter were mixed cellulose ester filters (Type AW-19, Millipore Corp, Bedford, MA). One of the LMJ samplers was mounted within a few inches of the point of operation of the process and is referred to hereafter as the point of operation (POO) sample. This sampler was designed to investigate the actual aerosol generated by the machining process. The second fixed sample was called the nearest worker location (NWL) sample and was mounted at a location that was rep-

resentative of the closest point at which the worker would routinely approach the tool and beryllium piece for observation or adjustment of the machining operation. This point was not the closest point at which the worker *could* approach, because some activities (such as tolerance measurements) might require very close approach distances for short periods of time, but it was closer than where workers were located routinely.

Two additional sets of samples were obtained in the breathing zone of the worker operating these machines by using lapel samplers powered by personal sampling pumps. The first sampler was referred to as the total beryllium (TB) sampler and consisted of a pre-loaded, closed-face, 37-mm cassette with a 0.8- μm pore-sized multiple cellulose ester filter (MAWP037AO, Millipore Corp, Bedford, MS). This sampling method is the personal sampling method currently recommended for beryllium in the current National Institute for Occupational Safety and Health (NIOSH) guidelines.²⁴ The TB sampling results provide a determination of the TB present in the breathing zone of the worker during the sampling period with little regard to the size of the particulate. This sample was collected by using a calibrated personal sampling pump sampling at a rate of approximately 2 L per minute.

The second personal sampler, a Series 290 Marple Personal Cascade Impactor (Graseby-Anderson, Atlanta, GA), was also attached to the lapel of the workers. The five stages used were designed to give cut points of 10 μm for stage 1, 6.0 μm for stage 2, 1.0 μm for stage 3, 0.6 μm for stage 4, and all particles less than 0.6 μm for stage 5. The collection stages were loaded with slotted cellulose ester filters designed for use with the personal cascade impactor and provided by Graseby Anderson. The final filter of the personal impactor (PI) was also a cellulose filter provided by Graseby-Anderson. The

samples were collected by using calibrated personal sampling pumps sampling at an approximate flow rate of 2.0 L per minute. Samples collected in this manner are referred to as PI samples.

Ambient air samples were taken from four fixed locations in the facility: three locations within the work area and one in the administrative area. The air samples were obtained over a 48-hour period by using 37-mm cassettes with cellulose ester filters, PIs, and the Lovelace Multijet Impactors. One of each type of sampler was used at each location for a total of three samples per location.

Both the LMJ impactors and the Marple Personal Impactors were loaded in a laboratory hood within the precision machining facility. The impactors were cleaned in a soap and water solution, dried, and loaded. Information provided by Graseby-Anderson indicated that particle bounce would not normally be a problem when using the cellulose ester media in the Marple Personal Cascade Impactors. In addition, work conducted by Lovelace Respiratory Research Institute with the LMJ samplers using cellulose ester filters suggested that filter greasing might not be necessary. However, to verify that there were no differences in the results between greased and ungreased filters, we collected some samples side-by-side by using the Marple Personal Impactors and Lovelace Multijet Impactors with and without a silicone grease (Dow 316 Silicone Spray, Dow Chemical Co, Midland, MI).

The fixed, LMJ samples at the POO and NWL were typically collected over an 8-hour shift. Because of the lower sampling rates of the personal samplers and the generally lower concentrations of beryllium to which they were exposed, the TB and PI samples were collected over two shifts. After sampling, the cascade impactors were returned to a laboratory located within the plant and unloaded in the laboratory hood.

The impactor collection substrates were packaged in petri dishes, sealed with tape, and shipped for analysis as a group. Filters from the cassette samples were removed from the cassette, packaged in petri dishes, sealed, and shipped with the other samples for analysis. All of the samples were shipped to an American Industrial Hygiene Association accredited laboratory for analysis, along with both blank and beryllium-spiked samples.

Sample Analysis

For the majority of the samples, the method of analysis was NIOSH Method 7300 using a Fisons Accuris Radial ICP Spectrometer.²⁴ This method had a limit of detection (LOD) of 0.007 µg/filter and a limit of quantification (LOQ) of 0.030 µg/filter. Some of the samples were analyzed by using NIOSH Method 7102 using a graphite furnace atomic absorption spectrometer.²⁴ This method had a LOD of 0.0003 µg/filter and a LOQ of 0.005 µg/filter. In both cases, the results used in the study were the value assigned by the laboratory, even if the reported results were less than the LOQ. Results below the LOD were assigned a value of 0.0035 µg/filter (1/2 the NIOSH 7300 Method LOD) for purposes of statistical analysis. Laboratory results are reported as a specific value down to the limit of detection for the method used.

PI sample data were analyzed in several ways. The percentage of beryllium mass for each stage was calculated to determine a median percent beryllium per stage. In addition, the time-weighted average (TWA) for each sample was calculated by adding the total mass from all of the stages together and dividing the result by the cubic meters of air sampled. The theoretical number of particles per stage was calculated, assuming the particles to be spherical in shape, by determining the characteristic physical diameter of the particle by dividing the aerodynamic characteristic diameter for that stage

(the geometric mean of the upper- and lower-stage diameters for 50% collection efficiency) by the square root of the particle density (considered to be 2.0 owing to the presence of beryllium oxide¹⁸). Appropriate treatment of the Cunningham slip correction was included for smaller particles. We calculated the characteristic mass of a particle on an impactor stage by using the following formula for particle volume times density:

$$(3.14 \times (\text{characteristic physical particle diameter})^3 / 6) \times \text{density of 2.0} = \text{characteristic particle mass.}$$

The resultant particle mass was divided into the total mass for that stage to determine a theoretical particle number for the sample collected on each stage. The theoretical surface area for the sample collected on each stage was then calculated by using the following formula:

$$3.14 \times (\text{particle diameter})^2 \times \text{particle number} = \text{theoretical surface area.}$$

We calculated the mass median aerodynamic diameter (MMAD) for each cascade impactor sample by plotting the cumulative percents for each stage on a log probability plot.²⁵ In a significant number of samples, more than 50% of the mass was found on either the first stage or the last stage of the impactor. In those instances, the MMAD was assigned a value 10 if the MMAD was greater than 10, or a value of 0.1 if the MMAD was less than 0.1.

Deposition rates in the human respiratory tract for the beryllium particulate were estimated by using the characteristic aerodynamic diameter for each impactor stage and then applying the information obtained in the LUDEP 2.0 program developed by the International Commission on Radiological Protection (ICRP). This program is based on the data presented in ICRP publication 66.²⁶ The model that we used assumes an oc-

cupational exposure with adult male physiology, light work, 100% nose breathing, and a monodisperse particle size equal to that of the impactor stage of interest and a particle with a density of 2.0. The value for total deposition was the percentage of the median percentage for each stage that would be deposited somewhere in the respiratory tract. The amount deposited in the lung was defined to be the percentage of the median percentage per stage that would be deposited from the trachea down to the alveoli. The alveolar deposition rate included the percentage deposited in the alveolar and bronchiolar regions of the lung.

Statistical Methods

One-way repeated measures of analysis of variance (ANOVA) models were fit to look for differences between process groups and sampler types. Although there were three factors (metal, sampler, and operation) in these analyses, interactions among operations, metals, and sampler types could not be evaluated. Variables that were not normally distributed were log-transformed for these analyses. Least squares means and their standard errors were calculated, and significant differences between them were determined by using Tukey's Honestly Significant Difference procedure. Although least squares means were used for the comparative analysis, the data were summarized by using arithmetic means and medians for the tables presented in this article. The median is a more appropriate summary statistic for data that are not normally distributed or that have extreme outliers. Wilcoxon's signed-rank test was used to compare the greased and ungreased sample pairs. All statistical tests were two-sided and were conducted using a significance level of 5%.

Results

A total of 336 samples were obtained, including 79 TB samples, 87 PI samples, 71 NWL samples, 87

TABLE 1

Comparison of TWA* Beryllium Concentrations in $\mu\text{g}/\text{m}^3$ Obtained From the Different Types of Samples Collected at a Precision Machining Plant

Sample Type	<i>n</i>	Mean ($\mu\text{g}/\text{m}^3$)	Median ($\mu\text{g}/\text{m}^3$)	Minimum ($\mu\text{g}/\text{m}^3$)	Maximum ($\mu\text{g}/\text{m}^3$)
POO*	56	7.19	0.60	0.02	122.31
NWL	64	0.91	0.20	0.01	18.13
PI	59	1.51	0.37	0.03	22.68
TB	64	1.48	0.29	0.03	41.48

* TWA, time-weighted average; POO, point of operation; NWL, nearest worker location; PI, personal impactor; TB, total beryllium.

POO samples, and 12 ambient air samples. In addition, we submitted 130 blanks using all of the filter types as well as 8 spikes that contained approximately 70 μg of beryllium/filter. Twenty-five (19.2%) of the blanks were reported by the laboratory to contain amounts of beryllium above the LOD, ranging from 0.00073 $\mu\text{g}/\text{filter}$ to 9.7 $\mu\text{g}/\text{filter}$ with a mean of 0.660 $\mu\text{g}/\text{filter}$, a median of 0.011 $\mu\text{g}/\text{filter}$, and a standard deviation of 2.24. The values of the spikes ranged from 75 to 86 $\mu\text{g}/\text{filter}$ (mean 80.28), with the exception of one outlier (0.93 $\mu\text{g}/\text{filter}$).

Of the total 336 samples, 243 were samples of the target processes. The 243 samples consisted of 64 TB samples, 59 PI samples, 64 NWL samples, and 56 POO samples. Descriptive statistics for the time-weighted average results obtained from the specific sampler types are presented in Table 1. The median TWA sample results ranged from a low of 0.20 $\mu\text{g}/\text{m}^3$ for the NWL samples to 0.60 $\mu\text{g}/\text{m}^3$ for the POO samples. The TWA results obtained at the POO were significantly higher than the results obtained at the NWL, PI, or TB locations ($P < 0.05$, from one-way ANOVA). The TWA beryllium levels found in the breathing zone by using the TB and PI sampling locations were not significantly different ($P > 0.05$, from one-way ANOVA). Contrary to our expectation, NWL sample results were significantly lower than the results found at the breathing zone ($P < 0.05$, from one-way ANOVA). This suggests that either the worker routinely ap-

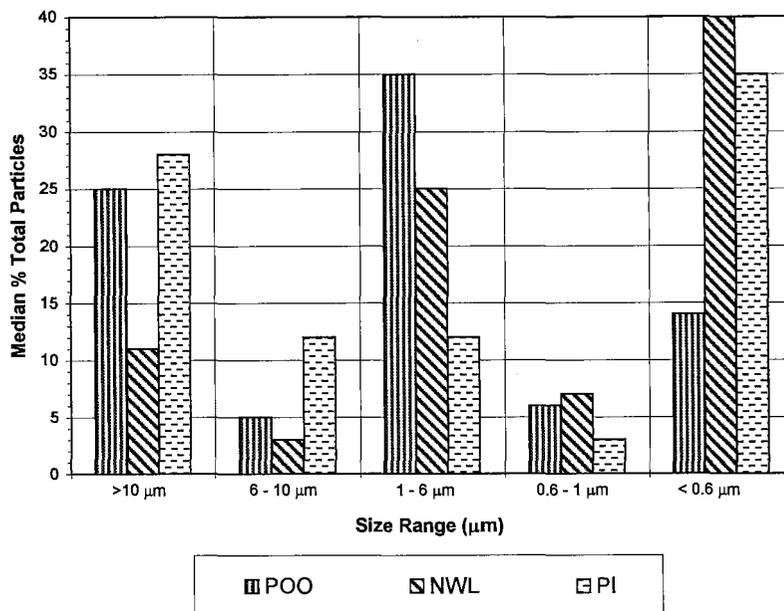


Fig. 1. Median percent of beryllium by sampling method for the five particle size ranges sampled.

proached the process closer than the location of the NWL or that the NWL location was not entirely in the plume of the aerosol in all cases. This difference highlights the reason that breathing zone samples must be taken to accurately predict actual worker exposures.

Figure 1 illustrates the median percent of beryllium collected in the five different particle size ranges sampled. The percentage of particles in the smallest size range (stage 5, $<0.6 \mu\text{m}$) was similar for both the NWL and the PI samples, with median percentages of 40% and 35%, respectively. Particles produced at the POO were generally of a larger size than the particles that reached the worker's breathing zone, with only 14% of particles at POO found

at stage 5. The largest particles (those collected on stage 1) were greatest for the PI samples (median 28%) and POO samples (median 25%), compared with only 11% for the NWL samples.

Because of the surprising observation that more of the large particles were in the PI samples than in the POO samples, we wanted to determine whether this was a consequence of sampling bias, such as particle bounce off the ungreased filter substrates on stage 1 of the LMJ. If particle bounce had caused the difference, it would be expected that the lower stages of the ungreased PIs would have the most material present owing to particles bouncing down from the higher stages. The results of a paired sam-

TABLE 2
Comparison of TWA* Exposure Concentrations in $\mu\text{g}/\text{m}^3$ by Process Sampled for Each Sampler Type

Process	POO*		NWL		PI		TB	
	<i>n</i>	Median ($\mu\text{g}/\text{m}^3$)						
Deburring	16	0.58	18	0.26	15	0.74	17	0.57
Grinding	13	2.21	13	0.65	11	0.34	12	0.26
Lapping	10	0.32	10	0.11	12	0.13	13	0.10
Lathe operations	6	4.08	6	0.27	7	0.60	7	0.40
Milling	11	0.18	17	0.18	14	0.25	15	0.20

* For definitions of abbreviations, see Table 1.

pling ($n = 8$ pairs) using the PI samples showed no difference between the greased and ungreased PIs either in total mass of beryllium per stage or in total mass of beryllium collected. There was a significant difference ($P = 0.02$) between the percent of total mass on the fourth stage of the greased impactors (median 9%) versus the ungreased PIs (median 2%). The fourth stage of the PIs generally collect the lowest amount of mass, and the observed difference may be due to the low amounts of mass on that stage.

The greased LMJs, on the other hand, had a significantly greater amount of beryllium on the first three of the total eight stages as well as a higher TB content for the overall sample than did the ungreased LMJs ($n = 9$ pairs, $P = 0.004$). Stages 4 to 8 were not found to have significantly different amounts of beryllium present, suggesting that the ungreased LMJs lost some of the larger particles (probably to the internal surface of the impactor) that should have been collected on the first three stages. These particles did not seem to collect on the lower stages of the LMJ. Because of this anomaly, the first two of the five stages (represented in Fig. 1) may underrepresent the actual amount of beryllium present in the NWL and POO samples.

The results of the ambient air samples were similar for all of the sampling devices, with levels ranging from $0.01 \mu\text{g}/\text{m}^3$ to $1.20 \mu\text{g}/\text{m}^3$. The median values ranged from $0.02 \mu\text{g}/\text{m}^3$ in the administrative area to

$0.07 \mu\text{g}/\text{m}^3$ in the work areas. The majority of the particulate was also less than $0.6 \mu\text{m}$ in aerodynamic diameter, with median percentages of 38% to 83% of the particulate collected on the last stage of the impactors.

Table 2 shows the median TWA results of the five different types of processes sampled for the different samplers. Grinding and lathe processes produced the highest median exposures at the POO, but at distances removed from the POO there were minimal differences between the exposures generated by the five processes. The medians for the lapel samples, both PI and TB, were similar for all of the processes, although lathe and deburring exposures were slightly higher than lapping, grinding, and milling exposures. Lower median levels of exposure were detected for deburring at the NWL location than for the other samplers. Deburring is performed by using a hand-operated grinding tool that is moved around the part to remove rough edges and, therefore, the POO is continually moving. Thus, the true POO for deburring may have been closer to the breathing zone than to the POO or NWL sampling site.

The median percent of beryllium per size range for the PI samples obtained for each process is shown in Figure 2. We typically observed a bimodal distribution of particle sizes with greater than 20% of the particle mass in the $>10 \mu\text{m}$ size range and greater than 30% of the particle mass in the smallest size range ($<0.6 \mu\text{m}$).

This bimodal distribution was observed even when normalized data were plotted (delta mass/delta log (d)). The greatest median percent of particle mass present in the smallest size range occurred in lathe and milling operations (57% and 44%, respectively). Lathe and deburring processes produced the lowest percentage of large particles in the first stage, suggesting that these operations produce an aerosol of generally smaller particle sizes at the breathing zone.

Because of the bimodal distribution and the lack of a log normal distribution, the calculation and use of a MMAD is not justified in the strict sense. To facilitate comparison of MMADs with the MMADs presented by other authors, we calculated approximate MMADs, which are summarized descriptively for each process in Table 3. As expected, the POO samples had a higher median MMAD than did the NWL or PI samples for all of the process types. Aerosols at the POO ranged from $2.6 \mu\text{m}$ for lathing processes to $5.3 \mu\text{m}$ in diameter for milling processes. The results obtained from the NWL and PI samples were similar and ranged from a low of less than $0.6 \mu\text{m}$ to a high of $3.1 \mu\text{m}$. Lathe operations generated the lowest median MMAD ($0.6 \mu\text{m}$) measured from the PI samples. Grinding produced the highest MMAD ($3.1 \mu\text{m}$).

Table 4 shows median respiratory tract deposition rates for the specific processes. The total median respiratory tract deposition rate varied from

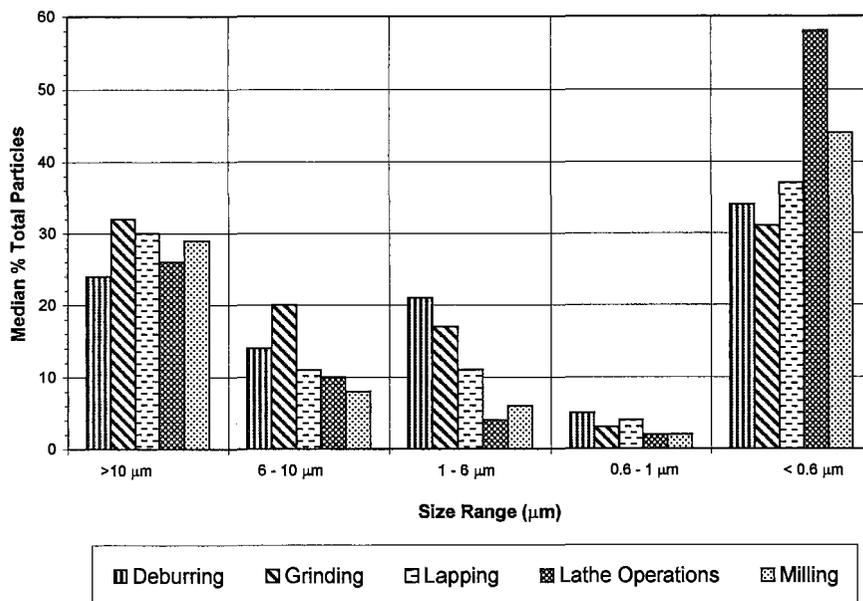


Fig. 2. Median percent of beryllium for PI samples by process for each size range.

42% for lathe operations to 59% for grinding operations. Lung and alveolar lung median deposition rates were similar in our model for all process types, ranging from 6% to 8%. This difference between total and lung deposition is due to the high deposition rates (29% to 34%) in the extrathoracic region of the respiratory tract for particles greater than 2.0 µm in aerodynamic diameter. Alveolar lung deposition rates are highest for the smaller diameter particles that are less than 2.0 µm in aerodynamic diameter, although the rate of deposition is much lower (1% to 13%).²⁶ These factors result in the similar lung and lower lung deposition rates observed for the different processes.

Table 5 presents the median numbers of particles theoretically generated per process for PI samples. The median particle numbers range from a low of 14×10^6 particles for ambient air to a high of 99×10^6 particles for deburring operations. The primary determinant of particle number is the median total mass obtained for that process. When we divided the total number of particles by the median mass per process to calculate the number of particles per µg of exposure, exposures produced

by lathe operations and in the ambient air tended to contain the highest levels of particles per unit mass.

The theoretical surface area of the beryllium particles is related directly to the particle numbers and, again, to the median mass for each of the processes. Median particle surface area per process is presented in Table 6 and ranges from 1.4×10^6 µm² for ambient air samples to 10.9×10^6 µm² for deburring operations. When we divided the particle surface area by the median mass per process, we observed that the ambient air and the lathe operations again had a higher surface area per µg of exposure.

Conclusions

Our review of the literature indicates that this is the first study designed to characterize the particle size of aerosols generated under normal beryllium machining conditions in a beryllium machining facility. It is also the first study employing personal cascade impactors to characterize the aerosol to which the beryllium operators are exposed. Our objective was to explore why machinists have higher CBD and beryllium sensitization rates than do most other beryllium industry workers. The results of the study show that

machining of beryllium liberates a high percentage of small diameter beryllium particles. Aerosols generated by machining processes were generally bimodal in distribution, with greater than 50% of the particulate less than 10 µm in aerodynamic diameter. This small particle size may result in high beryllium deposition rates and penetration into the deepest portion of the lung, where immune responses and the pathology of CBD develop.

All of the exposures monitored contained a high number of very small particles per microgram of exposure. The high number of particles may result in a higher surface area per microgram of exposure. Ambient air sample results within the facility had some of the highest numbers of particles per unit of mass, which may help explain why personnel exposed to the ambient air of beryllium manufacturing facilities have been found to be sensitized to beryllium or have developed CBD.

Before this plant-based study, Hoover et al²⁷ studied the particle size of aerosols generated during sawing and milling of beryllium under controlled conditions wherein the cuts were specified and repeated for the study. The machining was done dry, without cutting liquids, and all of the particle debris from the cutting operations was collected for particle size analysis. In a previous study, Hoover et al¹⁹ studied beryllium aerosols during a specific machining operation performing the final milling of beryllium metal, and during electron beam heating of a beryllium block using area cascade impactors. These data were collected during specific operations under controlled conditions. In neither case were personal cascade impactors used, and the studies generally characterized the aerosol only at the POO for the process monitored.

In both of the experimental studies, the researchers reported larger particle sizes for particle debris from the cutting process than we observed as an aerosol in this workplace study.

TABLE 3
Comparisons of Median MMAD* for the Processes Studied by Sampler Type

Process	POO		NWL		PI	
	<i>n</i>	Median (μm)	<i>n</i>	Median (μm)	<i>n</i>	Median (μm)
Deburring	16	3.2	18	1.2	15	1.6
Grinding	13	4.1	13	2.3	11	3.1
Lapping	10	2.6	10	1.2	12	2.3
Lathe operations	6	3.6	6	0.6	7	0.6
Milling	11	5.3	17	0.6	14	2.7

* MMAD, mass median aerodynamic diameters. For definitions of other abbreviations, see Table 1.

TABLE 4
Comparison of Median Percent and Site of Deposition for Beryllium Particles from Sampled Processes

Process	<i>n</i>	Total Deposition (%)	Lung Deposition (%)	Alveolar Deposition (%)
Deburring	15	54.4	8.3	6.9
Grinding	11	58.7	6.8	5.7
Lapping	12	50.8	7.1	5.9
Lathe operations	7	41.9	8.3	6.9
Milling	14	52.0	6.9	5.7

TABLE 5
Median Theoretical Numbers of Particles per Process Group for PI* Samples

Process Group	<i>n</i>	Median		
		Be Mass (μg)	Particle No./10 ⁶	Particle No./10 ⁶ /Be Mass (μg)
Deburring	15	1.42	99	69.5
Grinding	11	0.47	38	56.8
Lapping	12	0.31	20	80.9
Lathe	7	1.01	45	117.8
Milling	14	0.52	55	80.3
Ambient air	4	0.11	14	104.0

* PI, personal impactor; Be, beryllium.

The initial study characterizing the final machining of beryllium metal reported an aerosol with a MMAD of 3.5 μm.¹⁹ During the second study, Hoover et al²⁷ reported that the mass median diameter of all of the particle debris from the cutting operations ranged from 50 μm to 175 μm, depending on the depth of the milling cut. The results of our sampling were similar to the results from the first study in that, at the POO, the MMAD ranged from 2.6 μm to 5.3 μm, depending on the machining process sampled. The MMAD of the

aerosol present at the breathing zone of the operator, however, was substantially smaller and ranged from 0.6 to 3.1 μm. The MMADs observed at the POO for all of the processes were lower than the MMADs reported for the total particles that were produced and collected in Hoover et al's second study.²⁷ The reason for these differences may be that the size distribution is quite large for the particles that are removed by the machining processes, but that only the smaller size fraction is suspended as an aero-

sol. This reduction in the effective particle size of the dispersed material may be associated with the use of liquid cutting solutions in the present study. Other differences may be related to the cutting depths or the machining processes observed in the two studies, as well as the point at which the particulate was measured.

In general, the aerosols generated by the processes that we measured were very similar. Although samples obtained at the POO contained generally larger particulate, results obtained at the breathing zone of the operator were very similar between the different processes, with a substantial amount of the particulate observed on the last stage of the impactor (characteristic aerodynamic diameter of 0.24 μm). The lack of a large difference between the POO samples and the PI samples is most likely explained by two factors: the location of the sampler and variation within the process. In some instances, the beryllium concentration observed at the POO was less than at the PI sample. Assuming that the person operated that specific machine all day and that there was not a machine with a much higher exposure located nearby, the only explanation for this difference is that the POO sample was located outside of the plume generated by that machine. In one study, Hoover et al¹⁹ used an optical aerosol monitor to locate the plume generated by the machine that they sampled before setting up the sampling devices. In our study, the location was chosen by our best es-

TABLE 6
Median Theoretical Surface Area by Process Group for PI* Samples

Process Group	n	Median		
		Be Mass (µg)	Surface Area/10 ⁶ (µm ²)	Surface Area/10 ⁶ /Be Mass (µg)
Deburring	15	1.42	10.9	7.3
Grinding	11	0.47	4.0	6.1
Lapping	12	0.31	2.0	8.1
Lathe	7	1.01	4.7	11.4
Milling	14	0.52	5.6	8.0
Ambient air	4	0.11	1.4	10.6

* PI, personal impactor; Be, beryllium.

timate as to where the plume might be located as well as the ability to safely attach a sampler in that area. The safety factor was especially important because operations were being conducted as normal and a tool contacting a sampler could have resulted in injury to the operator. Samples taken using a real-time aerosol monitor provided to us by Los Alamos National Laboratory during our study suggested that the location of our sampler was not always optimal. The real-time analyzer results suggested, in fact, that the plume from some operations was very narrow and difficult to predict without the use of an optical particle counter or real-time analyzer. Thus, assessment of exposures in the breathing zone of the individual may provide more valid data and may be of great relevance for the machinist.

A second factor affecting the difference in particle size between the POO samples and the PI samples is the type of process monitored. In processes such as milling and grinding, the actual POO is easily pinpointed. In the case of deburring, for which a small grinding tool is used, the POO moves with the operator and may, at times, be closer to the operator than to the POO sampler. Other processes may consist of a number of operations, each of which may produce different particle sizes at different times during the sampling process. Particle size differences may be due to machine type, tool type, depth of cut, and type of beryllium metal or mixture used.

Exposures in the employee's breathing zone were very similar for all of the operations monitored and resulted in a large amount of small beryllium particles reaching the breathing zone of the employee. A median of 68% to 78% of the mass collected in the employee breathing zone was respirable (<10 µm in aerodynamic diameter) for all of the processes. This large number of small particles resulted in fairly uniform deposition rates for the processes monitored, especially in the air exchange areas of the lung, where beryllium deposition rates ranged from 7% to 9%. When total respiratory tract deposition was calculated by using the ICRP Model, deposition rates for the different processes ranged from 46% to 62%. Our data indicate, therefore, that the machining operations sampled during this study produce an aerosol that consists of small particles that are easily respirable and are deposited in large quantities in the respiratory tract, and in smaller quantities in the deepest portions of the lung.

During our study, median exposures measured at the breathing zone for all of the machining processes were less than the current OSHA permissible exposure limit or the American Conference of Governmental Industrial Hygienists Threshold Limit Value of 2 µg/m³. The median time-weighted PI sample results ranged from 0.13 µg/m³ for lapping processes to 0.74 µg/m³ for deburring operations, well below the current standards. Ambient air sam-

ples taken at several locations within the plant revealed 48-hour median concentrations of between 0.02 µg/m³ and 0.07 µg/m³. This ambient air aerosol was unique in that it was comprised of smaller particulate than the aerosol associated with the individual processes, with most of the aerosol collected in the lowest stage of the impactor (0.24-µm characteristic aerodynamic diameter). These results indicate that everyone within the plant was exposed to some degree to beryllium and that the background, ambient air exposure is to a fine particulate that achieves deep penetration into the lung.

Particle numbers and particle surface area may play important roles in the sensitization or disease process. Eisenbud² suggested that particle size, solubility, and solubility rates have been shown to have an effect on CBD rates. He also indicated that these physical factors may have been related to why community CBD rates in Ohio were higher than worker CBD rates even though exposures to workers were thought to be much higher. Hoover et al¹⁸ found that small particles form an outer oxide layer that results in characteristics similar to low fired beryllium oxide, which has been shown to have a greater toxicity than high fired beryllium oxides. The greater toxicity of the low fired beryllium oxide may be due to the solubility of the material following deposition in the pulmonary portion of the lung. Hoover et al¹⁸ suggested that the similar solubility of small particles coated with a layer of beryllium oxide may result in a similar toxicity. In addition, smaller particles may provide more sites for attack by the cellular immune system and result in a larger number of granulomas.

The calculations of theoretical particle numbers for the different processes presented in Table 5 indicate high particle numbers per microgram of exposure for all processes and especially for ambient air and lathe operations. This higher particulate count in ambient air may begin to

explain the cases of CBD found in individuals working in administrative and clerical positions in this and other plants, as well as the higher than expected number of neighborhood cases, when compared with worker cases, reported by Eisenbud.² Our study also indicates that lathe operations may produce a high number of very small particles per microgram of exposure and that lathe operations should receive extra control considerations. However, it should also be noted that all of the processes monitored produced large numbers of very small particles, which, if coupled with higher mass exposures, would certainly equal and exceed the numbers of small particles present in ambient air and milling operations. Our calculation of surface area was based on the number of particles and thus mirrored the results observed when looking at particle numbers, indicating that the two are linked and that processes resulting in high particle numbers also produce high surface area per microgram of exposure.

By virtue of the fact that this study was conducted in an actual working environment during normal operations, we were able to determine working aerosol characteristics for the processes monitored. Although this is advantageous in determining "real world" exposures, it also results in a number of problems that introduce limitations to the interpretation of the results of the study. Although every effort was made to look only at the exposure produced by a specific operation, the breathing zone samples (PI and TB samples) followed the worker and thus may actually represent exposure from more than one process. In addition, although an effort was made to monitor an individual who worked only with one type of beryllium metal, the use of more than one metal type per monitoring period was possible. It was also not possible to control for process factors such as specific cuts, dimensions, and tool size, although each has a potential effect on the aerosol characteristics of the process

in question. Our sampling effort reflects an average of the aerosol produced by the process monitored under varying conditions. These conditions may or may not be replicated in other beryllium industries, although instrumentation and machining processes are relatively consistent across the industry. Future research will be needed to validate our observations at other beryllium plants.

In addition to variation in the processes, there also may be variation in the ventilation control devices, control device location, and individual machine characteristics for the processes sampled. Individual operators adjusted their own beryllium control devices during the project, and the devices may not have been positioned as effectively for each process sampled. In addition, the results from different machines were grouped into specific process types. For examples, the results from a Bostomatic milling machine sampled as a milling process on one day were combined with the results from a horizontal bore mill taken at another time period. Thus the observations concerning milling operations represent an aggregate value, serving as an estimate for a number of different milling operations. This grouping limits the conclusions that can be drawn concerning specific types of milling. The results also suggest the need for close monitoring and control of machining aerosols involving beryllium metal.

In conclusion, beryllium machining as performed in industry today produces large numbers of fine respirable size particles. This may help explain the persistence of CBD occurring even at seemingly low levels of beryllium exposure. Exposure control efforts in the beryllium industry should focus on mitigating exposure to respirable beryllium particulates in the personal breathing zone of the worker. Future research should examine the relationship between machined particle size, mass,

and physiochemical properties and the incidence of CBD in industry.

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ORIGINAL ARTICLES

Mercury Toxicity due to Use of a Cosmetic Cream 4
Cheryl McRill, MD, MPH, Leslie V. Boyer, MD, Timothy J. Flood, MD,
and Luis Ortega, MD, MPH

Aerosols Generated During Beryllium Machining 8
John W. Martyny, PhD, Mark D. Hoover, PhD, Margaret M. Mroz,
MSPH, Kimberly Ellis, MS, Lisa A. Maier, MD, Karen L. Sheff, MS, and
Lee S. Newman, MD

**Circulating Natural Killer Cells in Retired Asbestos Cement
Workers** 19
Paul Froom, MD, MOccH, Nitzah Lahat, PhD, Estela Kristal-Boneh, PhD,
Chaim Cohen MD, MOccH, Yehuda Lerman MD, MPH, and Joseph Ribak
MD, MPH

**Why Most Workers With Occupational Repetitive Trauma do not
File for Workers' Compensation** 25
K. D. Rosenman, MD, J. C. Gardiner, PhD, J. Wang, MS, J. Biddle, PhD,
A. Hogan, PhD, M. J. Reilly, MS, K. Roberts, PhD, and E. Welch, JD

**Effectiveness of Early Physical Therapy in the Treatment of
Acute Low Back Musculoskeletal Disorders** 35
Gary C. Zigenfus, MSPT, Jiahong Yin, BS, PhD, Geneva M. Giang,
MBA, and W. Tom Fogarty, MD

**Psychosocial Factors At Work, Smoking, Sedentary Behavior,
and Body Mass Index: A Prevalence Study Among 6995 White
Collar Workers** 40
Chantal Brisson PhD, Brigitte Larocque MA, Jocelyne Moisan PhD,
Michel Vézina, MD, MPH, FRCP, and Gilles R. Dagenais, MD, FRCP

**Worksite Cardiovascular Wellness Programs As a Route to
Substance Abuse Prevention** 47
Max Heirich, PhD, and Cynthia J. Sieck, MPH

**Hearing Loss Among Construction Workers in Edmonton,
Alberta, Canada** 57
Patrick A. Hessel, PhD