

Airborne Release Fractions and Health Effects for Beryllium During Facility Accidents

J.C. Laul et al
Los Alamos National Laboratory

March 2006

jclaul@lanl.gov

LA-UR-06-1755

Beryllium Metal Bounding Airborne Release Fractions and Respirable Fractions for DOE Facility Accidents Analyses

Jofu Mishima, Los Alamos National Laboratory, consultant

Terry L. Foppe, Foppe and Associates

J.C. Laul, Los Alamos National Laboratory

Patrice M. McEahern, CALIBRE

David M. Pinkston, Lawrence Livermore National Laboratory

Louis F. Restrepo, Omicron Safety and Risk Analysis

LA-UR-05-1096, April 2005

Presentation Topics

- **Introduction and Purpose**
- **Beryllium (Be) Characteristics**
- **Oxidation of Be Metal < Ignition Temperature**
- **Ignition of Be Metal**
- **Reported Accidental Releases of Airborne Be Oxide**
- **Applicability of DOE-HDBK-3010 Values to Particulate Toxic Material Releases**
- **Summary of ARF & RF Values for Be Metal**
- **High, Moderate and Low Hazard Facility**
- **Health Effects of Beryllium (<10 um & >10 um)**
- **Sensitization and CBD Cases**

Introduction and Purpose

- **Be has special properties for use with nuclear materials**
- **Metal is a misnomer – “blue oxide” coating at room temperature**
 - **Rapidly re-established if disturbed**
- **Inhalation hazard from airborne releases**
 - **Table 1 shows ERPGs/TEELs**
- **Purpose: review literature and recommend ARFs and RFs for Be metal and its oxide**
- **Assess health effects of <10 um on short and long term basis**

Table 1

ERPGs/TEELs Values for Beryllium and its Compounds

Compound	ERPG-1 (mg/m ³)	ERPG-2 (mg/m ³)	ERPG-3 (mg/m ³)
Beryllium metal, Be	0.005	0.025	0.1
Beryllium hydroxide, Be(OH) ₃	0.025	0.25	20
Beryllium oxide, BeO	0.0125	1.25	10*

*** On Oxidation, Be is converted to BeO, which is 100 times less hazard than Be metal**

Beryllium Characteristics

- Physical characteristics of Be metal:
 - Formula weight: 9.02
 - Metal density (?): 1.85 g/cm³
 - Oxide density (?): 3.05 g/cm³
 - Melting point: 1,278 °C
 - Boiling point: 2,467 °C
 - 2% to 15% ARF from detonations
- Chemical characteristics:
 - Be – 97.6% to 92.0%;
 - BeO – 1.2% to 8.1%;
 - C, Al, Fe, Mg, Si – trace amounts
- Excellent corrosion resistance like Al
- Be fines are weakly explosive (S1)
- Be metal powder distribution in Figures 1 and 2

Figure 1: Cumulative Mass Fraction versus Particle Diameter
The Mass Median Diameter is $d_G \sim 50\text{-}\mu\text{m}$ ($d_{AED} 68\text{-}\mu\text{m}$)

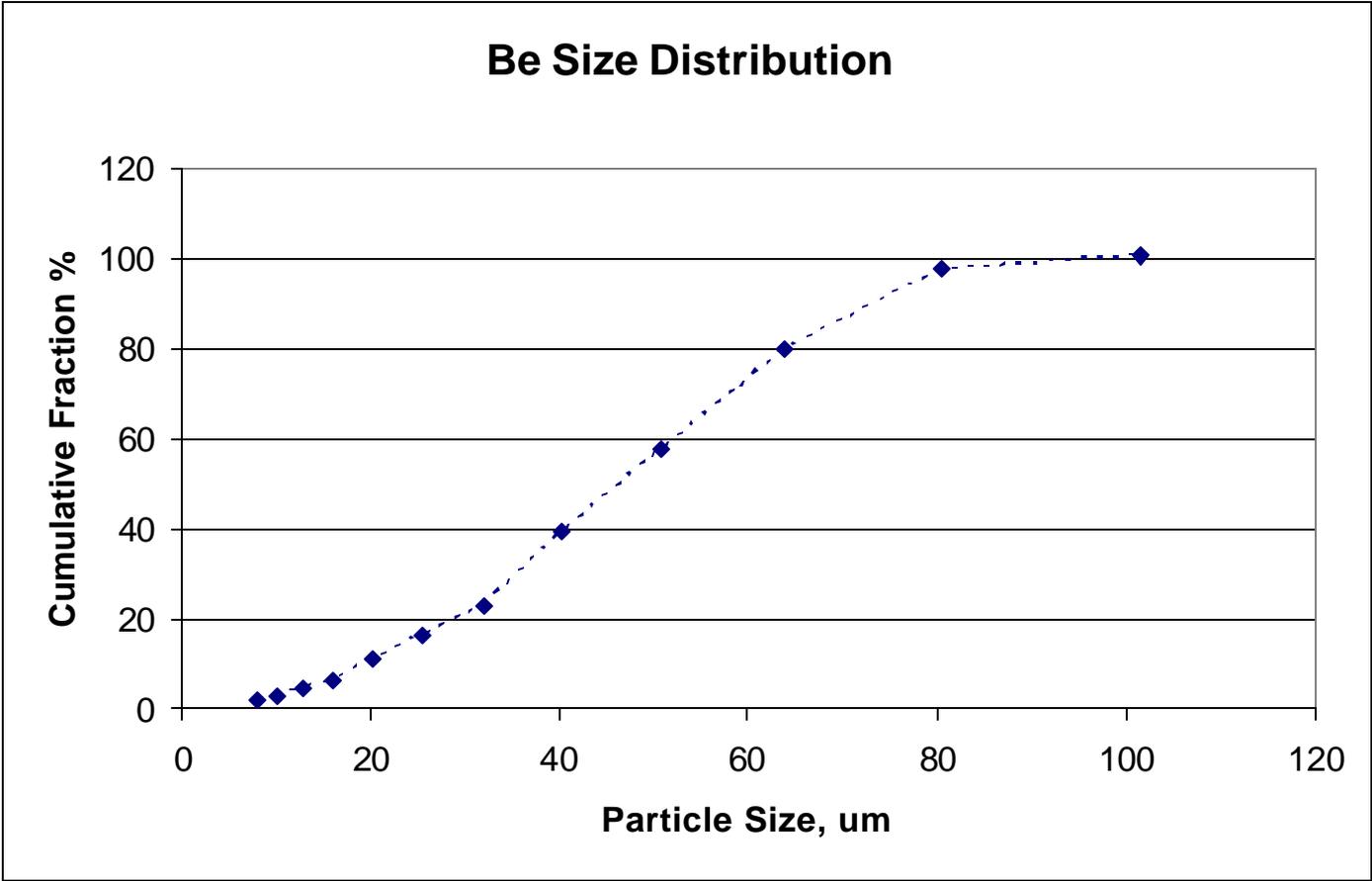
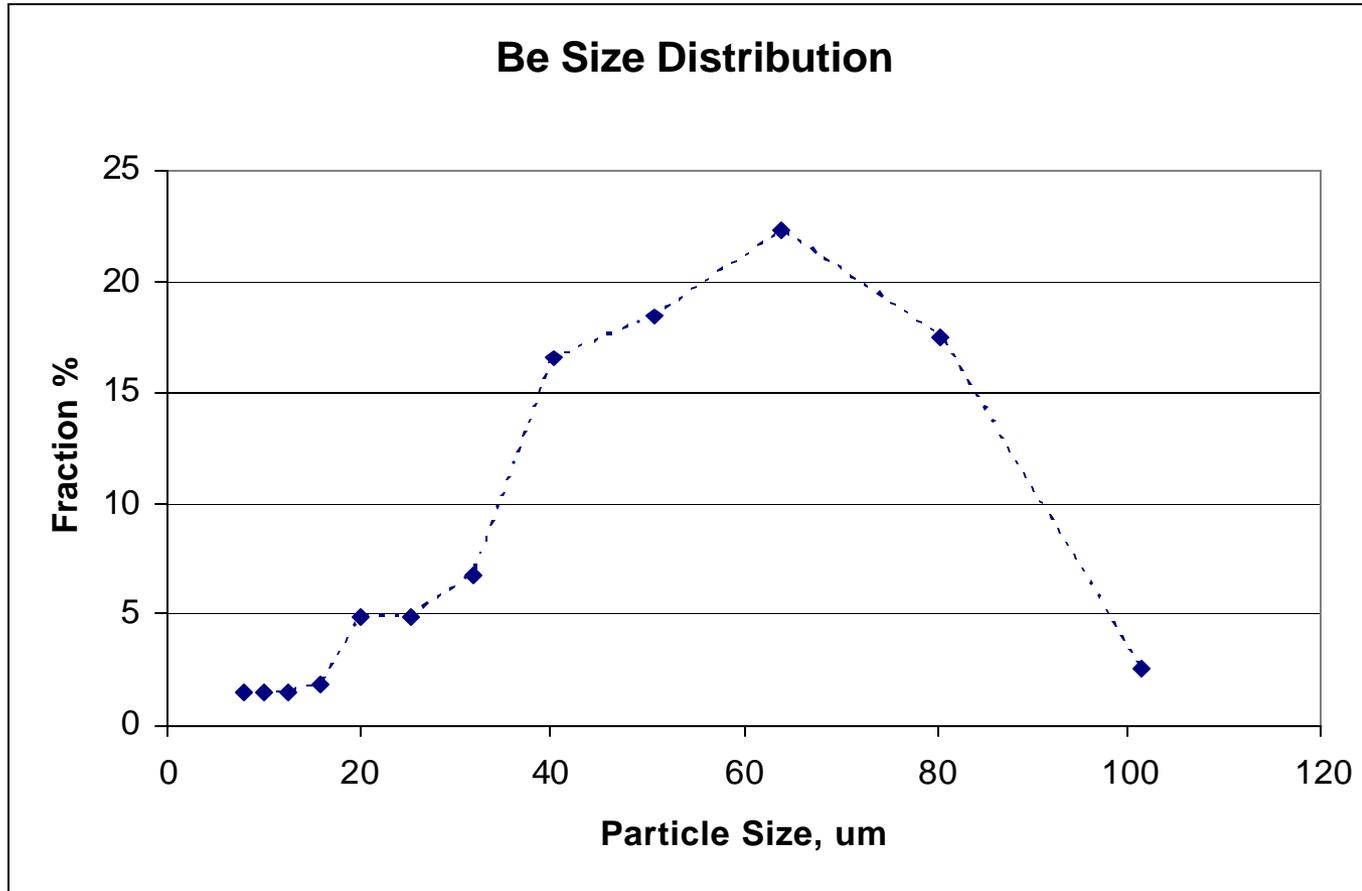


Figure 2. Particle Size Distribution of Brush-Wellman 205 Production Powder, Size vs. Mass Fraction



Oxidation of Be Metal < Ignition Temperature

- All metals have partial pressure of vapors above surface
- Experiments showed oxidation controlled by vapor release through blue oxide layer, surface-to-volume ratio, physical configuration, other factors
- Room temperature: sub-micron layer
- Elevated temperature: white “fluffy” layer
- Ignition occurs when “hot spot” forms a “flaw” and rapidly spreads over entire surface

Ignition of Be Metal

- Wide range of ignition temperatures reported.
- Nominal perspective centered around:
 - ~750 °C for dust < 20 μm if no hydrocarbon contamination
 - > 800 °C for chips/powder
 - > 1000 °C for turnings/swarfs
 - > 1264 °C for large, coherent metal – cannot ignite under most DOE nonreactor nuclear facility fires
- < half (40%) material converted to oxide retained by original particle
- Oxide particle same size as original oxide-encased metal particle
- Much more data in paper on higher temperatures > mp and bp of metal or oxide – very complex phenomena and contradictory experimental results
 - Not typical of DOE facility fires

Summary of ARF and RF Values for Encased Be Metal

Condition	Airborne Release Fraction (ARF)/Respirable Fraction (RF) Values			
	Large, Coherent Items	Powder/Chips	Turnings /Swarfs	Dust Layer
Explosion, detonation	1E-1/0.3	1E-2	1E-2	4E-1
Explosion, deflagration	<1E-6	1E-2	1E-2	4E-1
Explosive Release	<1E-6	1E-3	1E-2	1E-1/0.7
Fire, Be heated	3E-6	2E-5	2E-4	4E-1
Fire, Be ignited		4E-1	4E-1	4E-1
Fire, packaged combustible waste, waste ignited, Be heated	-	2E-5	-	3E-4
Fire, packaged combustible waste, waste and Be ignited	-	-	-	4E-1
Free-fall Spill	<1E-6	<1E-6	<1E-6	2E-3/0.3
Crush-Impact	<1E-6	<1E-6	<1E-6	1E-3/0.3
Shock-Vibration	<1E-6	<1E-6	<1E-6	1E-3/0.3
Resuspension	<1E-6	<1E-6	<1E-6	4E-5/hr (ARR)

Source Term Evaluation

Source term (ST) estimation is defined by use of a five factor formula.

$$ST = MAR \times ARF \times RF \times DR \times LPF$$

For conservative estimate, if RF, DR, and LPF values are unity, then

$$ST = MAR \times ARF$$

Release concentration estimate to a receptor location is defined as

$$\text{Concentration (mg/m}^3\text{)} = \frac{[?/Q \times ST]}{T}$$

T is typically 15 min (TWA). Concentration is then compared with ERPG/TEEL -1, -2, -3 values to evaluate consequence for worker and public.

Screening Criteria

No ARF/RF Required

- 40 CFR 302, Reportable Quantity (RQ)
- 40 CFR 355, Threshold Planning Quantity (TPQ)
- 29 CFR 1910.119 Process Safety Management (PSM), TQ

ARF/RF Required

- 40 CFR 68, Risk Management Plans (RMP) assessment (only solid releases), to evaluate the consequence to the public.
- For some DOE sites, ARF/RF values are required for High and Moderate hazard facility safety analysis for workers and public.
- DOE Guide G 151.1-1 Volume II *Hazards Surveys and Hazards Assessments*, Section 3.5.5 states:
 - ARF_xRFs need to be derived from material properties using basic physical and chemical principles, or Computer codes
- Nuclear DSAs have evaluated significant chemical particulate releases:
 - Pyrophoric metal reactions (e.g., Na)
 - Large quantities of powders (BeO)
 - Situations that could challenge ERPGs/TEELs onsite or offsite

DOE Complex Particulate Release Survey Results

	DOE Site	Chemical	ARF/RF	Comment
1	PNNL	Small quantities*	No	Hanford Sites: Hanford <i>Safety Analysis and Risk Assessment Handbook (SARAH)</i> , Rev. 1
2	ERC	Small quantities	No	
3	Fluor	Small quantities, except Cl ₂	No	
4	Mound	Small quantities	No	D & D Sites Uses SARAH , EPA Model
5	Fernald	Small quantities	No	
6	WVDP	Small quantities	No	
7	RFETS	Small quantities	No	
8	INEEL	Yes	DOE-HDBK-3010	Uses EPA-Model for clean air act Uses ES&H Manual, Vol. 1 LIR300-00-07.3; OST 300-00-06 Uses approach of BeO = UF ₆ ; BeO = ThO ₂ , etc,
9	SRS	Yes	DOE-HDBK-3010	
10	Pantex	Cl ₂	No	
11	SNL	Yes	DOE-HDBK-3010	
12	LLNL	Yes	DOE-HDBK-3010	
13	LANL	Be, Li	DOE-HDBK-3010	
14	Y-12(Oak Ridge)	Be, Ni, Th, Zr, U	DOE-HDBK-3010	

Applicability of DOE-HDBK-3010 (PU, U) Value to Particulate Toxic Material Releases

- If the element's physical and chemical properties are not altered by the accident stresses (fire, explosion, spill, loss of confinement, resuspension), particulate toxic materials generally behave similar to the surrogate materials used in the experimental studies of PU and U in DOE-HDBK-3010-94, then the selection of ARF/RF values are generally appropriate.
- Factors such as density, particle size, deposition rate, and other properties play an important role in the evaluation of the ARF/RF values.
- The sites that use ARF/RF values for chemicals, typically use ARFs/RFs that are based on DOE-HDBK-3010-94, which may not be always applicable.

ERPG-1, -2,-3 Values for Be

- Based on exposure up to one hr of increasing severity. The concentrations are short term exposure limits for protective actions and are based on the toxicity data that contribute the most to health effects.
- ERPG-3 is the “maximum airborne concentration, below which it is believed that nearly all individuals could be exposed for up to one hour without experiencing or developing life threatening health effects”.
- Exposure is largely through inhalation, although absorption through skin can also contribute to the total absorbed dose.
- The ERPGs values are based on total concentration of <10 um + >10 um particle sizes to a receptor. It is not known the proportion of <10 um to >10 um as a function of the receptor distance in a release scenario. However, the <10 um proportion would continuously increase with increasing distance relative to >10 um.

Beryllium Particle Impacts on Health Effects

Beryllium large particles are not likely to impair one's ability to take protective actions during a release event.

- Larger particles, due to deposition velocity, tend to fall out with increasing distance relative to smaller particles.
- Larger particles pose less health risk to the workers and public as compared to smaller particles (<10 μm).
- Health effects related to the exposure of Beryllium aerosol are associated with the deep lung in which the particles <10 μm AED deposit (e.g. Be sensitivity, CBD).

Beryllium: Acute and Chronic Diseases

- Be aerosol exposures can cause two major diseases: 1) Acute exposure ~ ERPG-3; 2) Chronic – a) Sensitization; and b) CBD. Sensitization can occur shortly to years after exposure and is precursor to the CBD.
- Not all those exposed to Be get sensitized.
- There is a lag time between the exposure and sensitization in those who get sensitized.
- The immune system response that results in CBD is generally localized in the part of the lung where respiration occurs and where only particles $<10 \mu\text{m}$ AED deposit.
- The CBD cases are generally far fewer than the sensitization cases. This is because the CBD occurs in fraction of those sensitized. The CBD symptoms may not appear sometime for >10 years. The CBD is treatable but not curable.
- Per Be Rule (10 CFR 850), several requirements are triggered by the action levels of $0.2 \mu\text{g}/\text{m}^3$ (air, 8 hr TWA). This dose is 62 times lower than the ERPG-3 exposure of $100 \mu\text{g}/\text{m}^3$. Other requirements address the potential for exposure and are not dependent on the action level.

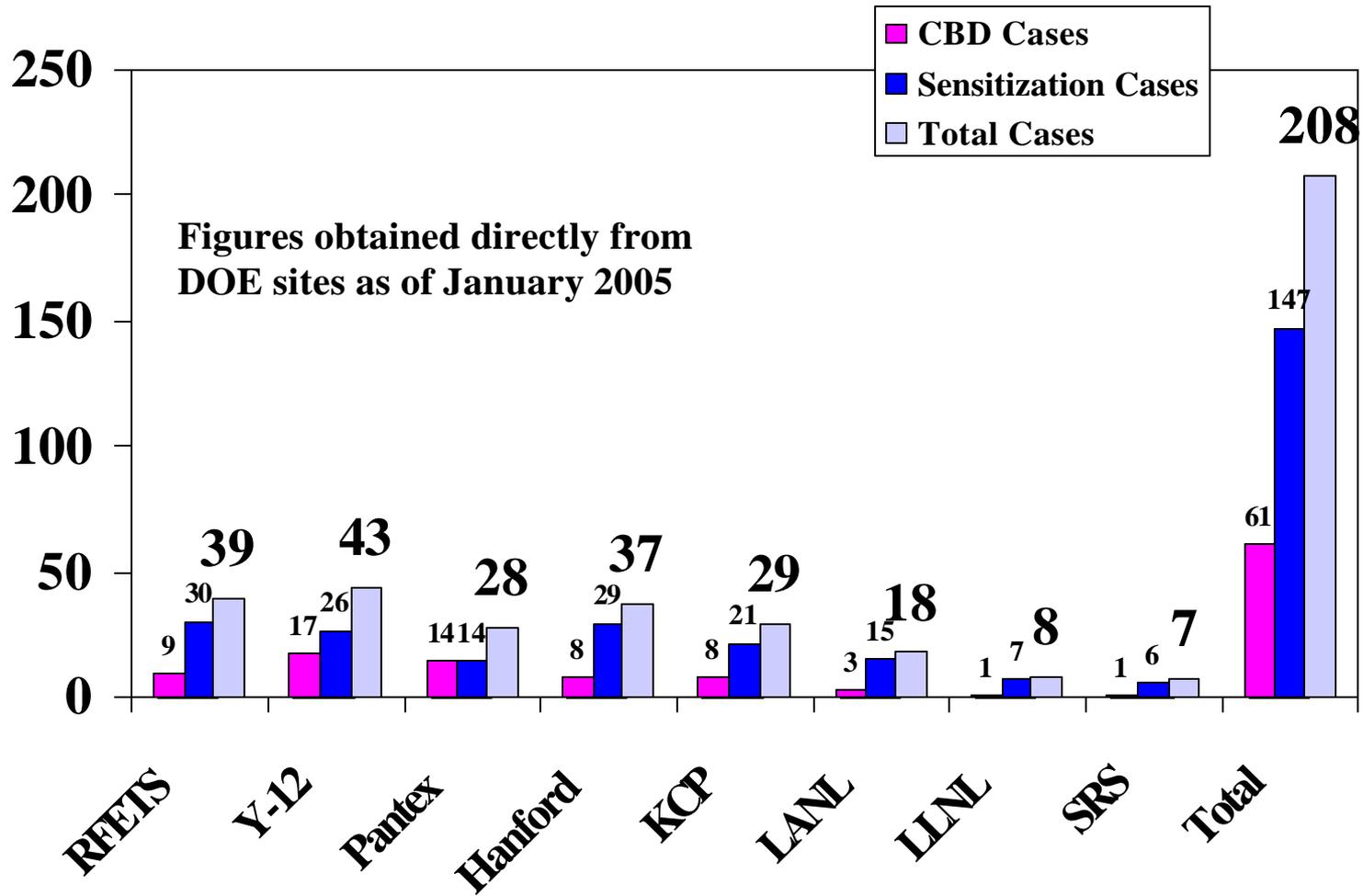
Prevalence of Beryllium Sensitization*

Current Employee Programs	Number of Individuals Tested	Confirmed Positive Be-LPT	Diagnosed CBD	Prevalence of BeS
Oar Ridge National Lab.	93	0		0.0%
Sandia National Lab.	222	0		0.0%
East Tenn. Tech. Park (K-25)	228	2		0.9%
Kansas City Plant	980	13	2	1.3%
Pantex	1239	19	4	1.5%
Nevada	641	13	1	2.0%
Hanford	573	16	4	2.8%
Oak Ridge Y-12	616	20		3.2%
LLNL	150	5		3.3%
Rocky Flats	729	27		3.7%
Los Alamos National Lab.**	1671	15	3	0.9%
Total	7142	129	14	1.8%

* David Weitzman, DOE, Chemical Management Workshop, Nov 4-6, 2003. Data up to 2003; Current Workers.

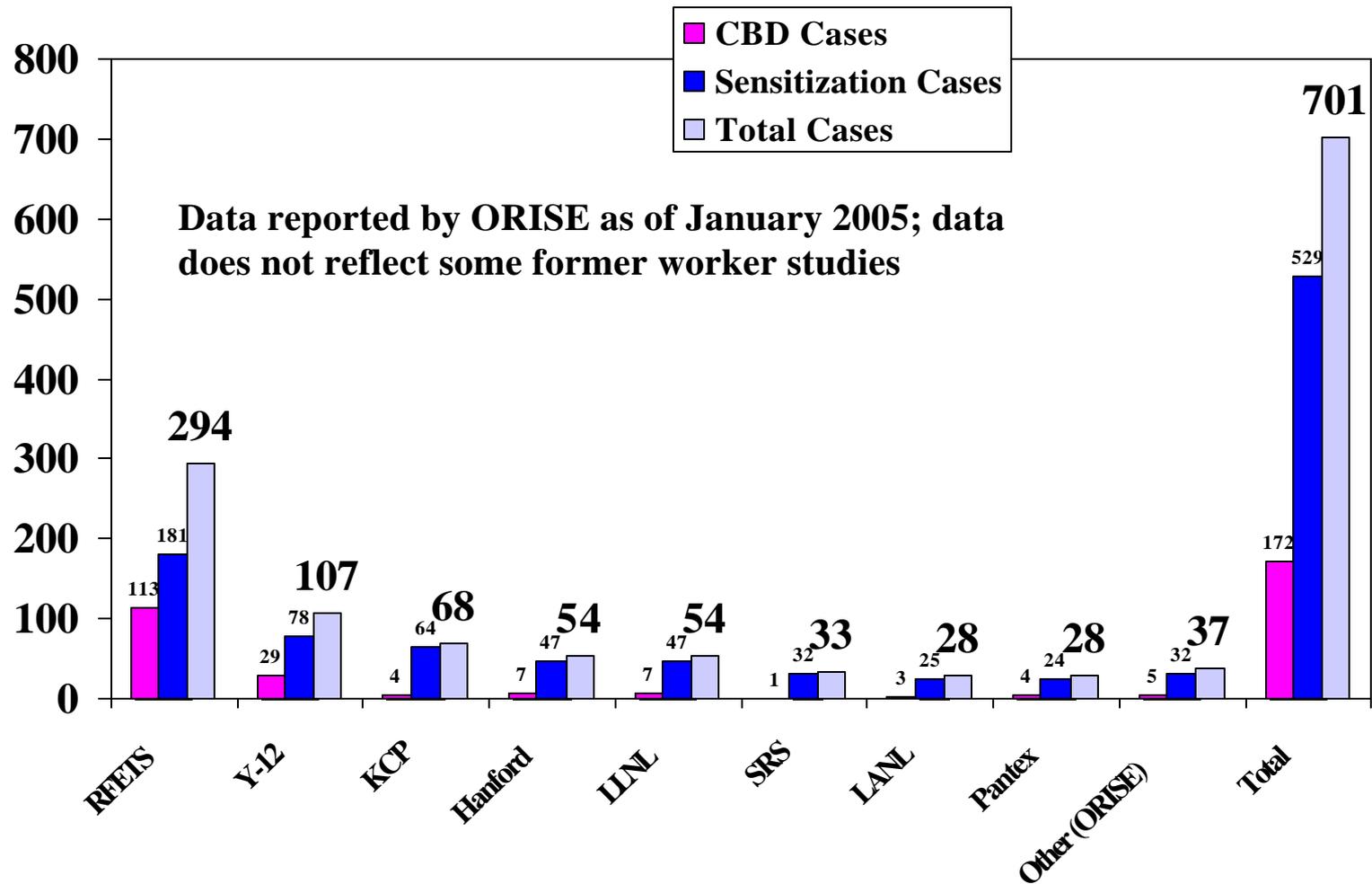
** Gary E. Whitney, HSR-5, LANL; Data up to 2005

DOE Statistics—Current Workers*



* From David Weitzman, DOE-EH-52

DOE statistics—Former Workers*



* From David Weitzman, DOE-EH-52

Hazard Category, Consequence Levels, Controls, and Operational Costs

Category	Criteria	Control*	Operational Cost
High	= ERPG/TEEL-3 or 2 Offsite	EC, DiD, AC, SMP	Very high
Moderate	= ERPG/TEEL-3 or 2 workers at 100m	EC, DiD, AC, SMP	Moderate
Low	Significant health effects to local workers	EC, AC, SMP	Low
Minor	Minor health effects to local workers	SMP	<Low

* EC = Engineered Control ; D&D = Defense-in Depth; AC + Administrative Control; SMP = Safety Management Program
DOE-STD-1186 is applicable for Specific Administrative Controls.

Conclusions

- **The ARF/RF values of Beryllium are governed by the particle size distribution and may not be reliable based on DOE-HDBK-3010.**
- **BeO is 100 times less hazard than Be metal, based on the ERPG-3 values.**
- **The ARF plays an important role in determining consequence for the worker and public and for High, Moderate and Low hazard category.**
- **<10 um (respirable) fraction by inhalation make into the lung compartment, where on deposition develop the conventional health effects – sensitization and CBD. The CBD cases are generally far less than the sensitization cases. This is because the CBD occurs in fraction of those sensitized.**
- **The CBD is mainly due to the immune system response of an individual worker exposed to Be particulates, regardless if the Be operation facility is a Low, Moderate and High hazard facility.**
- **This is a consequence or risk based decision a contractor and DOE/NNSA needs to make. Based on the new ARF/RF consideration, justification may be developed to downgrade a High hazard facility to a Moderate hazard facility.**
- **The level of controls (EC, DiD, AC, SMP) are more rigorous in High and Moderate hazard than in Low hazard facility. However, there are cost considerations, which are very high to operate a High hazard relative to a Moderate hazard and a Low hazard facility.**