REDISPERSION OF INDOOR SURFACE CONTAMINATION: A REVIEW

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Summary

The importance of surface contamination as a potential source of exposure to hazardous materials is discussed. Data from the literature concerning the resuspension of indoor surface contamination are presented. Reported procedures for quantitating surface contamination are compared. It is suggested that, despite its limitations, surface contamination monitoring may be useful in estimating potential risks from hazardous materials.

Introduction

Surfaces contaminated with radioactive, carcinogenic, toxic, or otherwise potentially injurious substances constitute a twofold risk: human exposure may occur through direct contact with a contaminated surface, or materials resuspended from such surfaces by physical agencies such as air currents or vibration may be inhaled. The existence of a relationship between the movements of personnel, sweeping, and other forms of activity and the amount of bacteria airborne in hospital wards was recognized as early as 1890 [1]. The importance of contact transmission and of inhalation exposure in the production of human illness is well known.

Measurements of surface contamination before and after a cleaning procedure allow estimation of the effectiveness of the decontamination process and of the potential risk represented by the residual contamination. This review is concerned with the resuspension of surface contamination into the air of enclosed work environments and the efficiency of removal of surface contamination. With data of this sort, one can predict airborne concentrations of hazardous materials from simple surface contamination measurements.

An international symposium on surface contamination was held in 1964 [2]. Much of the following discussion is based on information presented at that symposium. Aerosol physics, fundamental to a complete understanding of the factors governing the deposition and resuspension of particulate matter, is not included in this review; the subject has been covered elsewhere [3—5]. Information concerning outdoor resuspension of materials such as pesticides and radio-isotopes has also been reported [6—8], for example, but is not considered here.

Redispersion of surface contamination

The relationship between surface and airborne contamination is usually expressed as the ratio of the air concentration of the material of interest (A) to the surface concentration of the material (S). The ratio ($K = A/S$), usually called the resuspension or redispersion factor, is variously expressed in units of m^{-1}, cm^{-1}, or cm^2/m^3. In this paper, all values of K are expressed as m^{-1}. Most studies of redispersion have used radionuclides as tracers; some have employed chemical and microbial agents. Air and surface concentrations would therefore be expressed in terms of disintegrations per unit time, mass units, or organisms or colony-forming units, respectively. If $K = 10^{-6}$ m^{-1}, then one unit of surface contamination/m^2 generated 10^{-6} units of airborne contamination/m^3.

The earliest effort to relate surface contamination and physical activity to the consequent airborne contamination was that of Chamberlain and Stanbury [9]. Their data were used by Dunster [10] to infer, from values for maximal permissible concentrations of radionuclides in air, permissible levels of surface contamination. Since Dunster's work, many values for K have been reported (Table 1).

With some exceptions, the data were obtained using α- or β-emitting radionuclides under normal or experimental "working" conditions. The K factor increases, as expected, with any increase in operations that are likely to resuspend loose surface contaminants. The variation in K factors is considerable even though most of the values tabulated are averages: from 2×10^{-8} m^{-1} under quiescent conditions to 4×10^{-2} m^{-1} with vigorous sweeping of contaminated floors [17,18]. Even with so-called maximal surface disturbances, differences of three orders of magnitude have been reported [9,18]. (This last difference is probably due to the more vigorous resuspension procedure used by Mitchell and Eutsler and the fact that they used surface contamination values based on wipe tests which represented only a fraction of loose contaminant, whereas Chamberlain and Stanbury's values [9] were based on total surface contamination and, therefore, include nontransferable material.)

Brunskill noted that "The resuspension factors . . . can, under many practical conditions, be one or two orders of magnitude greater than the resuspension factor assumed by Dunster . . ." [22]. Others have also reported K factors significantly greater than those proposed by Dunster [16,19,20,25,26]. The variation in K values that can occur even when very similar procedures are carried out by the same experimenters is illustrated by the data of Glauberman et al. [23]. The two orders of magnitude difference found with floors contaminated by plutonium and uranium may be due to the difference in the room sizes which affected the resultant aerosol concentrations, a confined area being compared with the same area of a much larger room. Similar reasoning can be used to explain the fact that one order of magnitude difference in K factors was obtained in the small rooms when cart movement was superimposed on the air turbulence created by the fans, whereas no significant

difference in aerosol concentration (or K values) was detected in the large rooms under similar conditions [23].

The relatively high K factors (2 orders of magnitude greater than those suggested by Dunster) obtained by others indicate the hazard that may be expected from the process of removing contaminated laboratory uniforms in change rooms [20,22]. Others have studied the amounts of uranium released from contaminated uniforms during typical work procedures [28].

In one study, an attempt was made to demonstrate a quantitative relationship between surface contaminant concentration and the amount resuspended by air turbulence [27]. However, it was found that aerosol concentration did not increase with increase in the density of surface contamination and, therefore, the resuspension coefficient tended to decrease.

Also of interest, but not included in Table 1, are values obtained by Fish et al. showing the fractions of loose floor-contaminant (cupric oxide) that settled on various parts of the clothing of a person exposed for 90 min to the aerosol produced by two 10 min periods spent in lightly sweeping the floor, and by the movements required to collect surface and air samples [21]. The settled fraction of contaminant particles varied from about 0.04 on the chest to about 0.7 on the insteps.

The usefulness of the concept of the redispersion factor is limited by the variability of the values obtained. The variability of K may be attributed to the nature of the contaminant, the nature of the surface, and the means by which surface contamination is estimated.

Estimation of surface contamination

Since it is usually not the total amount of contaminant on a surface, but the loose fraction that can be effectively aerosolized, the determination of surface contamination should measure the latter. Probably the most questionable value in the calculation of resuspension factors is the surface contamination, since, in most cases, the fraction of loose contamination removed by the test procedure was not known. Variously named wipe, swipe, smear, or swab tests have been used frequently for the estimation of loose surface contamination. In these procedures, about 10 cm^2 of dry filter paper is firmly wiped over a measured surface area (usually 100 to 1000 cm^2) and the contaminant removed is measured [10,13,16,18,22,29—34]. It is frequently stated that such procedures remove about 10% of the total surface contaminants which represents "loose" material available for resuspension in the air [10,23,31]. Other estimates run as low as 1 or 2% of the total, only 10% of the "loose" component being removed by smears [22]. In at least one case the radioactivity removed by smear samples was considered to represent 100% of the transferable surface contamination [11].

In a study in which nearly all of the surface contamination was loose (thorium dioxide dust, ThO$_2$, was allowed to settle overnight in a small test chamber) about 24 to 75% of the total material was removed from a variety

TABLE 1

Selected resuspension factors

Materials	Surface contamination measurement	Operating conditions	K (m^{-1})	Remarks	Ref.
^{131}I-labelled brick and plaster dust	Total by ratemeter	Active work in open	2×10^{-6}	Air sampled at 4–6 ft. from floor.	9
		Active work in confined, unventilated space	4×10^{-5}	Particle size determined.	
UF$_4$	Loose by wipe	Normal	$2.5{-}20 \times 10^{-5}$	Air sampled up to 4 ft. from floor.	11
		Added ventilation and vibrations	2×10^{-3}		
Ra and U	Total by ratemeter	Normal; highly contaminated plant	Ra: 1.5×10^{-7}	Values calculated from floor contamination data.	12[a]
	Loose by wipe	Normal; plant with good hygiene	Ra: 9×10^{-7}		
		Normal	U: 1.3×10^{-5}		
U compounds	Loose by wipe	Normal; 10 min spot air samples	5×10^{-4}	Air sampled at face level.	13
		Normal; 8 h shift air samples	3.6×10^{-5}		
U	Total by ratemeter	Ore sampling plant, operational	$2{-}10 \times 10^{-6}$	Data collected after removing all loose dust by vacuuming the floors.	14[a]
		U reduction plant, operational	$5{-}140 \times 10^{-5}$		

Material	Sample type	Operation	Value	Remarks	Ref.
Be and compounds	Loose by wipe	Loading and unloading Be blocks	$0.1-9 \times 10^{-3}$	Used mean surface contamination values (size of areas not specified).	
		Cleaning shelves and blocks	$0.14-3.7 \times 10^{-2}$	Used mean of Be block contamination.	
		Preparation of Be cyclotron targets	3.4×10^{-3}	Used mean values.	
		Be compound synthesis	9.3×10^{-3}	Used estimated values.	15[a]
		Warehouse inventory	$3.6-140 \times 10^{-3}$	Used mean air values. (Data from 1st annual survey)	
U compounds	Loose by wipe	Normal; 10 min spot air samples	4.5×10^{-4}	Air sampled at face level. Particle size determined for UO_2F_2	16[a]
		Normal; 8 h shift air samples	1.3×10^{-4}	Urine samples analyzed.	
Plutonium oxide		Quiescent	2×10^{-8}	Aqueous solutions or suspensions of contaminants dried on various floor coverings.	
	Loose by wipe	14 steps/min for 1 h	1×10^{-5}		
		36 steps/min for 1 h	5×10^{-5}		17
Plutonium nitrate		Quiescent	2×10^{-8}	Particle size determined:	
	Loose by wipe	14 steps/min for 1 h	1×10^{-6}	10—20% of respirable size.	
		36 steps/min for 1 h	5×10^{-6}		
Be	Loose by wipe	2 men sweeping vigorously	4×10^{-2}	Unventilated storeroom.	
		2 men sweeping vigorously after vacuuming floor	1×10^{-2}	Air sampled 3 ft from wooden floor (90% of particles nonrespirable).	18
Micro-organisms	Known amount of bacteria applied to floor	1 person walking	2×10^{-3}		19

(continued)

Materials	Surface contamination measurement	Operating conditions	K (m^{-1})	Remarks	Ref.
Various radionuclides	Soil, sand, and dust monitored with ratemeter	Quiescent conditions	1×10^{-6}	Values suggested for indoor conditions based on outdoor studies	20
		Operating conditions	1–10×10^{-5}		
		Handling contaminated clothing	1–10×10^{-4}		
ZnS and CuO dust	Loose (not specified)	Vigorous work & sweeping	2×10^{-4}	Asphalt tile floor, unventilated room.	21
		Vigorous walking	4×10^{-5}		
		Light work	9×10^{-6}	Particle size determined.	
		Light work & light sweeping, fans on	7×10^{-4}		
Pu	Total by ratemeter	4–6 persons walking over contaminated floor	1.2×10^{-4}	Concrete floor brushed each shift, soap and water wash once a day. Air sampled at 1.5 or 4.5 ft. from floor. 9 air changes/h	22
	Loose by wipe	2 persons repeatedly changing contaminated clothing	2.8×10^{-3}	Unventilated, small room	
		2 persons walking continuously in clean clothing	3.8×10^{-3}		
Pu and U	Loose by wipe	Pu plant, small room, fans on	1×10^{-3}		23
		Pu plant, small room, fans on, continuous cart movement	1×10^{-2}		
		U plant, large room, fans on	1×10^{-4}	Air sampled 5 ft. from concrete floor.	
		U plant, large room, fans on, continuous cart movement	3×10^{-4}	Unventilated rooms.	

Material	Source	Conditions	Fan	K factor	Remarks	Ref.
Various α and β emitters	Probably total by ratemeter	Ventilation on; no operations		α: 1.9×10^{-6} β: 7×10^{-7}	Values calculated from floor contamination data.	24*
		Ventilation off, workers enter room		α: 1.9×10^{-6} β: 1.1×10^{-6}		
		Ventilation on; floors rubbed with cotton by remote control		α: 2.1×10^{-4} β: 6.4×10^{-4}		
Ba³⁵SO₄	Not specified	Strike surface with metal plate; fan on		9×10^{-4}	BaSO₄ deposited in test cylinder (4 ft × 1 ft diam.).	25
⁸⁹Sr ⁶⁰Co	Known amounts applied to test sheets	Wind tunnel for 1 h		Sr: 1.2×10^{-4} Co: 3.3×10^{-4}	Solutions dried on surfaces of plastic, steel or painted test sheets.	26
	Not specified	Wind tunnel for 1 h		Sr: 2.8×10^{-2}	Deposited on plastic sheets by ultrasonic aerosol generator for 1 h.	
Be powder	Known amounts applied to surface	Surface conc. (mg/m²)	Fan[b]		Aqueous suspension dried on PVC plate.	27[a]
		0.67	off on	1.5×10^{-6} 13×10^{-6}		
		6.7	off on	2×10^{-7} 7×10^{-7}		
		67	off on	1×10^{-8} 2×10^{-7}		
Ammonium fluoroberyllate	Known amounts applied to surface	Surface conc. (mg/m²)	Fan[b]		Aqueous solution dried on PVC plate.	27[a]
		0.67	off on	1.5×10^{-6} 10×10^{-6}		
		6.7	off on	8×10^{-7} 7.4×10^{-7}		
		67	off on	7.4×10^{-8} 3.1×10^{-7}		

[a] K factors calculated from reported data. [b] Fan in test chamber produced an air flow of 1.8 m/sec.

of surfaces by wiping with filter paper [30]. Forty-four to 86% was removed by pressing adhesive paper to the surfaces. Adhesive paper removed loose ThO_2 dust from surfaces significantly more reproducibly than filter paper wipes. Barry and Solon, who measured the removal of uranium oxide formed naturally on plates of uranium metal, obtained more consistent results with filter paper wipes than from adhesive paper [34]. These contradictory results may be explained by the nature of the association of the contaminant to the surface (an oxide must be more securely bound to its native metal than a dust is to the surface on which it settles). The efficiency of removal of transferable contaminant by wipes or adhesive papers also varies with the type of surface and its degree of roughness [17,30,31,33].

Some reports on the comparison of wet or dry paper wipes indicate that the efficiency of removal depends upon the type of contaminated surfaces as well as the hydrophilic properties of the contaminant [31,33]. Washing the surface with water, with or without added detergent, removed 40 to 50% of the contaminant [22]. However, the contamination removed may have included "fixed" material that would not have been aerosolized under the conditions used for determining K factors and, therefore, does not accurately represent the hazard to personnel.

For measuring microbial contamination, Rodac nutrient agar plates are pressed against the surface to determine the number of loose, colony-forming particles [35—37]. However, neither this procedure nor swabbing appeared to be reliable for quantitating loose bacterial surface contamination, since swabs or plates repeatedly applied to the same surface area gave no decrease in colony counts with successive samples [35]. A similar result was obtained when beryllium was wiped from a wooden surface [18].

A possibly better method has been devised for estimating transferable radioactive surface contamination: the "smair" test uses air impingement to resuspend loose particles which are collected on a filter [30]. Using standardized procedures, this sampler was less efficient in removing loose contamination from a variety of surfaces than were wipe and adhesive paper techniques (1.3 to 33%, as compared with the higher values cited above). However, it was considered to be more reliable for estimating the fraction of transferable contaminant. For example, while wipes or adhesive paper removed similar fractions (38 to 56%) of the ThO_2 dust from greased or ungreased concrete surfaces, the smair technique removed only 1.3% from the greased, as compared with 22% from the ungreased, thus better correlating with the relative aerosol risks. These values were obtained by sampling for 6 seconds at an air velocity of 30 m/sec. Under these standardized conditions, approximately 20% and 65% of ThO_2 particles (0.5 and 5 μm in diameter, respectively) were removed from stainless steel. At an air velocity of 90 m/sec, the values increased to about 70% and 90%. When 21 individuals measured surface contamination using the three methods, personal variance was much lower for the smair technique; wiping led to the greatest variability. The smair technique also removed a relatively constant fraction of particles from plexiglas sheets

of varying surface roughness, whereas the efficiency of the wipe and adhesive paper methods decreased with increasing roughness.

Referring to the smair test, Shapiro commented that while such procedures " . . . are more realistic than transferable contamination tests in evaluating the surface contamination hazard, and correlate better with observed airborne levels, they simulate only partially the mechanisms by which particles are removed from surfaces and produce sustained airborne concentrations" [25]. He acknowledged the absence of a relationship between the wipe technique for estimating surface contamination and potential aerosol hazard. However, he recommended that test procedures be used which simulate the conditions under which surface contamination becomes a hazard, whether by translocation without aerosol formation or by aerosolization resulting from personnel movements.

Others have suggested that it is futile to seek a quantitative correlation between surface contamination and health hazard; they recommend that such measurements of contamination be used as a " . . . qualitative indication of total presence or absence of contaminant" [29]. Several experiments have indicated that the real inhalation risk cannot be estimated from resuspension factors based on measurements of airborne particles, 80 to 90% of which may be too large to penetrate the pulmonary barriers [17,18]. Still others point out that " . . . air-surface contamination relationships derived at one facility will not be valid at another facility except by coincidence" [23]. Utnage concluded that the results of his survey " . . . show no correlation between surface contamination and air concentration . . . " [14]. However, these data were obtained by measuring total floor contamination with an α-survey meter after removing loose dust by vacuuming.

Table 2 summarizes data from the literature that give an indication of the effectiveness of several procedures for removing loose contamination from surfaces.

Other factors affecting resuspension

Particle size, shape, and density determine aerodynamic behavior, which is important not only in regard to resuspension, but also to the likelihood of the particle reaching the gas exchange portion of the lung. Cohesive forces between particles determine the size of agglomerates, which will affect their resuspension and respirability [38]. Adhesive forces between particles and substratum may be decisive in determining whether or not particles become airborne [29,38,39]. The nature of the surface will not only affect adhesiveness, but surface porosity and roughness will also be major factors in determining the relative amounts of fixed and loose (transferable) contaminant associated with the surface [17,25,29,30,35,36]. Hall made an interesting observation: a "smooth" surface has many topographic characteristics of the Rocky Mountains when viewed from the vantage point of a bacterium [35]. Micron- and submicron-sized particles may be relatively "fixed" in

TABLE 2

Efficiency of methods used for sampling loose surface contamination

Contaminant	Method	Surface		Efficiency of removal (%)		Remarks	Ref.
UO_2	Wipe[a]	Uranium plates		No data		Values within 30% of mean	34
UO_2	Adhesive paper[b]	Uranium plates		No data		Not nearly so reproducible as wipes.	34
Pu	Wipe[c]	Granolithic concrete		Assumed 10% of loose removed, equivalent to 1–3% of total		Total by α-ratemeter. Floor brushed each shift, wet mopped with soap each day. 43–50% of total removed by wash with detergent.	22
PuO_2	Wipe[d]	Paper PVC Waxed linoleum Unwaxed linoleum		Not given 14 58 20		Water suspension of PuO_2 applied to surface and dried. $Pu(NO_3)_2$ soln. was absorbed by paper and gave lower recoveries (0.1–0.2%).	17
Na acetate (^{3}H) Paraffin (^{3}H)	Wipe[e]	Shellstone Fiberglass	Na Acetate Paraffin Na Acetate Paraffin	Dry wipe 5–10 18–23 20–30 15–30	Wet wipe 7–19 12–17 26–32 5–6		31
Be	Wipe[f]	Wood		3% of total per wipe. About 20% of loose for each of 3 wipes over same area.		60% of loose removed by 3 wipes, other 40% by water and cloth. Large amount of total remained embedded in wood.	18

	Method	Surface	Data (%)					Notes	Ref
Various α & β emitters	Wipe[g]	Various smooth surfaces	50% under "ideal" conditions 10% under "more practical" tests					300 cm² usually wiped; 1000 cm² gave more consistent results.	32
ThO₂ dust	Wipe[h]	Various[k]	A	B	C	D	E	Total = loose dust (~1 μm particles ~10^6/cm²) measured by α scintillation probe.	30
			70.6	50.5	23.5	39.5	37.5		
	Adhesive paper[i]		73.4	67.7	73.4	55.5	43.5		
	Smair[j]		26.5	10.5	6.6	22.0	1.3		
Various α & β emitters	Wipe[d]	Stainless steel	21% of total activity						24
³H-thymidine	Wipe[l]	Stainless steel	86.1					Soln. air-dried on surface. Counting efficiency not affected by scintillator type or position of paper in vial.	33
		Glass	70.4						
		PVC	28.2						
		Wood	4.2						

[a] Whatman #41 paper (1-1/8" diam.) on #5-1/2 rubber stopper; rubbed 10 times over width of uranium plate (~3.5"). ZnS phosphor scintillation detector for counts.

[b] 1-1/8" diam. disk pressed with rubber stopper on uranium plate. ZnS detector. 3M #216 and Simon Adhesive Co. #2 adhesive tapes used.

[c] Dry filter paper wiped over 900 cm² and counted with ratemeter.

[d] Details not given.

[e] Whatman #1 paper (wet or dry). Counted in liquid scintillation counter (wet wipes were dried before counting).

[f] Whatman #41 paper (5" × 8") wiped once (back and forth) over 1 ft².

[g] Whatman #1 paper (5 cm diam.) wiped firmly over 100, 300, or 1000 cm². Counted in drawer attached to ratemeter.

[h] Whatman #50 paper on #5 rubber stopper; rubbed over 1-8 different 5.8 cm diam. spots/sample.

[i] 3.8 cm square paper pressed with #10 stopper against 1-14 different spots/sample.

[j] 5 cm² head held for 6 sec/spot (with air flow of 30 m/sec) on 4-40 spots/sample.

[k] Data selected for: A = smooth formica, B = stainless steel, C = rough fiberboard, D = concrete, E = greased concrete.

[l] Various filter papers and paper towel, dry or wetted with water. Counted in liquid scintillation counter.

microscopic crevices, especially when ground in by traffic or when carried deeper into porous surfaces by capillary forces during periods of high humidity or actual wetting by water or other fluids. Floor contamination is often the only parameter considered in determining resuspension factors, but contamination on walls, ceilings, piping, ducts, work benches, equipment, etc., which may well contribute to the total of material made airborne by air movement and mechanical vibrations, is usually ignored. Taking into account these surfaces further complicates the determination of an average value for surface contamination [12,24,26] and contributes to the uncertainty in the resuspension factor.

The K factor is also influenced by the method used to effect redispersion of surface contaminants. Studies with radionuclides and bacteria have demonstrated that resuspension may result from any of several forces or a combination of them such as: abrasion by the movements of people and equipment, mechanical vibrations, electrostatic forces, and fluid forces including air turbulence [11,17,21—23,29,38,40]. Air currents not only dislodge particles from surfaces, but also dilute and remove airborne materials, and thus reduce the air concentration used in calculating resuspension factors.

Conclusion

From the above considerations, it is difficult to ignore the doubts expressed concerning the validity and usefulness of resuspension factors. In an excellent review of surface contamination as related to the determination of "decision levels" for 180 radioisotopes, Healy warned that the resuspension factor is a simple idea for which " . . . the conditions of application and measurement must be rigidly defined because the concept is not completely applicable to all situations," and " . . . care should be used in extrapolating such coefficients very far beyond the conditions existent at the time of the measurement" [41]. This idea had also been expressed earlier by Glauberman et al. [23]. However, the recommendation by Shapiro that K factors be determined for each set of conditions as they are encountered would be impractical in most cases [25]. Nevertheless, in some situations the potential hazards may be sufficient to warrant the use of K factors, their imprecision notwithstanding. In those cases, a range of values should be adopted that would be sufficiently inclusive to permit the estimation of aerosol hazards to be anticipated by determining the concentration of loose surface contaminants. The K factor values selected would depend upon the surfaces under consideration, the contaminant(s), and the nature of the activities performed.

Surface contamination monitoring has not been generally applied to materials such as carcinogens and other toxic chemicals due to the need for sensitive methods for detection and quantitation, but " . . . if the techniques of measurement were easier it is likely that the occupational hygienist would find the monitoring of surface contamination to be a technique as useful to him as it is to his radiological colleague" [42]. Guidelines for permissible

doses of carcinogens have not been developed; it is usually assumed that any exposure is to be avoided. For this reason, as well as the need for special procedures for the detection of each carcinogen, or small groups of carcinogens having similar, reactive, chemical moieties, the application of K factors to the monitoring of carcinogens has not been feasible in the sense that it has been used in radionuclide contamination studies. However, the K factor could be of value in determining the maximal aerosol production to be expected from a measured amount of transferable surface contamination by carcinogens, bacteria, or any other obnoxious material having the requisite physical properties for aerosol formation.

Recently, studies have been initiated at the Los Alamos Scientific Laboratory for determining the limit of detection (LOD) values for certain of the aromatic amines designated by OSHA as cancer-suspect agents [43]. Sensitive chemical spot tests have been applied to samples obtained from various surfaces by a wipe procedure combined with chromogenic, fluorogenic, and thin-layer chromatographic techniques that permit the determination of LOD values of less than 0.2 μg/cm^2. In some cases, LOD values of a few ng/cm^2 have been attainable. The development and application of such procedures to other types of chemical moieties may allow the general application of K factors to areas in which chemical carcinogens are handled.

Finally, in addition to the hazard from resuspended surface contaminants, one must not neglect other sources of aerosol formation associated with specific procedures such as the routine methods associated with processing and manipulating radionuclides, carcinogens, and other potentially hazardous materials.

Acknowledgements

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