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A PRELIMINARY RADIOLOGICAL ASSESSMENT OF RADON EXHALATION FROM PHOSPHATE GYPSUM PILES AND INACTIVE URANIUM MILL TAILINGS PILES

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EPA Review Notice

PREFACE

The Eastern Environmental Radiation Facility (EERF) participates in the identification of solutions to problem areas as defined by the Office of Radiation Programs (ORP). The Facility provides analytical capability for evaluation and assessment of radiation sources through environmental studies and surveillance and analysis. The EERF provides technical assistance to the State and local health departments in their radiological health programs and provides special analytical support for Environmental Protection Agency Regional Offices and other federal government agencies as requested.

This study is one of several current projects which the EERF is conducting to assess environmental radiation contributions from naturally occurring radioactivity.

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Abstract

The EPA Office of Radiation Programs has conducted a series of studies to determine the radiological impact of the phosphate mining and milling industry. This report describes the efforts to estimate cumulative working level months (CWLM) from radon-222 daughters produced from radium-226 in phosphate gypsum piles and how these estimates compare with CWLM from inactive uranium mill tailings piles.

Two Florida phosphate gypsum piles were selected for radon exhalation rate measurements. Indoor radon concentration, indoor working level, and individual and population CWLM were computed from the exhalation rate and source size for each source category. The calculated results for each source category are tabulated and compared.

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TABLES

I. Introduction

Previous reports (1,2,3,4) have described radiological conditions in and around phosphate mining and milling operations located in Florida. Information on radioactivity distributions in phosphate industry products, byproducts, and waste; dose assessment of phosphate industry personnel; dose estimates to the general population from particulate emissions; and radium-226 concentrations in ground water are included in these publications (1,2,3,4). In Polk County, Florida, and several neighboring counties large deposits of phosphate ore are found. Associated with this phosphate material are significant quantities of natural radioactivity including economically recoverable amounts of uranium contained in the phosphoric acid which is produced from the phosphate ore. The radium-226 activity tends to be concentrated in the waste gypsum (1). The waste gypsum pile then becomes a potential source of radon.*

For many years inactive uranium mill tailings piles have been recognized as a source of relatively large quantities of radon. To test the hypothesis that phosphate gypsum piles may also be a source of relatively large amounts of radon, radon exhalation rate studies were undertaken and recently completed at two phosphate gypsum piles.These exhalation rate data have been converted to radon source terms so that for a nearby residence indoor radon concentration, indoor working level, and individual and population cumulative working level months (CWLM) estimates can be determined by utilizing atmospheric dispersion modeling. Similar calculations were undertaken for inactive uranium mill tailings piles. The uranium mill tailings data used in the calculations were previously published in several EPA reports (5,6,7). The results for each source category are compared.

Some initial work on this subject, which included other large area sources of radon, can be found in a previous EPA publication (5). A theoretical relationship (8) which was developed for inactive uranium mill tailings piles was applied to all source categories because measured radon exhalation rate data were not available at that time for every source category of interest. Based on actual measurements of exhalation rate, the source terms for source categories other than inactive uranium mill tailings piles can be much lower than predicted by this theoretical relationship (8). While the use of this theoretical relationship gives a conservative estimate, there is a danger of grossly overestimating the radon exhalation rates (e.g., the radon exhalation rate for a slag pile resulting from the combustion of coal in producing electrical power) for sources other than inactive uranium mill tailings piles. This theoretical relationship (8) can also give overestimated predictions for inactive uranium mill tailings piles.

As implied in the title of this publication, only radon from the waste piles are considered in this report. Other sources of radon exist at both phosphate and uranium milling operations, but are not discussed in this report. These other sources of radon are thought to be at least an order of magnitude less important than the sources presented. A discussion of airborne particulates, while very important, is not addressed in this publication.

* In this report radon means radon-222.

II. General Description of a Florida Phosphate Gypsum Pile

Phosphate gypsum piles in central Florida range in size from 20 to 130 hectares with a height of 20 to 30 m. The piles range in age from new to several decades old. A gypsum pile is created by slurrying the waste gypsum from the phosphoric acid production building. The slurry is diverted to different portions of the active pile by creating dikes that channel the slurry. The gypsum settles out and part of the water is recycled back to the phosphoric acid production area. When the pile dries out, a crusty surface forms which minimizes wind erosion. If this crusty surface is disturbed, (e.g., by a bulldozer which is used to construct the diking), the wind is free to erode the powdery composition underneath the crusty surface. Modeling of this dusting or resuspension of particulate material is not addressed in this report.

The previous description does not apply to all gypsum piles in all parts of the country. For example, wind erosion and resuspension of gypsum occur with gypsum piles in Idaho (9) because the pile has a sandy consistency which is a function of the type of phosphate ore from which the gypsum is derived and ore benefication prior to phosphoric acid production (1,2). The difference in rainfall rates for Florida and Idaho also influences the amount of dust suppression.

III. Estimation of Source Terms

Table 1 summarizes the information that is required to estimate the radon-222 source term for each source category. The mean exhalation rates are presented for each type of pile. The exhalation rate is expressed in terms of radon activity released per unit area per unit time (e.g., pCi/m^2-min). The exhalation rate depends on a number of variable factors, including the radium-226 specific activity in the pile material, the emanating power (i.e., the amount of radon released per unit generated) of the pile material, atmospheric pressure, and the diffusion coefficient (which includes the moisture content of the pile) for the radon in the pile material. Also, ice and water cover over the pile can drastically reduce the exhalation rate (10). This rate may be determined experimentally by several methods, including the two most common ones utilizing charcoal canisters (10) and 55 gallon drums (accumulator technique) (11). Details of these methods are adequately described in past reports (10,11).

Phosphate Gypsum Pile Exhalation Rates

Table 2 contains a series of exhalation rate results obtained by the charcoal canister method (10) at phosphate gypsum piles. Data are presented for two phosphate gypsum piles sampled at various times and locations (i.e., old and new sections of each pile). The old (inactive) section of each pile constitutes a portion of the overall pile that is not presently being worked (i.e., new gypsum is not being added to this section of the pile). It may include gypsum that has been present on the pile for a number of years. The new (active) section is an area of the overall pile where new material is being slurried to the pile. For the most part, canisters were placed on relatively dry areas of each pile, primarily on the outer edges of the pile. Ideally, the canisters should have been distributed over the entire pile to account for the spatial distribution of radon flux. Since these were operational piles, practical limitations precluded ideal sampling.

Table 1

Source Terms

Inactive Uranium Mill Tailings Piles	Size (hectare)	^{226}Ra Specific Activity (pCi/g)	Exhalation Rate pCi/m^2-min	Source Term Ci/yr	Background **Ci/yr
Maximum (U_{max})	43.3 (370)*[6]	896[6]	66,000	15,000[5]	5.7
Minimum (U_{min})	0.8 (50)*[6]	356[6]	28,000	120[5]	0.1
Shiprock (U_{sh})	35.3 (335)*[7]	700[6]	5,600	1,040[7]	4.6
Average (U_{av})	15.1 (200)*[6,7]	620[6]	39,000	3,200	2.1
Range	0.8 - 43.3[6]	50-980[6]	3,800-72,000	120-15,000[5]	ND
Phosphate Gypsum Piles					
		Range			
Pile A (P_A)***	75.4 (490)*	25(19.2-32.2)	1,600	620	9.7
Pile B (P_B)***	81.7 (510)*	27(12.8-42.8)	1,600	680	10.6
Background					
Worldwide bkg (B_W)	NA	1	25[14]	NA	NA
Polk County FL bkg (B_p)	NA	0.5	18[15]	NA	NA
Active Uranium Mill Tailings Pile					
Typical (U_{act})	60.0 (437)*[17]	560[17]	9,500	3,000[17]	7.9

[ ] Reference number inside brackets.
* Equivalent radius in meters within parentheses.
** Background soil source term if pile were not present (B_W is used).
*** Piles are associated with same plants as given in Partridge et al[3].
NA Not applicable.
ND No data.

Table 2

Exhalation Rates

Phosphate Gypsum Pile	Exhalation rate (pCi/m^2 -min)	
Pile A		
Old section	1.06×10^3	
"	7.22×10^3	
"	1.61×10^3	8/28/78-9/01/78*
"	1.24×10^3	
"	5.07×10^2	
"	1.17×10^3	
"	1.00×10^3	
"	1.51×10^3	9/05/78-9/08/78*
"	8.29×10^2	
"	1.43×10^2	
New section	6.30×10^3	
"	1.26×10^3	
"	1.97×10^3	8/28/78-9/01/78*
"	1.71×10^2	
"	1.39×10^3	
"	2.24×10^2	
"	2.85×10^2	
"	1.70×10^3	9/05/78-9/08/78*
"	1.24×10^3	
"	1.23×10^3	

* Charcoal canister placement dates (e.g., the first group of canisters were placed on the old section 8/28/78 and removed 9/1/78).

Table 2 (Continued)

Exhalation Rates

Phosphate Gypsum Pile	Exhalation rate (pCi/m^2-min)	
Pile B		
Old Section	1.55×10^2	
"	7.72×10^2	
"	8.87×10^2	7/17/78-7/21/78*
"	2.50×10^2	
"	8.37×10^2	
"	2.22×10^3	
"	1.67×10^3	
"	4.24×10^2	7/24/78-7/28/78*
"	3.71×10^2	
"	1.58×10^3	
"	8.00×10^2	
"	2.36×10^3	
"	1.14×10^2	8/07/78-8/11/78*
"	2.46×10^3	
"	5.39×10^2	
New Section	3.79×10^2	
"	2.40×10^3	
"	8.11×10^2	7/17/78-7/21/78*
"	1.09×10^3	
"	1.13×10^2**	
"	1.94×10^3	
"	8.07×10^3	7/24/78-7/28/78*
"	1.96×10^3	
"	1.38×10^3	
"	7.74×10^2	
"	5.01×10^2	
"	3.00×10^3	8/07/78-8/11/78*
"	1.12×10^3	
"	3.92×10^3	

* Charcoal canister placement dates.
** Wet charcoal canister measurement.

An attempt was made to fit both sets of pile data (table 2) to both a normal distribution and a log normal distribution (12,13). Gypsum pile A results did not follow either distribution while pile B results did approximate a log normal distribution. The arithmetic mean exhalation rate for pile A was calculated by assuming a normal distribution, (because pile A results more closely approximate a normal distribution), while pile B exhalation rate is based on an arithmetic mean estimated by assuming a log normal distribution and calculating a geometric mean and a geometric standard deviation. The arithmetic mean exhalation rate for each pile is 1600 pCi/m^2-min, calculated independently of each other as described above.

The exhalation rate measurements for each pile (table 2) vary over almost 2 orders of magnitude. This variation can be explained in part by the nonuniform distribution of radium-226 in the pile material and moisture content throughout the pile. One example of the effect of moisture on exhalation rate can be seen in the pile B result (table 2) described as a "wet charcoal canister measurement." This particular result is the lowest reported individual exhalation rate for the two piles. The time of sampling is an important aspect of attempting to explain these variations. For example, the arithmetic mean for both the old and new sections of pile A for the sampling period, 8/28/78 - 9/1/78, is approximately the same. This is also the case for the sampling period, 9/5/78 - 9/8/78, but the resulting arithmetic means for the two sampling periods differ by more than a factor of 2. This difference possibly can be accounted for by changes in average barometric pressure, total rainfall (if any) during the sampling period, and charcoal canister placement (i.e., location on the pile). The ability of the charcoal canister technique to reproduce the same result under identical conditions should also be mentioned as a plausible explanation for variation. The ability of the counting system to analyze the charcoal canister with precision and accuracy should also be considered. Method reliability and sensitivity are discussed by Countess (10). The reproducibility of this method is expressed in terms of coefficients of variation, corrected for counting errors, ranging from 0.06 to 0.15 (10). A typical lower limit of detection is 2 pCi/m^2-min (10).

Based on an intercomparison study with the Polk County Health Department (PCHD), Winter Haven, Florida, it is felt the actual analysis of the charcoal canister to determine the exhalation rate is not a major contributor to the wide variation in exhalation rates. Table 3 summarizes the results from this intercomparison. The charcoal canisters used in the intercomparison are the same ones placed on the gypsum piles. After the canisters were removed from the gypsum pile, they were first counted by the PCHD and then by the EERF. To determine whether there was any significant difference between results, a simple statistical test was applied to the data. The simplifying assumption was made that if the mean of the differences between corresponding values of each data set was zero or near zero, then there was no significant difference. The mean of the differences plus or minus one standard deviation of the mean encompasses zero; therefore, it is assumed that either set of measurements is equally valid. By choice EERF values were used in tables 1 and 2.

Meteorological parameters such as barometric pressure, wind speed, ambient temperature, humidity or any other factors that might potentially influence radon exhalation rate are not available for this report. One of the purposes of this report is to present phosphate gypsum pile radon exhalation rate data. To utilize this data, at least one simplifying assumption must be made, i.e., the arithmetic mean of the data for each pile represents an annual average condition. A more representative value would result from an entire year's collection of data. A year's worth of data negates the need to know the meteorological parameters identified previously. Unfortunately, collecting data for a year is unfeasible at this time.One other option is to apply the previously discussed meteorological parameters to the radon exhalation rate data and calculate a modified arithmetic mean exhalation rate. The resulting exhalation rate would at best be highly speculative since the relative effect of each meteorological parameter on exhalation rate is not well understood. Therefore, it is felt that the previously indicated values of 1600 pCi/m^2-min for each phosphate gypsum pile are reasonable values to assume for the purposes of this publication.

Inactive Uranium Mill Tailings Pile Exhalation Rates

The exhalation rates for the uranium mill tailings piles (except Shiprock) are estimated by using a theoretical relationship involving the radium-226 specific activity of the tailings material (8). A correction for pile wetness (15% decrease), pile thickness (5% decrease), and stabilization (25% decrease) was also made (6). The 25% decrease for stabilization was only applied to stabilized piles (6). Since uranium mill tailings piles are highly variable in size, radium-226 specific activity, and exhalation rate (6), several situations are included for comparison purposes (i.e., maximum and minimum source terms, average source term, range of source term dependent parameters, and the Shiprock pile). The Shiprock pile exhalation rate was determined by averaging the results from several different measurement techniques (7). The estimated exhalation rate of the Shiprock pile is probably the most reliable value for the inactive uranium mill tailings pile source category. The theoretical equation (8) used to generate the other exhalation rates tends to be conservative, i.e., it overestimates the actual exhalation rate especially when the radium-226 specific activity in the pile material is overestimated. For example, if the theoretical relationship (8) had been used to compute the Shiprock pile exhalation rate, a value of 41,000 pCi/m^2-min (compared to 5,600 pCi/m^2-min : table 1) would have been reported after accounting for pile wetness, pile thickness, and stabilization. The large difference in values is due mostly to the relative importance of stabilization on reducing the exhalation rate. Measurements made before and after stabilization showed a reduction in exhalation rate of slightly greater than 8 for several feet of earth cover over the pile (7). In this report, stabilization was only given a 25% reduction credit for two feet of earth cover (6,8). Before stabilization, the measured exhalation rate was approximately 46,000 pCi/m^2-min (7). A value of 54,000 pCi/m^2-min is calculated by the theoretical relationship (8) after accounting for pile wetness and pile thickness. Looking at a second pile (U_{max}) which is the same as the Salt Lake City, UT, pile, the measured exhalation rate has been reported to be about 19,000 pCi/m^2-min for a series of short term measurements (11), while the value in table 1 (66,000 pCi/m^2-min) was obtained from the theoretical relationship (8) modified for pile wetness and thickness. Again, the predicted value is greater than the measured one. Therefore, in general, any comparisons with phosphate gypsum piles tend to bias the inactive uranium mill tailings piles on the high side since the source terms are thought to be overestimated for the inactive uranium mill tailings pile source category.

Table 3

Intercomparison of Exhalation Rate Measurements

Charcoal Canister #	Exhalation Rate ($aCi \cdot cm^{-2} \cdot sec^{-1}$)*** PCHD*	EERF**
71	1709.79	1479.0
58	357.17	257.8
84	1150.94	1287.0
91	1572.69	1395.0
44	558.33	416.5
43	4132.28	3993.0
79	1489.86	1351.0
61	3017.48	1813.0
85	393.55	187.7
46	880.84	630.4
4	2996.49	3707.0
20	2367.40	2791.0
36	599.84	706.8
42	538.37	618.5
55	2770.23	2639.0
69	2796.42	3238.0
77	11968.23	13450.0
148	2758.84	3259.0
181	1830.78	2304.0
11	1426.57	1333.0
15	3750.83	3936.0
27	178.99	190.4
34	3764.96	4102.0
47	878.56	898.4
52	1322.76	1290.0
71	961.40	835.6
74	5402.35	4958.0
154	1888.22	1869.0
121	6815.69	6534.0
82	1480.04	1767.0
9	1096.43	1204.0
126	2315.52	2677.0
75	2055.20	2072.0
48	752.89	845.5
68	9139.21	10500.0
V†	1774.77	2105.0
151	2617.57	3277.0
37	205.84	284.4
1	1977.97	2319.0
13	1698.39	1957.0
163	1424.81	1670.0

† PCHD designator.

Table 3 (Continued)

	Exhalation Rate ($aCi \cdot cm^{-2} \cdot sec^{-1}$)***	
Charcoal Canister #	PCHD*	EERF**
54	2385.03	2513.0
50	1201.70	1382.0
33	192.84	238.5
175	396.82	372.6
42	440.48	475.3
156	2464.38	2840.0
125	1984.42	2059.0
22	1869.96	2053.0

* Polk County Health Department, Winter Haven, FL; results that are given are as reported to EERF; no significance should be attached to the number of digits reported.

** Eastern Environmental Radiation Facility (USEPA), Montgomery, AL; computer generated results; no significance should be attached to the number of digits reported.

*** Units used by PCHD; aCi = 10^{-18} Ci; 1 aCi/cm^2-sec = 0.6 pCi/m^2-min or 1 pCi/m^2-min = 1.67 aCi/cm^2-sec.

† PCHD designator.

Background Exhalation Rates

Referring to table 1, the worldwide background exhalation rate (25 pCi/m^2-min) is an average value for the entire world for background soil (14). The average U.S. value is approximately the same as the worldwide rate (14). The exhalation rate for Polk County, Florida, background soil was calculated to be 18 pCi/m^2-min, the computed arithmetic mean from accumulator method results (15).

Radon Source Terms

Again referring to table 1, the radon source terms in units of Ci/yr are based on the mean exhalation rate for each source and a representative source area expressed in terms of hectares (1 hectare = 10,000 m^2) and an equivalent radius. These source terms are used as input into a computer code which calculates individual and population doses (16). The computer generated doses are converted to radon concentration, working level, and CWLM. The computer code (16) was written to output doses directly without giving radon concentration and working level; hence, the doses are transformed by hand calculations to concentration and working level.

Discussion of Table 1 Results

By addressing the source size and radium-226 specific activity information in table 1, an almost reciprocal relationship exists between radium-226 specific activity and source size which accounts for the relatively large source terms for phosphate gypsum piles. Even though radium-226 specific activity for uranium mill tailings piles is large in most cases, the pile areas are much smaller, in almost every instance, than the phosphate gypsum piles. A pattern occurs when comparing exhalation rates and source terms. The average uranium mill tailings pile (U_{av}) exhalation rate is much greater than either of the two phosphate gypsum pile exhalation rates, which reflects the greater radium-226 specific activity of the U_{av} pile. Looking at the source terms for each category, the difference is much smaller than was seen previously with the exhalation rate comparison. The relatively large phosphate gypsum piles nullify a large portion of the difference.

Of interest is the radon contribution background soil would make if each pile were not present. The source terms for background soil are presented in table 1 in the last column. In all cases the pile radon source term is much greater than its corresponding background soil component.

Active Uranium Mill Tailings Pile

A typical active uranium mill tailings pile was included for academic interest. The source size for the U_{act} pile is approximately four times larger than that of the U_{av} pile (table 1) but only 25% of the pile is contributing to the radon source term (17). The other 75% of the pile includes the tailings pond and wet beach areas which are assumed to contribute negligibly to the radon source term (17). The exhalation rate for U_{act} in table 1 is averaged over the entire pile. This rate computed for the radon contributing portion of the pile yields an exhalation rate of 38,000 pCi/m^2-min or approximately the same as for U_{av}.

Table 4

Air Concentration, Working Level,

and

Annual Cumulative Working Level Months (CWLM)

Source	Indoor Radon Concentration (pCi/l)(a)	Indoor Working Level(a)	Population Within 80 km	Individual CWLM/Year(a)	Population CWLM (Person-CWLM/year)(b)
U_{max} (c)	3.2	.02	850,000	0.45 (4.0E-3)*(g)	3000
U_{min} (d)	0.07	.0005	36,000	0.01 (1.0E-5)(g)	0.5
U_{sh} (e)	0.45	.003	43,000	0.06 (2.0E-4)(g)	7
U_{av} (d)	1.8	.01	36,000	0.25 (3.0E-4)(g)	10
P_A (f)	0.19	.001	1,300,000	0.03 (2.0E-5)(g)	30
P_B (f)	0.21	.001	1,200,000	0.03 (3.0E-5)(g)	40
U_{act} (d)	1.4	.01	36,000	0.20 (3.0E-4)(g)	10

(a) 800m from the center of each pile for the maximum sector.
(b) Within 80 km of the pile.
(c) Based on Salt Lake City, UT, meteorological data.
(d) Based on Grants, NM, meteorological data.
(e) Based on Farmington, NM, meteorological data.
(f) Based on McCoy AFB (Orlando, FL) meteorological data.
(g) CWLM/year to an average individual within 80 km of the pile (within parentheses).
* 4.0E-3 is equivalent to 4.0×10^{-3}

IV. Radiological Impact Assessment

The computer code AREAC (16) was used to assess the radiological impact of each radon source. A ground level release is assumed in every instance. The source term (Ci/yr) for each source is combined with representative meteorological data to calculate outdoor radon concentrations. The simplifying assumption is made that over a year's period the indoor radon concentration attributable to each pile will approach the annual average outdoor radon concentration resulting from atmospheric dispersion of the pile radon. Working level exposures associated with indoor radon are calculated assuming an indoor exposure at 70 percent equilibrium (18) (i.e., 100 pCi/l radon = 0.7 working level). All reported values (table 4) of radon concentration and working level are for a structure located 800 m from the center of the pile in the maximum wind direction.

To obtain individual CWLM estimates, the indoor radon concentration is multiplied by a CWLM conversion factor (1 pCi/l of radon-222 is equivalent to 0.14 CWLM/year at 75% occupancy) (19). As would be expected, the individual CWLM estimates for the uranium mill tailings piles are typically greater than for phosphate gypsum piles (table 4). The same simplifying assumptions made in calculating indoor radon concentration apply to CWLM predictions.

The population CWLM predictions (person-CWLM/year : table 1) are noteworthy. Due to the relatively large population centers near the Polk County phosphate gypsum piles, the population CWLM for phosphate gypsum piles are significantly greater than for the average inactive uranium mill tailings pile (U_{av}). The U_{av} pile which is thought to be fairly typical in its population distribution for that source category (i.e., a low population density within 80 km of the pile). The preceding comparison also applies to a typical active uranium mill tailings pile (U_{act}). At least one exception to the aforementioned remarks is the uranium mill tailings pile located close to Salt Lake City. With a combination of a large source term (same as U_{max})and proximity to Salt Lake City, the resulting population CWLM greatly exceeds those for P_A and P_B piles.

V. Summary and Conclusions

Florida phosphate gypsum pile exhalation rate results are provided. These are compared with those calculated for inactive uranium mill tailings. Indoor radon air concentration, indoor working level, and individual and population CWLM were derived from the exhalation rate and source size using the computer code AREAC for each source category. The computed results for each source category are tabulated and compared. The following conclusions are drawn.

(A) The maximum individual CWLM/year exposure due to radon emanation from a typical inactive uranium mill tailings pile is significantly greater than from a typical Florida phosphate gypsum pile which is attributable to the greater radon source term associated with a typical inactive uranium mill tailings pile.

(B) The population CWLM/year exposure within 80 km of a typical Florida phosphate gypsum pile is as great or greater than from a typical inactive uranium mill tailings pile which is a reflection of a greater average population density within 80 km of a typical Florida phosphate gypsum pile.

(C) In order to better define the total risk associated with each source category, a comprehensive radiological analysis is needed. This involves the combining of individual pile results from the entire United States. The best available estimate of the local meteorology, population distribution and source term ideally should be incorporated into this effort. Total health effects for each source category can then be determined.

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