Dr. Denwood F. Ross Jr.
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U.S. Nuclear Regulatory Commission
Washington, D.C. 20555

Re: NUREG-0668 Staff Review of "Radioecological Assessment of Wyhl
Nuclear Power Plant"

Dear Dr. Ross;

I realize that comments on the NRC review of the Heidelberg Report
were due to your office by the end of October, but being relatively
new in this area of study I am at least a couple of monthes behind.
I hope you will be able to accept these comments from a physician
who is concerned about the possible health effects of the current
nuclear power program.

I must say that, perhaps because of my lack of prior involvement with
the NRC and its publications, I was somewhat surprised at the overall
tone of the reply to the Heidelberg group. It seemed unnecessarily
antagonistic and certainly uncritical of current NRC thought on the
subject of the biological implications of nuclear power plants. The
Heidelberg report raises a number of inovative approaches which if
properly integrated could raise our current understanding of this
very important subject. It seems to me that to be entirely critical
of the Heidelberg study without reexamining one's own assumptions
ignores the spirit of science to which we should be striving.

For example, it seems resonable that the soil to plant transfer factors
of cesium and strntium will vary depending on the type of soil and the
type of vegetable, and also depend on other aspects such as concentration
of these isotopes in relation to depth of roots, biological availability,
etc. The attempt to quantify this as done in the Heidelberg report,
although possibly not completely accurate as you have pointed out,
certainly is not without merit, and seems more realistic than assigning
the same value to all vegetables in all soils. Also, given the fact
that our nuclear reactors sit in a variety of climates, soil types,
and agricultural settings, an individual assesment of each power station
would really be needed in order to tell what the predicted or actual
effects of releases might be. This does not appear to be taken into
account by the NRC, but is more specifically addressed by the Heidelberg
report. It would seem that there are some lessons to be learned.

I was very interested to read of the ongoing field measurments of levels
of incorporated nuclides as this seems like one of the best ways to
begin to develop a science of environmental effects of released radio-
nuclides. Unfortunately in NUREG-0668 the data was only touched on
lightly with few details, and so does not represent a critique of

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either the Heidelberg or the NRC's assesment of pathways or actual
amounts of incorporated radionuclides. Obviously, in the assesment
of the amount of absorbed nuclides in plant or animal one must be aware
of the prevailing meteorlogical conditions. A field upwind but very
close to a site of release will get much less exposure than a field
at some distance but downwind from the same site, (ref. 1). It is
impossible to say from the data that you present whether this was taken
into account during the sampling. Perhaps the range you mention in
the tables represent differences in location and not differences in
time as you have suggested. Alternatively, it could represent changes
in wind direction or variability in amounts of releases from the nuclear
plants. Again in order to access this particular aspect, each plant
would have to be looked at on an individual basis with regard to pre-
vailing wind directions, specific releases, and ongoing sampling of
a variety of plants, milk, and animals.

I would like to point out that although the predictions of the Heidelberg
group in terms of radiation incorporated into plants and animals does
not fall into the same range as measured by the individual utility
companies, if one takes into account the fact that the measured releases
are significantly less than predicted, the incorporation is well within
the predicted range. For example, Heidelberg calculations of Cesium 137
incorporation into vegetables is based on .4 Ci/yr release, the actual
measured release is given as .0026 Ci/yr, or a ratio of .0065. All
measured values for cesium 137 in vegetables were within or greater
than the range which is predicted from the Heidelberg figures using
this correction factor. The same is also true of Iodine 131 and
cesium 137 in milk. This would suggest to me that the Heidelberg
interpretations of the dispersal of nuclides into the environment is
correct. Nuclear facilities releasing amounts of Cesium 137 and other
nuclides in the range predicted by the Heidelberg group (ref. 2) may
be contaminating the environment to a correspondingly greater extent.

I also find it particularly disturbing that the people who stand to
lose the most by disclosing unusual amounts of releases and environ-
mental contamination are the very ones who are providing the measurments.
This represents an obvious conflict of intrest and would be more appro-
priately handled by an unenvolved party such as the Environmental Pro-
tection Agency or the U.S. Geological Survey.

In summary, I would like to suggest that the study of the environmental
effects of normally functioning nuclear power plants from the perspective
of incorporation of released nuclides into surrounding flora and fauna
should be made into an exact science and is not served by polar attacks
on opposing views, but rather by critical collection and analysis of data.
We currently have the functioning experiments in the form of our existing
nuclear powered electrical generating stations. The scientific community
and the public at large would be well served by an in depth impartial
analysis of the environmental and potential health effects of the releases
from these plants. This study would have to be independant of the industriies
agencies, and individuals who have a vested intrest in the outcome. From
the tone and content of NUREG-0668, it is apparent that this study will not
come from the NRC.

                    Sincerely,

                    Thomas H. Roberts M.D.

SCIENCE

Plutonium Hazard in Respirable Dust on the Surface of Soil

Carl J. Johnson, Ronald R. Tidball and Ronald C. Severson

Plutonium Hazard in Respirable Dust on the Surface of Soil

Abstract. *Plutonium-239 in the fine particulate soil fraction of surface dust is subject to suspension by air currents and is a potential health hazard to humans who may inhale it. This respirable particulate fraction is defined as particles ≤ 5 micrometers. The respirable fraction of surface dust was separated by ultrasonic dispersion and a standard water-sedimentation procedure. Plutonium concentrations in this fraction of off-site soils located downwind from the Rocky Flats Nuclear Weapons Plant (Jefferson County, Colorado) were as much as 380 times the background concentration. It is proposed that this method of evaluation defines more precisely the potential health hazard from the respirable fraction of plutonium-contaminated soils.*

Methods of evaluating Pu inventories in soils are important because of the possibility of soil contamination near Pu processing plants and nuclear generating stations and areas where Pu has been accidentally released—for example, at Palomares. Spain, and Thule, Greenland, where Pu was released in airplane accidents in January 1966 and January 1968. respectively. Evaluation of Pu (*1*) in the soil is of special importance in contaminated areas that are now considered for residential development. One such area is in the vicinity of the Rocky Flats Nuclear Weapons Plant (Jefferson County, Colorado), which is currently operated by Rockwell International for the Energy Research and Development Administration (ERDA). Activities at the plant include processing radioactive chemicals and making weapons from radioactive metals (*2*).

The Colorado State Health Department in 1973 proposed an interim standard for soil contaminated with Pu, setting the maximum allowable concentration at 2 disintegrations per minute per gram (dpm/g) (*3*). Land with Pu concentrations in excess of the standard would require ameliorative treatment before residential development could be approved. However, the standard fails to define "soil." Either single or composite samples of the soil at a depth of 0 to 0.5 cm from numerous locations in a development area are required. Because such samples include soil particles much too large to be resuspended or inhaled, the possible risk to health cannot be properly evaluated (*4*). Further, no provision is made to prevent the treated soil from being recontaminated by redeposition of Pu from more highly contaminated soils upwind. This redeposition mechanism potentially exists because winds in the area exceed 30 km/hour for 500 to 600 hours yearly. Wind speeds commonly reach 130 km/hour or more, with winds blowing predominantly to the east and southeast toward the Denver metropolitan area (Figs. 1 and 2).

The plant is located about 16 km northwest of Denver and about 8 km from the cities of Boulder, Westminster, and Arvada. Approximately 200.000 people live within 16 km and 600.000 people within 32 km of the plant. Residential development is now proposed within about 5 km of the plant (Fig. 1), involving as many as 3000 homes or a potential population of about 10.000 persons (*5*).

Since the plant began operation in 1953, there have been two major fires (1957 and 1969), a large release of Pu to off-site soils from a spill of metal-laden cutting oil, and an accidental release of Pu to the air in 1974. The major sources of off-site contamination are considered to be emissions from the 1957 fire and the oil leakage from corroded barrels of contaminated cutting oil that were stored
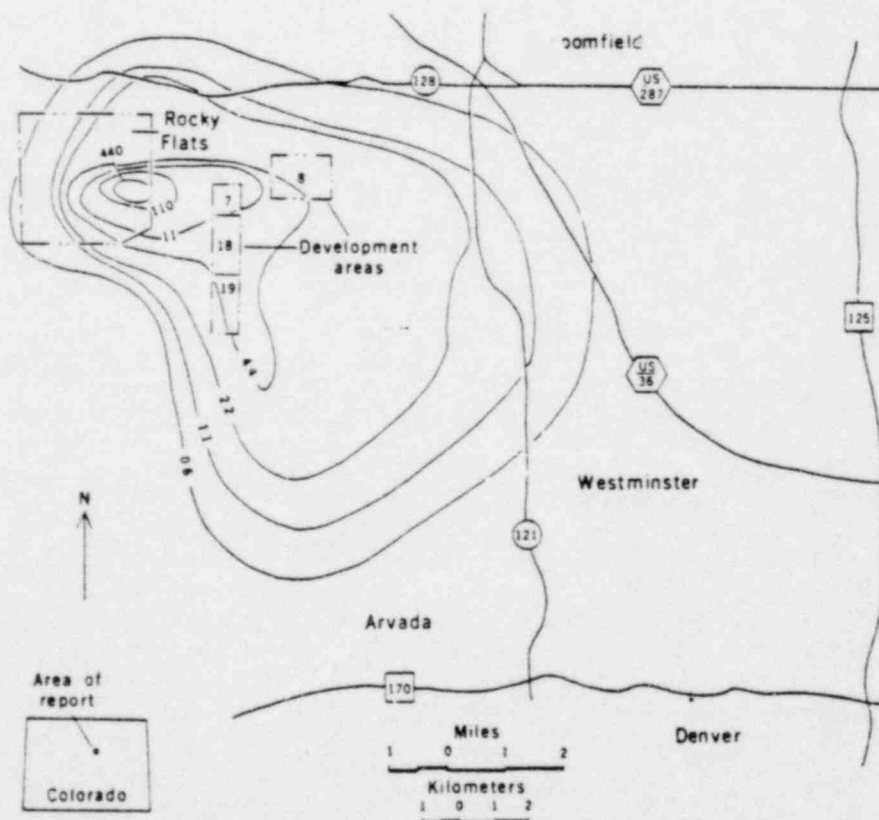

Fig. 1. Rocky Flats Nuclear Weapons Plant and proposed housing development area. Isopleths are labeled in disintegrations per minute per gram of whole soil, calculated from values in (*2*).

outside beginning in 1958 (2, 6). Although leakage of the barrels was first detected in 1964, storage in this manner continued until 1968. The oil-spill area has since been partially covered with asphalt.

A survey of Pu inventories in off-site soils was conducted by the Health and Safety Laboratory of the Atomic Energy Commission in 1970 (2). The results are used in Fig. 1. An off-site area of more than 50 km² had concentrations of Pu in excess of 10 mc/km². Soil samples were collected to a depth of 20 cm, which was considered sufficiently deep to account for the total deposition of Pu. The Pu inventory was based on the weight of the whole soil sample, including plant parts but excluding larger rocks.

Contaminated soils must be measured against a background of Pu released during atmospheric weapons testing. Of the 300 to 500 kc of Pu released worldwide, about 10 to 15 kc is estimated to be present in the soils of the United States and less than 10 percent is still suspended in the atmosphere. In Colorado the background level has been estimated to be 0.04 pc/cm² or 0.08 dpm per gram of whole soil (7).

Sampling localities, each about 4 ha in area, were selected within proposed residential development areas downwind from the plant (Fig. 1). Several sampling sites were randomly selected within each locality. In addition, one stream-sediment sample in section 18 and one sample of eolian sediment in section 19 were collected. Within a 4-m² area at each site, when the ground surface was dry, a representative quantity of loose, surficial (about 0 to 0.5 cm deep) soil material was collected with a clean brush and a clean plastic container. This area provided a sufficiently large composite sample. All samples were compared against a background level estimated from a control sample collected about 23 km south-southeast of the Rocky Flats plant.

The samples were analyzed in random order so that any systematic laboratory error would be converted to a random error. The objective in sample preparation was to disperse the soil microaggregates to expose the Pu as much as possible. Each sample was sieved through a 2-mm stainless steel screen; only the material that passed through the screen was retained for analysis. Approximately 50 g of material in the size fraction ≤ 2 mm was placed on a steam bath and treated with hydrogen peroxide to oxidize the organic material, particularly that present as grain coatings or cementing agents.
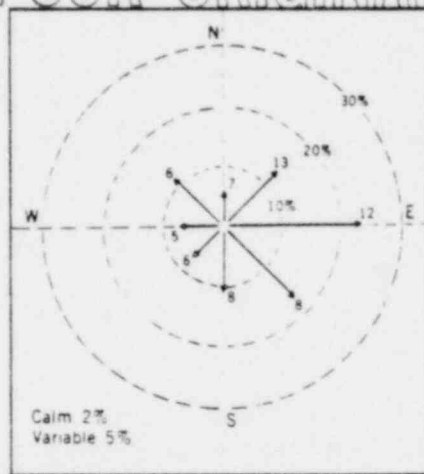

Fig. 2. Rose diagram showing average direction and velocity of wind at Rocky Flats for 1953 to 1970. Arrows point in the direction of wind movement; velocity (miles per hour) is given at the end of each arrow; concentric circles show frequency of wind direction (2).

Table 1. Analyses of Pu in respirable dust (size fraction ≤ 5 μm in soil material ≤ 2 mm) and in whole soil. In column 2 respirable dust is shown as the percentage of whole soil (≤ 2 mm). Localities are shown in Fig. 1. Values are given as disintegrations per minute per gram of material ≤ 5 μm; for Health Department samples these are averages of two analyses. Relative enrichment (RE) is the ratio of measured value to background value.

Local-ity	Health Department samples			Contractor's samples (13)	
	Respirable dust (%)	Pu in respirable dust			
		dpm/g	RE	dpm/g	RE
Section 7					
7-1	36.1	83	180	13.5	169
7-2	41.4	59	130		
7-3	17.9	120	270	14.1	176
7-4	18.8	170	380		
Section 18					
18-1	19.8	36	80	0.2	2.5
18-2	29.6	24	53	0.14	1.8
18-3	27.0	26	58	2.96	37
18-4	25.1	40	89	0.14	1.8
Section 19					
19-1	62.2	1.4*	3.1		
19-2	46.3	2.1†	4.7	0.23	2.9
19-3	36.5	1.3†	2.9		
Section 8					
8-1	47.7	1.1†	2.4	0.05	0.6
8-2	51.2	1.0†	2.2		
8-3	42.1	3.8†	8.4		
8-4	31.5	2.8†	6.2		
8-5	47.0	9.6	21	0.72	9.0
8-6	48.8	8.1	18		
8-7	37.6	11	24		
8-8	33.3	7.7	17		
8-9	24.8	14	31	0.72	9.0
8-10	25.7	19	42		
8-11	34.5	9.4	21		
8-12	37.9	6.9	15		
Background					
Control	48.9	0.45	1	0.08	1

*Eolian sediment from ground surface. †Sample from recently disturbed soil.

The samples were washed and filtered to remove soluble salts and dispersed with a 300-watt ultrasonic probe for 15 minutes (8).

Particles were separated according to size by a standard water-sedimentation technique (9). Sodium metaphosphate was added when necessary to avoid flocculation. The suspension containing the desired size fraction was collected and freeze-dried (10). The sedimentation technique ary measure of the "effective" an of particles with irregular shapes that have settling rates equivalent to those of spheres of the same diameter and density. The threshold parameters used were based on particles of plutonium oxide having an effective maximum diameter of 5 μm and a density of 11.46 g/cm³. The soil particles separated include other mineral grains that have an equivalent maximum settling velocity, but that also have some combination of smaller density and larger diameter. The size fraction thus defined is hereafter called the soil material ≤ 5 μm or "respirable dust" because we assume this size fraction to be an adequate measure of the respirable material.

Plutonium concentrations are given in Table 1. The results shown are the averages of the determinations for duplicate split samples by two laboratories (1). Averaging the two determinations is justified because a t-test indicates no significant difference between determinations at P = .05. Analytical procedures used for Pu were those described by Talvitie (11, 12).

Our estimate of background is 0.45 dpm per gram of soil material ≤ 5 μm. Samples from areas immediately downwind from the plant show evidence of Pu contamination considerably above background in the respirable dust. The sampling area in section 7, which is about km immediately downwind from the plant area, has the largest amounts of Pu: 59 to 170 dpm/g. Sites 7-3 and 7-4 are on the flat crest of a low ridge that trends east from the plant, and sites 7-1 and 7-2 are on the north-facing slope of the same ridge but about 12 m lower in elevation. Section 8, which is on the principal wind vector but about 5 km downwind, has 1 to 19 dpm/g. Although sample sites in section 18 are nearly the same distance from the plant as those in section 7, we measured less Pu there, perhaps because section 18 lies away from the principal wind vector. Section 19, which is farther from the plant and from the principal wind vector, has the lowest values. Eolian sediment (sample 19-1) that was derived from a freshly plowed field upwind

had only 1.4 dpm/g. The field is located in a zone of expected contamination about 3.3 km southeast of the plant area. Plowing tends to distribute the Pu throughout the plowed layer and reduce its probability of entrainment for the present, but it also creates a more erosive surface until plant cover is reestablished. Stream sediment collected in section 18 contained 9.4 dpm/g. This sediment could be susceptible to wind erosion during periods when the stream was at low flow or dried up.

The distribution of Pu in respirable dust may be compared with previous determinations by other methods in the same areas (13). However, the different denominators used to express concentrations make it inappropriate to draw direct comparisons between Pu in respirable dust and in whole soil. The present maximum allowable level in Colorado, 2 dpm per gram of whole soil, represents an enrichment of 25 times the background level for whole soil (0.08 dpm/g). Corresponding enrichment factors based on respirable dust are given in Table 1: for example, sample 7-1 contains 180 times more Pu in the respirable dust fraction than is contained in our background sample.

The surficial soil materials at different locations contain different percentages of material $\leq 5 \mu m$ (Table 1). The Pu concentration per gram represents a potential dose rate. This concentration is to be distinguished from a total inventory of respirable Pu. The greatest long-term hazard, expressed as the largest total inventory, occurs at sites where both the concentration per gram and the percentage of material $\leq 5 \mu m$ are high.

Several sampling sites appear to have been disturbed recently by land-development activity. In particular, sites 8-1 through 8-4 lie in a zone where elevated levels of Pu could be expected. These samples have Pu enrichment factors of no more than about eight times background, compared with 15 to 40 times background only 0.6 km closer to the source (samples 8-5 through 8-12). Disturbance of the ground surface may account for this diminished concentration, or the local topography may have produced a fallout "shadow."

Estimates of health hazards from Pu have been made on the basis of air-monitoring data (14), measurements of total soil inventories of Pu (2, 6), and measurements of Pu concentration on the surface of the soil (13). Air-monitoring data are of importance in estimating human exposure through inhalation, and have been used with soil-contamination data to esti-

mate resuspension factors (15). These factors are influenced by the moisture content of soil, wind speed, elevation above ground, type of ground cover, and presence of paved surfaces. Mechanical disturbances such as those produced by plowing, vehicular traffic, construction work, or street sweeping can resuspend surface dust. However, air-monitoring data and resuspension factors do not account for the total Pu hazard in residential areas (16). Some examples of other types of potential exposure to Pu in the respirable dust on the surface of the soil are listed below.

1) Children playing on the ground or adults working outdoors can pick up mud and dirt on their shoes and clothing and thus introduce dust into their homes. Washing and drying of contaminated clothes can release significant amounts of dust through the exhaust of the dryer. Such a mechanism of exposure has been demonstrated by the finding of characteristic neoplasms in the wives of asbestos workers (17, 18). Dust can also enter a house through windows and ventilating systems and be resuspended by household vacuuming and other cleaning operations (19). Pets track in fine particulates, which may eventually become airborne. These conditions can occur even if a developed area has grass cover.

2) Children playing outdoors may eat food with soiled hands and in other ways ingest or inhale unusual amounts of dust. Heavy-metal intake by this method was found to be important in children with lead poisoning who lived near a smelter at El Paso, Texas (20).

3) Heavily used playgrounds tend to be dusty, and strenuous playground activity can result in suspension and inhalation of dust by children.

4) Local resuspension of dust may occur in the preparation and maintenance of domestic gardens. Plutonium that has been plowed under before residential development may again be exposed by digging for gardens.

The present regulatory code in Colorado requires that soils with radiation contamination that surpass the interim standard of 2 dpm per gram of whole soil must receive special treatment to reduce the hazard to acceptable levels (7). Mixing the soil by plowing is presently accepted as one technique for treatment of contaminated land. We believe that this is an insufficient treatment because (i) plowing tends to displace the Pu particles from the ground surface to some position at depth, but the Pu is still in the soil; (ii) a treated area can be recontaminated either from untreated land upwind

or from subsequent accidental releases of radioactive material; and (iii) subsequent disturbance of the soil by construction activity or cultivation by home gardeners may expose Pu particles at the surface again.

Useful data for evaluating the health hazards of Pu contamination of soil may be obtained by determining the amount of Pu in the respirable-dust fraction (material $\leq 5 \mu m$) on the surface of the soil. It would be more realistic to base interim standards on the respirable-dust fraction because the very small particles in this fraction have the greatest potential for suspension and inhalation.

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References and Notes

1. Samples were analyzed for ^{238}Pu, ^{239}Pu, and ^{240}Pu in laboratories of both the Colorado State Health Department and the Energy Research and Development Administration. Both ^{239}Pu and ^{240}Pu are reported as ^{239}Pu; ^{238}Pu is not reported, but values ranged from 0.04 to 12.4 dpm/g.
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3. R. L. Cleere, unpublished memorandum.
4. C. J. Johnson, unpublished report.
5. _____, in *Proceeding of Public Hearings on Plutonium and Other Trans-Uranium Elements*, vol. 2, *Proceedings of Hearings in Denver, Colorado, January 10, 1975* (No. ORP/CSD-75-1, Environmental Protection Agency, Washington, D.C., 1975), pp. 208-224.
6. S. E. Poet and E. A. Martell, *Health Phys.* **23**, 537 (1972).
7. Final Report of the Lamm-Wirth Task Force on Rocky Flats to the Colorado State Health Department, Denver, 1 October 1975. The following relationship is given in this report: (mc/km²) × 0.2 = dpm/g.
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11. N. A. Talvitie, *Anal. Chem.* **43**, 1827 (1971).
12. _____, *ibid.* **44**, 280 (1972).
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14. M. A. Thompson and D. D. Hornbacher, *Dow Chem. USA Rocky Flats Div. Publ. RFP-ENV-74* (1974).
15. J. W. Healy, *Los Alamos Sci. Lab. Publ. LA 5483-MS, UC-41* (1974).
16. Despite the fact that air-monitoring data do not corroborate the redistribution of Pu, resuspension of contaminated dust from the ground surface has occurred, because we do find Pu widely scattered downwind.
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18. P. Champion, *Am. Rev. Respir. Dis.* **103**, 293 (1968).
19. N. M. Lefcoe and I. I. Inculet, *Arch. Environ. Health* **30**, 565 (1975).
20. B. F. Rosenblum, J. M. Scholtz, J. J. Chisolm, *U.S. Morb. Mortal. Wkly. Rep.* **22**, 405 (1972).
21. We are indebted to G. Lucas of the Colorado School of Mines, who provided assistance in the sample design; M. Hanrahan of the Colorado State Health Department, who assisted in the survey procedure; and to D. L. Bokowski, ERDA, and C. Rising, Colorado State Health Department Laboratories, who performed the analysis for plutonium.

17 February 1976; revised 27 April 1976

Table 5

RADIOLOGICAL SURVEILLANCE STUDIES AT THE OYSTER CREEK BWR NUCLEAR GENERATING STATION

3.3.9 Estimated annual radionuclide discharges. The effluent values discussed in the proceeding parts of Section 3.3 provide the radioactivity source terms for planning environmental measurements. The total discharged radioactivity and the associated radiation doses (discussed in Section 3.3.10) based on estimates from measured values are as follows:

Radionuclide			Estimated annual release,* Ci
Gases			
TRITIUM (as HT)	12.3	-yr.	8.9×10^{-1}
(as HTO)			2.7×10^{1}
CARBON 14	5730.	-vr.	9.1
NITROGEN 13	10.	-min.	$1. \times 10^{3}$
KRYPTON 83M	1.86	-hr.	3.1×10^{4}
KRYPTON 85M	4.48	-hr.	6.9×10^{4}
KRYPTON 86	10.7	-yr.	1.7×10^{2}
KRYPTON 87	76.3	-min.	1.3×10^{5}
KRYPTON 88	2.80	-hr.	1.4×10^{5}
KRYPTON 89	3.16	-min.	8.3×10^{2}
XENON 133	11.9	-d.	3.7×10^{2}
XENON 133M	2.25	-d.	5.1×10^{3}
XENON 135	5.29	-d.	1.6×10^{5}
XENON 135M	15.65	-min	8.9×10^{4}
XENON 136	9.15	-hr.	3.0×10^{5}
XENON 137	3.83	-min.	1.5×10^{3}
XENON 138	14.17	-min.	6.2×10^{4}
Particles and ^{131}I			
CHROMIUM 51	27.7	-d.	2.5×10^{-1}
MANGANESE 54	313.	-d.	5.7×10^{-2}
IRON 55	2.7	-yr.	4.1×10^{-1}
IRON 59	44.6	-d.	8.2×10^{-3}
COBALT 58	71.3	-d.	3.2×10^{-2}
COBALT 60	5.26	-yr.	2.1×10^{-1}
ZINC 65	244.	-d.	6.0×10^{-2}
STRONTIUM 89	50.5	-d.	3.2×10^{-2}
STRONTIUM 90	28.5	-yr.	1.8×10^{-4}
MOLYBDENUM 99	2.76	-d.	6.6×10^{-1}
IODINE 131	8.06	-d.	1.7×10^{1}
CESIUM 134	2.07	-yr.	1.2×10^{-1}
CESIUM 136	13.	-d.	5.7×10^{-2}
CESIUM 137	30.0	-yr.	2.3×10^{-1}
BARIUM 140	12.8	-d.	1.4
CERIUM 141	32.8	-d.	4.7×10^{-2}
NEPTUNIUM 239	2.34	-d.	8.5

* Except for ^{3}H (as HT), ^{14}C (as CO_2 and ^{85}Kr, the annual release represents the sum of the pathways; annual release of the former radionuclides are based on stack measurements. Values apply for an average stack release rate of 3.9×10^{4} uCi/s of gross radioactivity.