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13. ABSTRACT <p>A brief summary of recent literature on the nature of atmospheric releases of radioactive particulates by the nuclear industry is presented. This is to supplement the Task 1 report A005-1. The report first reviews the nature of natural radioactivity in the atmosphere and then discusses the occurrence of uranium and plutonium in air. The various fission and activation isotopes and nuclear industry related isotopes present in the environment are finally discussed.</p> <p>00000000</p>			

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FOREWORD

ENVIRONMENTAL CONTAMINATION BY THE NUCLEAR INDUSTRY

This is a rapid time-limited survey of the literature on atmospheric releases of radioactive particulates by the nuclear industry and is supplemental to the contractually required literature survey reported in IITRI Report No. C6239-A005-1. After completion and reporting of the more general survey on atmospheric aerosols, it was suggested that the survey be expanded in the specific direction of active particulates released in operations of the nuclear industry, especially nuclear reactors. This report covers that specific topic.

Respectfully submitted,

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## TABLE OF CONTENTS

	Page
1. INTRODUCTION . . . . .	1
2. PRELIMINARY SURVEY OF LITERATURE ON RADIOACTIVE PARTICULATES ASSOCIATED WITH ENVIRONMENTAL CONTAMINATION BY THE NUCLEAR INDUSTRY . . . . .	2
2.1 Natural Radioactivity . . . . .	2
2.2 Uranium and Plutonium in the Atmosphere . . . . .	4
2.3 Fission and Activation Isotopes in the Atmosphere . . . . .	5
2.4 Nuclear Industry Related Isotopes in the Environment . . . . .	9
3. REFERENCES . . . . .	22

### Figure

1	Processes in the Atomic Energy Industry . . . . .	6
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## LIST OF TABLES

Table		Page
1	Average Content of Radioactive Elements and Their Active Isotopes in Igneous Rocks . . . . .	2
2	Production Rates, Tropospheric Concentration and Detection of Cosmic Ray-Produced Radionuclides . . . . .	3
3	Some of the More Important Radionuclides Produced by Nuclear Reactions . . . . .	5
4	Isotope Concentrations in Air at the Novo-Voronezh Nuclear Power Station . . . . .	11
5	Principal Particulate Radionuclides in Gaseous Waste Release Assuming 99% Filter Efficiency, 1.0 Ci/sec Noble Gas Stack Release Rate After 30 minute Hold-up Time . . . . .	12
6	Airborne Waste Data, Studsvik and Agesta . . . . .	13
7	Characteristics of Eight Nuclear Power Plants . . . . .	14
8	Average Annual Stack Release Rates - Gross Radionuclides ( $\mu$ Ci/sec) . . . . .	15
9	Typical Releases of Particulate Radionuclides Reported for a Single Year . . . . .	17
10	Summary of Gross Radioactive Waste Discharges - 1966 . . . . .	17
11	Stack Effluent Radionuclide Concentration Nov. 11-13, 1964 Air Particulate Filter . . . . .	18
12	Yearly Average Concentration of Intermediate and Long-Lived Gamma Emitting Isotopes in BGRR Stack Effluent . . . . .	19
13	Inferred Gamma Dose Rates ( $\mu$ /hr) at Several Field Sites . . . . .	20

# ENVIRONMENTAL CONTAMINATION BY THE NUCLEAR INDUSTRY

## 1. INTRODUCTION

The examination of the subject of active particulate contaminants in the environment adjacent to nuclear facilities would not be complete without consideration of normal background radiation and of fallout from weapons testing. Background radiation from natural radioisotopes ordinarily exceeds that introduced by the nuclear industry except during accidental releases or adverse atmospheric conditions. Conversely, weapons testing in the atmosphere can release high levels of tropospheric radioactive fallout downwind of the test area, and stratospheric injection of fission products results in worldwide fallout which takes place over a period of several years. Thus long-lived fallout particles from weapons tests will continue to descend to the earth's surface for some time, although in decreasing amounts unless replenished in a new series of atmospheric tests or deployment in military situations.

The subject of fallout from nuclear weapons has been detailed in literally thousands of reports and publications over the years and no attempt has been made to cover this subject except by brief reference to some of the more recent literature citations. In a like manner, natural radioactivity in the atmosphere is also briefly covered as a prelude to coverage of the nuclear industry.

2. PRELIMINARY SURVEY OF LITERATURE ON RADIOACTIVE PARTICULATES ASSOCIATED WITH ENVIRONMENTAL CONTAMINATION BY THE NUCLEAR INDUSTRY

2.1 Natural Radioactivity

Soils and rocks contain trace amounts of radioactive elements. Table 1 shows the average content of various radioisotopes in igneous rocks according to Rankama (1). While soil derived aerosols are present in significant amounts in the atmosphere, their contribution to natural radioactivity is insignificant. The daughter products of radon and thoron, which emanate from uranium and thorium in the ground, do contribute greatly to atmospheric radioactivity, especially the former.

Table 1

AVERAGE CONTENT OF RADIOACTIVE ELEMENTS AND THEIR ACTIVE ISOTOPES IN IGNEOUS ROCKS

Element	Abundance (parts per million)	Radioisotope	Abundance (parts per million)
K	25,000	<sup>40</sup> K	3.08
Rb	350	<sup>87</sup> Rb	97.48
Th	0.14	<sup>232</sup> Th	0.14
La	18.23	<sup>138</sup> La	0.02
Nd	23.9	<sup>147</sup> Nd	1.34
Sm	6.47	<sup>147</sup> Sm	0.98
Eu	0.75	<sup>152</sup> Eu	0.02
Re	0.05	<sup>187</sup> Re	0.03
Th	11.5	<sup>232</sup> Th	11.5
U	4.0	<sup>235</sup> U	0.0002
		<sup>238</sup> U	0.03
		<sup>232</sup> Th	3.97

Jacobi, et al. (2), studied the attachment of RaA and RaC' to atmospheric particulates. He concluded that the activity is concentrated around 0.1  $\mu$  radius and between 0.015 and 0.5  $\mu$ , and that 50% of the activity was attached to particles larger than 0.1  $\mu$  radius. These relationships were similar in outside air, indoors and in uranium mines. The much shorter half life of thoron (54.5 sec) compared to radon (3.83 days) greatly

reduces the amount of thoron which can leave the soil before decay and attachment to soil particles. Therefore, radon and its decay products will dominate in the atmosphere above the soil.

Cosmic rays interact with atmospheric gases to produce  $H^3$ ,  $C^{14}$  and  $Be^7$  as well as a number of other isotopes (3), Table 2. Carbon 14 is produced by the  $N^{14}$  capture of cosmic ray neutrons. The low incident cosmic ray flux of approximately two neutrons per second per square centimeter of the earth's surface (4) leads to low concentrations of the isotopes compared to the concentration of diluent gases and particulates. As with the radon and thoron daughters, the cosmic ray produced radionuclides will often be associated with atmospheric particles as single atoms or molecules. Tritium, on the other hand, exists in the atmosphere principally in the form of water vapor which precipitates in rain and snow.

Table 2

PRODUCTION RATES, TROPOSPHERIC CONCENTRATION,  
AND DETECTION OF COSMIC RAY-PRODUCED RADIONUCLIDES <sup>a</sup>

Radionuclide	Half-life	Atmospheric production rate (atoms $cm^2/sec.$ )	Tropospheric concentration (dpm/kg air) <sup>b</sup>
$^7Be$	$2.7 \times 10^6$ years	$1.5 \times 10^{-2}$	$7 \times 10^{-4}$
$^{14}C$	$3.1 \times 10^4$ years	$1.1 \times 10^{-2}$	$1.5 \times 10^{-3}$
$^{14}C$	5568 years	1.8	7.6
$^{28}Si$	500 years	$1.6 \times 10^{-4}$	$1.2 \times 10^{-6}$
$^3H$	12.3 years	0.25	$7 \times 10^{-2}$
$^{22}Na$	2.6 years	$5.6 \times 10^{-3}$	$6.7 \times 10^{-5}$
$^{35}S$	88 days	$1.1 \times 10^{-3}$	$7.8 \times 10^{-3}$
$^7Be$	53 days	$8.1 \times 10^{-2}$	0.63
$^{32}P$	25 days	$6.8 \times 10^{-4}$	$7.6 \times 10^{-3}$
$^{32}P$	14.3 days	$8.1 \times 10^{-4}$	$1.1 \times 10^{-3}$
$^{24}Na$	15.1 hours		
$^{32}S$	2.9 hours		
$^{36}Cl$	55 minutes	$1.6 \times 10^{-3}$	
$^{36}Cl$	37 minutes		

<sup>a</sup> From Perkins and Nielsen (3)

<sup>b</sup> Disintegrations per minute per kilogram of air.



Thuronyi (5), Shaw (6), and Lowder, et al. (7), have surveyed the literature on radiation in the atmosphere and Junge (8) has written a chapter on this subject in his book on air chemistry and radioactivity.

## 2.2 Uranium and Plutonium in the Atmosphere

The concentration of U over the Atlantic Ocean (9) in 1959 was found to be 0.0022 to 0.0041 ng U/m<sup>3</sup> while at Sutton, England in 1967-68 the concentration was 0.062 ng/m<sup>3</sup>. Shleien, et al. (10), showed that background levels of Pu<sup>239</sup> in ambient air are approximately 10<sup>-15</sup> to 10<sup>-16</sup> gram Pu<sup>239</sup>/m<sup>3</sup>. Smorodintseva, et al. (11), sampled atmospheric air in the USSR in 1965-66 and found from 1.62 to 12.0 x 10<sup>-21</sup> Ci/l of plutonium-239 with an average of 3.6 and 5.0 x 10<sup>-21</sup> Ci/l for 1965 and 1966, respectively, and with an increase in Pu<sup>239</sup> activity in the spring.

Hoffman and associates (12) have shown that plutonium 244 occurs in nature by chemically isolating 8 x 10<sup>-15</sup> g of Pu<sup>244</sup> from 85 kg of bastnasite ore from the Mountain Pass, California, mine of the Molybdenum Corporation of America. The terrestrial abundance of 10<sup>-18</sup> g Pu<sup>244</sup> per gram of bastnasite suggests that natural background levels are insignificant compared to artificial sources.

Welford and Baird (13) found an average uranium concentration of 0.4 ng/m<sup>3</sup> on filter samples collected in New York City during January and February 1965. Industrial sources can contribute significantly to uranium in the surface air. Martin, et al. (14), recorded a concentration of 13.3 ng U/m<sup>3</sup> at a point 3.7 miles downwind from the 1400 MWe coal fired (1.1 ppm U) power plant at Widows Creek, Alabama, a level which was 35 times higher than the highest level measured around the West Valley nuclear fuel reprocessing plant by McEachern (26).

## 2.3 Fission and Activation Isotopes in the Atmosphere

Radioactive particulates from nuclear events such as fission or activation are artificial radioactive nuclides which are frequently the same regardless of whether the source is a nuclear reactor, a nuclear or thermonuclear bomb, or a plant reprocessing spent nuclear reactor fuel. Fission products are radioactive fragments from the fissioning of uranium or plutonium and the concentration of fission products peaks at about mass numbers 95 and 135 (15). Some of the more important fission products and induced radionuclides are shown in Table 3 (16). Figure 1 shows some of the major processes in the atomic energy industry which can contribute at any point to air pollution by radioisotopes.

Table 3

### SOME OF THE MORE IMPORTANT RADIONUCLIDES PRODUCED BY NUCLEAR REACTIONS

Fission products	Half-life
$^{90}\text{Sr}$	50 days
$^{90}\text{Sr}$	28 years
$^{137}\text{I}$	65 days
$^{131}\text{I}$	8.05 days
$^{131}\text{I}$	20 hours
$^{137}\text{Cs}$	30 years
$^{140}\text{Ba}$	12.8 days
$^{137}\text{Cs}$	285 days
Induced radionuclides	Half-life
$^{60}\text{Co}$	5.3 years
$^{14}\text{C}$	5730 years
$^{59}\text{Fe}$	44.5 days
$^{55}\text{Fe}$	2.7 years
$^{60}\text{Co}$	5.3 years

<sup>3</sup>H Hydrogen-3 (tritium) is also produced in fission.

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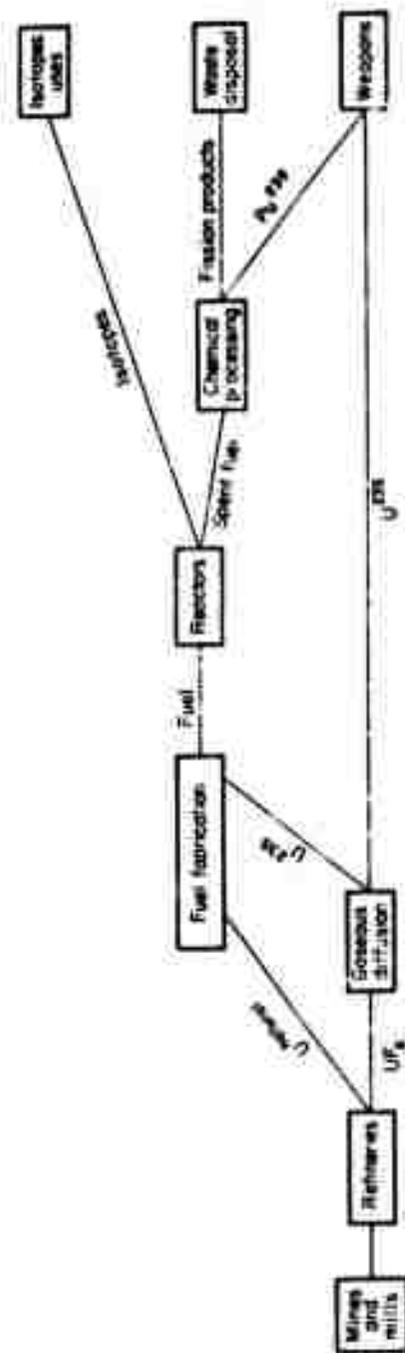


Figure 1. PROCESSES IN THE ATOMIC ENERGY INDUSTRY

During and after World War II, inadequate dust collection devices were initially provided for uranium refineries, and Klevin (17) reports that over 100 tons of uranium were discharged from two plants over a 14-year period. Soil concentrations within 2000 ft of the plants were elevated above the normal background of the region, about 5  $\mu$ g of U/gm of soil.

Air cooled reactors can emit radioactive dust from neutron activation of inert dust particles, and erosion of interior surfaces as well as from defective fuel cladding, but filtration of the supply air and the effluent gases will arrest nearly all of these particles. Filtration of vented gases from boiling water and pressurized water nuclear reactors can also eliminate or minimize radioactive dust emission. Reactor accidents provide the greatest risk of pollution, but reactor containment and conservative safety measures minimize this possibility.

The Windscale reactor accident in England in 1957 (18) was the only reactor accident that resulted in significant environment contamination. A too rapid temperature rise in the fuel cartridges caused fuel failure and ignition, resulting in the release of fission products, mainly  $I^{131}$ , to the atmosphere. The reactor was air cooled, necessitating direct discharge of the contaminants to the atmosphere after filtration to remove particulates. Water cooled reactors, as opposed to the Windscale air cooled reactor, are confined in containment vessels capable of confining any releases to the atmosphere.

When used fuel is reprocessed, the greatest hazards come from gaseous iodine and krypton isotopes, and the enormous amount of radioactivity in the fuel presents transportation, handling, and processing hazards which could easily lead to the release of low levels of particulate radioactive isotopes even during routine operations.

Even though effluent gases are filtered with 99.95+% efficient filters in the atomic energy industry, some of the more penetrating submicron particles, i.e., 0.2 to 0.5  $\mu$ , will

be passed by a HEPA filter. While the amounts passed may be insignificant from a health standpoint, they are certainly detectable and measurable. As pointed out by Hasenclever (19), the passage of very small amounts of radioactive submicron particulates by a high-efficiency filter is a problem that currently exists in nuclear facilities, not only from penetration of the filter itself, but also from leaks and by-passing of filter media, an ever present possibility. During normal operation in a nuclear installation, the formation of radioactive aerosols is not as predictable as the formation of radioactive gases. Particulates can be generated during normal operation from leaks in fuel elements, corrosion of activated components, direct activation of dusts and gases in radiating tubes, and wear of contaminated surfaces, such as control mechanisms, pumps, and the moving parts.

Nuclear weapons exploded in the atmosphere also produce contamination due to fallout of radioactive dust, principally  $Zr^{95}$ ,  $Nb^{95}$ ,  $Ce^{144}$  and  $Sr^{90}$ . The basic measurements on atmospheric radioactivity associated with nuclear weapons tests conducted in the atmosphere from 1945 to 1963 (some 511 megatons) are contained in reports submitted from many nations to the United Nations Scientific Committee on the Effects of Radiation (20).

Pierson (21) also noted the peak fallout of the long-lived  $Sr^{90}$  and  $Cs^{137}$  in 1963 which dropped steadily until 1967 when a third phase of nuclear weapon testing was started. A half-residence time of eleven months for long-lived fission products injected into the stratosphere was noted.

By counting individual hot particles on high volume filter samples taken over a period of three months in 1966, Beck and Kuroda (22) were able to trace fallout particles from the third Chinese nuclear explosion of May 9, 1966, for four complete cycles around the earth. Chen and Kuroda (23) have examined

this data further and find a positive correlation between the variation of meteorological conditions and the daily fluctuations of the particle concentration measured in ground-level air at Fayetteville, Arkansas.

The size spectra of radioactive aerosols in the atmosphere were reported by Schumann (24) who used a Goetz aerosol spectrometer to collect natural radon daughter and thoron daughter aerosols as well as fission product aerosols arising from nuclear test explosions. Results were compared with semi-empirical calculations and with data obtained from the literature.

Konstantinov, et al. (25), have reported on fallout levels and concentrations of gamma-radiating products in the surface air in the vicinity of Moscow from 1962 to 1967. During this period the fallout levels and concentrations in air trended downward from one to two orders of magnitude for  $Ce^{141}$ ,  $Ce^{144}$ ,  $Ru^{103}$ ,  $Ru^{106}$  plus  $Rh^{106}$ ,  $Zr^{95}$  plus  $Nb^{95}$ ,  $Mn^{54}$ , and  $Cs^{137}$ .

#### 2.4 Nuclear Industry Related Isotopes in the Environment

The concentration of uranium in surface air at New York State urban and rural locations was measured by mass spectrometric and fission track analyses (26). Average uranium concentrations ranged from 0.10 to 1.47  $ng/m^3$  (0.035-0.47  $f Ci/m^3$ ). Each area indicated that a significant correlation exists between the uranium concentration and the density of total suspended particulates (TSP), indicating that present levels of uranium in the air are derived primarily from natural sources and non-nuclear industrial activity. The high U/TSP ratios found downwind of a nuclear fuel reprocessing plant 3.5 miles northwest of West Valley, N.Y., showed release of  $U^{235}$  enriched uranium to the local environment. Uranium concentrations were approximately doubled in the downwind surface air at a distance of 1-2 miles away. The amount of  $Pu^{239}$  was about 1% of the uranium ion beam in the mass spectrometric analysis of the samples surrounding the nuclear fuel reprocessing plant.

Breslin and Glauberman (27) measured airborne radioactive dust dispersed by the wind from exposed uranium tailing piles, and the concentrations of the dust decreased in a fairly consistent pattern with distance from the piles. All of the samples were analyzed for total alpha, uranium, thorium 230, radium 226, and lead 210. The highest concentrations were observed at a uranium concentrating mill located in a dry desert region where intermittent extreme gustiness associated with "dust devils" was a characteristic condition.

Rivera-Cordero (28) discusses airborne effluents from the nuclear fuel cycle including environmental contamination at fuel reprocessing plants and releases of radioactive products during normal operation of nuclear power plants.

Examination of HEW data (29) led McEachern, et al. (26) to calculate levels of 90 and 300 ng U/m<sup>3</sup> near gaseous diffusion plants at Paducah, Kentucky, and Piketon, Ohio, respectively, and 6900 ng U/m<sup>3</sup> near a nuclear fuel fabrication plant at Fernald, Ohio, based on the assumption that natural uranium rather than enriched uranium was the predominant uranium isotope. There is some question as to the validity of this assumption as discussed by Kalmon (30), but in defense of the assumption, it does not appear likely that the highly enriched material is preferentially lost at the gaseous diffusion plant. The activity of the low enriched material containing overwhelming quantities of U<sup>238</sup> (about 10<sup>7</sup> atoms U<sup>238</sup>/atom U<sup>235</sup>) in such plant operations would predominate over the combined activities of the other uranium isotopes present unless operations with the costly enriched uranium are much more susceptible to loss to the atmosphere than in the case of the feed and depleted material stream.

Eisenbud and Petrow (31) have reported on studies of the release of radioactivity from nuclear and fossil-fuel power plants, especially uranium.

Cochran, et al. (32), measured  $U^{238}$  levels of less than 6 ng  $U/m^3$  in air at the West Valley nuclear fuel reprocessing plant in 1969.

A summary is given by Kornilov (33) of radioactive waste investigations during some years at different plants in Russia. Aerosols and other effluents are treated in detail. It was noted that dust from dumping areas for solid wastes from hydro-metallurgical plants contaminate the soil and plant life for a distance of about 200 meters.

The concentration of radioactive aerosols in the atmosphere of a Russian nuclear power station was determined (34). Concentrations of long-lived  $\beta$ -emitters ranged between  $2 \times 10^{-13}$  and  $7 \times 10^{-13}$  Ci/l. The long-lived fraction consists of  $Fe^{59}$ ,  $Zr^{95}$ ,  $Zr^{97}$  and  $Co^{60}$ . Concentrations found in the atmosphere are shown in Table 4.

Table 4

ISOTOPE CONCENTRATIONS IN AIR  
AT THE NOVO-VORONEZH NUCLEAR POWER STATION

Isotope	Concentration, Ci/l	Isotope	Concentration, Ci/l
Rb <sup>86</sup>	$3.9 \times 10^{-12}$	Sr <sup>89</sup>	$5 \times 10^{-15}$
Rb <sup>89</sup>	$5 \times 10^{-15}$	Y <sup>91</sup>	$10^{-14}$
I <sup>131</sup>	$1 \times 10^{-13}$	Zr <sup>95</sup>	$1.1 \times 10^{-13}$
Cs <sup>138</sup>	$3.3 \times 10^{-12}$	Zr <sup>97</sup>	$5 \times 10^{-15}$
Ba <sup>141</sup>	$2 \times 10^{-13}$	Ru <sup>106</sup>	$5 \times 10^{-15}$
Ba <sup>142</sup>	$5.6 \times 10^{-13}$	Cs <sup>137</sup>	$10^{-14}$
Cr <sup>51</sup>	$10^{-14}$	La <sup>140</sup>	$5 \times 10^{-15}$
Fe <sup>59</sup>	$7 \times 10^{-14}$	Ce <sup>144</sup>	$10^{-14}$
Co <sup>60</sup>	$5 \times 10^{-15}$	rare earths	$1.7 \times 10^{-12}$

Philbin and Whipple (35) have made an environmental survey for the Nine Mile Point Power Reactor based on operating experience at similar plants such as Dresden (36) and Big Rock Point (37). The empirically based radionuclide release so obtained is shown in Figure 5 for particulates only, assuming a 99% filter efficiency.

Table 5

PRINCIPAL PARTICULATE RADIONUCLIDES  
IN GASEOUS WASTE RELEASE ASSUMING 99% FILTER EFFICIENCY,  
1.0 Ci/sec NOBLE GAS STACK RELEASE RATE  
AFTER 30 MINUTE HOLD-UP TIME

<u>Radionuclide</u>	<u>μ Ci/sec</u>	<u>Radionuclide</u>	<u>μ Ci/sec</u>
Rb <sup>88</sup>	1.2 x 10 <sup>3</sup>	Y <sup>94</sup>	1.4 x 10
Rb <sup>89</sup>	1.2 x 10 <sup>3</sup>	Cs <sup>137</sup>	4.6 x 10 <sup>-3</sup>
Rb <sup>90</sup>	5.8	Cs <sup>138</sup>	2.4 x 10 <sup>3</sup>
Sr <sup>89</sup>	4.8 x 10 <sup>-1</sup>	Cs <sup>139</sup>	3.4 x 10 <sup>2</sup>
Sr <sup>90</sup>	1.7 x 10 <sup>-3</sup>	Ba <sup>139</sup>	2.4 x 10 <sup>2</sup>
Sr <sup>91</sup>	1.7 x 10	Ba <sup>140</sup>	7.2 x 10 <sup>-1</sup>
Sr <sup>92</sup>	2.2 x 10	Ba <sup>141</sup>	2.8 x 10
Sr <sup>93</sup>	1.2 x 10	La <sup>140</sup>	5.6 x 10 <sup>-3</sup>
Y <sup>90</sup>	1.5 x 10 <sup>-3</sup>	La <sup>141</sup>	4.6
Y <sup>91</sup>	2.2 x 10 <sup>-3</sup>	La <sup>143</sup>	3.0
Y <sup>91m</sup>	2.2	Ce <sup>141</sup>	1.2 x 10 <sup>-3</sup>
Y <sup>92</sup>	2.0	Ce <sup>143</sup>	5.8 x 10 <sup>-2</sup>
Y <sup>93</sup>	2.6		

Bergstrom (38) has reported on airborne wastes released from the Studsvik and Agesta reactors in Sweden, Table 6. Note that releases below 1% of the legally permissible releases are recorded as zero, and the aerosols are unspecified  $\beta$  emitters.

Table 6  
AIRBORNE WASTE DATA, STUDSVIK AND AGESTA

Nuclide (category)	Recipient Capacity for Single Nuclide Ci/yr	Permissible Simultaneous Releases Ci/yr	Fallout Source Corresponding to Air Concn Ci/yr		Maximum Releases 1960-67 Ci/yr	Notes
			max	min		
<b>Agesta</b>						
	40 m stack					
Noble gases ( $^{87}\text{Kr}$ equivalents)	800 000	800 000	—	—	20	$^{41}\text{Ar}$ from shield cooling, constant
Halogens ( $^{131}\text{I}$ ) grazing period	40	20	—	—	0	No detectable releases
rest of the yr	12 000	1 200	—	—	9	" " "
Aerosols unspecified $\beta$	4 000	4 000	160	3	0	" " "
"II	8 000 000	—	3 000	120	110	1967; no control necessary)
<b>Studsvik</b>						
	87 m stack	for each source				
Noble gases ( $^{87}\text{Kr}$ equivalents)	2 000 000	200 000	—	—	4 000	R2 reactor, mainly $^{41}\text{Ar}$
Halogens ( $^{131}\text{I}$ ) grazing period	20	10	3	0	about 1	Isotope center 1967
rest of the yr	30 000	320	3	0	about 10	Hot gamma cells 1962
Aerosols unspecified $\beta$	11 000	1 100	400	7	about 1	R2 reactor, fission products
unspecified $\alpha$ ( $^{210}\text{Po}$ )	6	0.6	—	—	0	

Note: The natural  $^{222}\text{Rn}$  concentration corresponds to a source of the order of 10,000 Ci/yr.

The characteristics of eight nuclear power plants in the United States are shown in Table 7. Brinck and Kahn (39) have tabulated annual stack release rates, Table 8, in  $\mu$  Ci/sec for different years through 1967. While the reactor outputs vary over one order of magnitude, the release rates vary over nine orders of magnitude. The boiling water reactors are not as clean as the pressurized water reactors and release rates are generally higher at higher power levels. The release rates also

Table 7

## CHARACTERISTICS OF EIGHT NUCLEAR POWER PLANTS

Name	Shippingport	Dresden I	Yankee	Indian Pt. I	Big Rock Pt	Elk River	Humboldt	Pathfinder
AEC docket no.	None	50-10	50-29	50-3	50-155	115-1	50-133	50-130
Reactor type	PWR	BWR	PWR	PWR	BWR	BWR	BWR	BWR
Manufacturer	W	GE	W	BW	GE	AC	GE	AC
Current reactor heat output (MWt)	231	700	600	615	240	58.2	240	190
Current plant electrical output (MWe net)	60	200	175	270	70.4	22	68.5	56.5
Date of initial criticality	12/10/57	10/15/59	8/19/60	8/2/62	9/27/62	11/19/62	2/16/63	3/24/64
Date of initial full power operation	7/1/58	6/29/60	11/10/60	1/25/63	3/21/63	2/11/64	8/1/63	—
Location	Pa.	Ill.	Mass.	N.Y.	Mich.	Miss.	Calif.	S. Dak.

Manufacturer—W: Westinghouse, GE: General Electric, BW: Babcock & Wilcox, AC: Allis-Chalmers.

Table 8  
 AVERAGE ANNUAL STACK RELEASE RATES - GROSS RADIONUCLIDES ( $\mu$  Ci/sec)

	1959	1960	1961	1962	1963	1964	1965	1966	1967
Shippingport	$4.4 \times 10^4$	$9.2 \times 10^4$	$3.6 \times 10^5$	$3.8 \times 10^4$	$2.4 \times 10^5$	$7.6 \times 10^5$	$1.0 \times 10^3$	$1.0 \times 10^3$	$1.0 \times 10^4$
Dresden I	—	$< 5.0 \times 10^5$	$< 1.3 \times 10^5$	$< 9.0 \times 10^5$	$2.3 \times 10^3$	$1.7 \times 10^4$	$2.0 \times 10^4$	$2.4 \times 10^4$	$6.3 \times 10^5$
Yankee	—	$2.0 \times 10^5$	$6.3 \times 10^5$	$6.9 \times 10^4$	$2.3 \times 10^4$	$3.3 \times 10^5$	$4.1 \times 10^5$	$7.6 \times 10^5$	$1.4 \times 10^4$
Indian Pt. I	—	—	—	—	$2.0 \times 10^4$	$3.5 \times 10^4$	$1.0 \times 10^6$	$1.2 \times 10^6$	$7.5 \times 10^4$
Big Rock Pt.	—	—	—	—	—	$1.6 \times 10^5$	$6.7 \times 10^5$	$2.0 \times 10^4$	$9.1 \times 10^5$
Elk River	—	—	—	—	—	$3.0 \times 10^4$	$7.1 \times 10^6$	$4.1 \times 10^4$	$1.2 \times 10^5$
Humboldt Bay	—	—	—	—	$2.0 \times 10^6$	$2.1 \times 10^4$	$1.0 \times 10^4$	$5.1 \times 10^5$	$3.2 \times 10^4$
Pathfinder	—	—	—	—	—	—	—	$1.8 \times 10^5$	$2.5 \times 10^5$

\*Does not include values for Nov. and Dec.

increase with the operating age of reactor. It should be noted that the incidence and magnitude of repair and experimental modifications (fuel element changes, etc.) can greatly affect average release rates. Design changes will also affect releases. Typical releases of radionuclide particulates reported for a single year are given in Table 9 as given by Brinck and Kahn. A summary of the gross radioactive waste discharges in 1966 from the eight nuclear power stations is shown in Table 10. Almost all of the radioactivity in gaseous waste originated in three boiling water reactors. For 1966 approximately 1,500,000 Ci of gaseous waste was released by these reactors.

Several investigators (40-43) have reported on the radioactive contamination released at the Windscale reactor in England, as discussed earlier in this document (18). Such accidents involving contamination of the external environs are quite rare, with Windscale only involving significant accidental particulate releases which could be detected some distance away as far as the Netherlands (41) and Poland (43).

McClintock (44) has reported that the efficiency of the glass-tex cloth exit air filters at the BNL-BGRR air-cooled reactor is only 80% for particulates in the 0.3  $\mu$  range. Previously, Potter (45) had determined that the particulate material in the filtered exhaust gases had a geometric mean diameter of 0.88  $\mu$  with a standard deviation of 2.38  $\mu$ . This illustrates that despite the presence of HEPA filters in a nuclear facility, the air effluent can contain a perceptible amount of radioactive particles.

Hull (46) has reported on stack effluent from the BGRR air-cooled graphite moderated nuclear research reactor at the Brookhaven National Laboratory, Table 11. These results were reported on filter samples taken in 1964. The data confirm that the isotopes present in the largest concentrations have relatively short half-lives, and that most of the longer lived isotopes in

Table 9

TYPICAL RELEASES OF PARTICULATE RADIONUCLIDES  
REPORTED FOR A SINGLE YEAR  
(Total Activity for 1965 Unless Otherwise Indicated)

	<u>Airborne Particulates (Ci)</u>
Shippingport	--
Dresden	--
Yankee	26.1*
Indian Pt.	--
Big Rock Pt.	--
Elk River	40,000
Humboldt Bay	--
Pathfinder	81,000*

\*For 1966

Table 10

SUMMARY OF GROSS RADIOACTIVE WASTE DISCHARGES - 1966

	<i>Liquid Waste</i> (Ci)	<i>Solid Waste</i> (Ci)	<i>Gasous Waste</i> ( $\mu$ Ci/sec)	<i>License Limit</i> ( $\mu$ Ci/sec)	<i>Average Reactor Power Level</i> (MWt)
Dresden	11.5	173	23,900	$7 \times 10^3$	547
Yankee	0.01	492	0.076	—	518
Indian Pt.	.05	1.5	1.16	—	362
Shippingport	0.06	N.A.*	0.001	—	N.A.*
Big Rock Pt.	0.8	3,100	20,300	$1 \times 10^6$	171
Humboldt Bay	1.77	0	8,100	$5 \times 10^4$	80
Elk River	0.17	82.1	11.4	—	39
Pathfinder	0.05	0	179	$1.5 \times 10^4$	21
Total	65	3,850*	52,500		1,740*

\*Values for Shippingport not available.

the sample would be so diluted in the environment (below  $10^{-15} \mu \text{Ci/m}^3$ ) as to be difficult to detect above the background levels of radiation.

Table 11

STACK EFFLUENT RADIONUCLIDE CONCENTRATION  
 NOV. 11-13, 1964 AIR PARTICULATE FILTER  
 (with half lives  $>0.1$  hr)

Activation Isotopes	$T_{1/2}$	$\mu\text{Ci/m}^3$	Fission Product		
			Isotopes	$T_{1/2}$	$\mu\text{Ci/m}^3$
$^{24}\text{Na}$	15.0 h	472 $\pm$ 25%	$^{90}\text{Mo-Tc}$	14 m	7,590 $\pm$ 25%
$^{82}\text{Br}$	35.7 h	111 $\pm$ 25%	$^{131}\text{I}$	53 m	5,830 $\pm$ 25%
$^{65}\text{Zn}$	245 d	2.4 $\pm$ 25%	$^{95}\text{Zr-Nb}$	6.7 hr	4,020 $\pm$ 25%
$^{60}\text{Co}$	5.26 y	0.7 $\pm$ 50%	$^{97}\text{Zr-Nb}$	17 hr	181 $\pm$ 25%
$^{203}\text{Tl}$	47 d	1.0 $\pm$ 50%	$^{131}\text{I}$	20.8 hr	1,310 $\pm$ 25%
			$^{90}\text{Mo}$	66 h	156 $\pm$ 25%
			$^{131}\text{I}$	8.05 d	130 $\pm$ 25%
			$^{140}\text{Ba-La}$	12.8 d	108 $\pm$ 25%
			$^{239}\text{Np}$	2.10 d	6.8 $\pm$ 50%
			$^{95}\text{Zr-Nb}$	65 d	3.3 $\pm$ 50%
			$^{103}\text{Ru}$	40 d	2.0 $\pm$ 50%
			$^{140}\text{Ce}$	32.5 d	1.1 $\pm$ 50%
			$^{141}\text{Ce}$	285 d	1.0 $\pm$ 50%
			$^{137}\text{Cs}$	30 y	0.6 $\pm$ 50%
			$^{106}\text{Ru}$	1.0 y	0.4 $\pm$ 50%
<b>Total</b>		<b>587</b>			<b>19,310</b>

Later samples (47) taken in 1966 at the BNL-BGRR reactor with a sampling train showed that  $\text{Co}^{60}$ ,  $\text{Zn}^{65}$ ,  $\text{Sb}^{124}$ ,  $\text{Zr-Nb}^{95}$ ,  $\text{Ru}^{103}$ ,  $\text{Ru}^{106}$  and  $\text{C}^{137}$  were largely present in the filterable particulates while  $\text{Sc}^{75}$ ,  $\text{Hg}^{203}$ ,  $\text{I}^{131}$ ,  $\text{Ba-La}^{140}$ ,  $\text{Ce}^{141}$  and  $\text{Ce}^{144}$  were more dominant adsorbed constituents on the charcoal, Table 12.

Beck, et al. (48), have measured the field gamma spectra at various sites in the United States including one at the Argonne National Laboratory (ANL). The natural gamma dose rates from potassium, uranium, and thorium exhibit changes with time at a given location, Table 13. These variations can be explained

Table 12

YEARLY AVERAGE CONCENTRATION OF INTERMEDIATE AND LONG-LIVED GAMMA EMITTING ISOTOPES IN BGRR STACK EFFLUENT

Sample Component	Activation Isotopes (pCi/m <sup>3</sup> )									
	<sup>60</sup> Co	<sup>65</sup> Zn	<sup>75</sup> Se*	<sup>113</sup> Sb	<sup>203</sup> Hg	<sup>60</sup> Co	<sup>65</sup> Zn	<sup>75</sup> Se*	<sup>113</sup> Sb	<sup>203</sup> Hg
	Conc.	%	Conc.	%	Conc.	%	Conc.	%	Conc.	%
Particulate	1.7	96	8.0	96	1.4	27	2.3	82	1.0	5
Charcoal	0.1	4	0.3	4	3.8	73	0.5	18	20.3	95
Total	1.8	100	8.3	100	5.2	100	2.8	100	21.3	100

Sample Component	Fission Product Isotopes (pCi/m <sup>3</sup> )									
	<sup>95</sup> Zr- <sup>95</sup> Nb	<sup>102</sup> Ru	<sup>106</sup> Ru	<sup>137</sup> I	<sup>137</sup> Cs	<sup>140</sup> Ba- <sup>140</sup> La	<sup>144</sup> Ce	<sup>144</sup> Ce	<sup>144</sup> Ce	<sup>144</sup> Ce
	Conc.	%	Conc.	%	Conc.	%	Conc.	%	Conc.	%
Particulate	1.4	77	2.3	60	1.1	79	19.	4.5	2.0	92
Charcoal	0.4	23	1.6	40	0.3	21	413.	95.5	0.2	8
Total	1.8	100	3.9	100	1.4	100	432.	100	2.2	100

\*Identification tentative.

Table 13  
**INFERRED GAMMA DOSE RATES ( $\mu$ /hr) AT SEVERAL FIELD SITES**

Town & Location	Date	Total Gamma				Natural Gamma				Fallout Gamma				Total F.O.
		Gamma	K	U	Th	Total	<sup>137</sup> Cs	<sup>54</sup> Mn	<sup>90</sup> Zr- <sup>90</sup> Nb	<sup>137</sup> Rh	Other			
Pelham, N.Y. (Park)	4-30-65	6.9	2.1	1.4	2.5	6.3	0.6	0.1	—	0.1	—	—	0.8	
	5-21-65	7.9	2.2	1.3	3.1	6.6	0.8	0.2	—	0.2	—	0.2	1.4	
	7-29-65	7.7	2.2	1.5	3.0	6.5	0.8	0.1	—	0.1	—	0.2	1.2	
Mamaroneck, N.Y. (Open Field)	4-30-65	6.8	2.0	1.3	2.8	6.1	0.6	0.1	—	0.1	—	—	0.8	
	5-21-65	7.8	2.2	1.3	3.4	6.9	0.7	0.2	—	0.2	—	0.1	1.2	
	7-29-65	8.0	2.2	1.3	3.3	6.8	0.6	0.2	—	0.2	—	0.2	1.4	
Greenwood Lake, N.Y. (Lot)	4-26-65	8.7	2.0	1.4	4.3	7.7	0.6	0.1	—	0.2	—	0.1	1.0	
	5-18-65	9.1	2.1	1.9	4.1	8.1	0.7	0.1	—	0.2	—	0.1	1.1	
	5-24-65	8.6	2.1	1.7	4.0	7.8	0.6	0.2	—	0.2	—	0.1	1.1	
Carlisle, Pa. (Lawn of Army War College)	8-3-65	7.7	2.4	1.5	3.0	6.9	0.5	0.1	—	0.2	—	0.1	0.9	
	10-1-63	9.5	3.0	1.5	3.1	7.6	—	—	1.6	0.3	—	—	1.9	
	4-5-63	11.2	3.3	1.5	3.6	9.4	—	—	3.4	1.0	—	—	4.4	
Argonne, Ill. (Field, ANL)	8-5-65	8.9	2.0	2.2	2.4	6.6	0.5	0.1	—	0.3	—	1.2( <sup>41</sup> Ar)**	2.1	
	10-3-63	10.8	2.8	1.7	3.0	7.5	—	—	2.5	0.3	—	—	2.8	
	10-15-62	9.5	2.4	1.7	3.0	7.1	—	—	1.9	0.6	—	—	2.5	

\*The difference between the value of the total gamma dose rate and that of the total natural gamma dose rate.

\*\*During the measurement an apparent escape of <sup>41</sup>Ar from a nearby reactor produced a mean gamma dose rate of about 1  $\mu$ /hr.

by variation in the radon content of the lower atmosphere and in the moisture content of the soil. In most cases the natural gamma dose rate accounted for more than 80% of the total gamma dose, but at ANL the natural gamma dose rate was only 69-75% of the total, indicating the possibility that reactor operations contributed to the increased proportion of fallout gamma. In fact, an escape of  $A^{41}$  from a nearby reactor at ANL was indicated in the sample of 8-5-65.

A number of additional references were encountered in this brief time limited literature search which were not abstracted or examined, but which may be of interest should the reader desire to delve more deeply into the subject (49-85).

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