Determining Criteria for the Disposal of Iodine-129

L. L. Burger

October 1980

**Prepared for the U.S. Department of Energy
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SUMMARY

The basic consideration in the disposal of the ^{129}I produced by the nuclear power industry is that humans must be protected from unacceptable radiation risks. Existing standards prescribe maximum concentrations in air and water and, more recently, a maximum release per unit of electrical power production.

This report examines the global quantity, distribution, and rate of movement of ^{127}I (natural iodine), naturally produced ^{129}I , and anthropogenic ^{129}I . The ^{129}I released earlier as a result of nuclear activities over the past few decades is not uniformly dispersed. But the possibility of much greater dispersion exists and, therefore, of much greater dilution than was previously attempted. The potential for dilution with respect to either the ^{129}I concentration or the $^{129}\text{I}/^{127}\text{I}$ ratio far exceeds the minimum required for acceptable exposure to mankind. For utilizing the dilution principle, it is preferable to package and dispose of ^{129}I separately from other fission products. The deep ocean is seen to be the logical location for ultimate disposal.

A set of 14 basic items is described that can be used to set criteria for storage and disposal of ^{129}I . It is suggested that preliminary standards be developed on these and perhaps other items to apply to 1) temporary storage and transportation, 2) disposal to a dry environment with a time limitation on calculated behavior, and 3) disposal to the deep ocean with complete release permitted in 10^3 yr. Early quantification of some of these items will permit better decisions on further research and development needed for iodine removal or control, fixation, and disposal.

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DETERMINING CRITERIA FOR THE DISPOSAL OF IODINE-129

INTRODUCTION

A major obstacle to selecting a storage or disposal system for ^{129}I is that criteria have not been established. Thus no standard exists against which the suitability of a concept or process can be judged. Without these criteria, there is no quantitative measure for the importance of such factors as thermal stability of the fixation material, leach resistance of the package, long-term reliability of the container, disposal site characteristics, isotopic dilution, etc.

The basic considerations are that human life must be protected from unacceptable risks and that the actual selection of a particular system should be based upon a cost/benefit analysis of those systems capable of meeting or exceeding the requirements of existing radiation protection standards. Such an analysis should include as a minimum the following factors:

- estimate of potential radiation dose reduction provided
- monetary cost
- expenditures of material resources and energy required for implementation
- capability to fix ^{129}I resulting from all collection methods
- ability of the waste form to withstand the range of environmental conditions encountered between manufacture and final disposal, including accidents
- ability to recycle its own waste streams so that any ^{129}I releases from the process itself are acceptably low
- adaptability to changing societies.

From these and perhaps other issues a set of measurable criteria must be established, sufficiently definitive to permit the scientist or engineer to evaluate a proposal or a process and sufficiently lucid to preclude political misinterpretation.

This paper does not attempt to specify definite criteria or standards, but rather its goal is to identify factors that are important and to use these to suggest possible fixation, storage, or disposal options. Reprocessing of light water reactor fuel is assumed for the discussion, but this restriction is not required. Likewise, although nearly all the iodine is released from the irradiated fuel during the reprocessing step, any conclusions reached here should apply to all parts of the nuclear fuel cycle.

GENERAL CONSIDERATIONS

In the control and disposal of radioactive wastes ^{129}I is unique from several points of view:

- Its long half-life, $t_{1/2} = 15.7$ million years ($1.7\text{E-}4 \text{ Ci/g}^{(a)}$) and low disintegration energy, total energy, $\beta^- = 0.15 \text{ MeV}$, $\gamma = 0.04 \text{ MeV}$, make it almost nonradioactive.
- On the other hand, the biological importance of iodine (thyroid specificity) decrees environmental evaluation of all iodine isotopes.
- The chemistry of iodine is complex. The element and many of its compounds have high vapor pressures. It exists in many valence states, is chemically reactive, and forms a variety of inorganic and organic compounds.
- The fission yield is relatively high, $\sim 0.8\%$ via the ^{129}I chain^(b) (Walker 1977), and large quantities are produced in the generation of nuclear power.
- Natural iodine, ^{127}I , is abundant throughout the biosphere.
- ^{129}I has always been present in nature, although in very low concentrations.
- The concept of isolation until radioactive decay abates the hazard becomes meaningless in terms of an anthropocentric time frame.

(a) In this report powers of 10 are written as a number following E, e.g., $\text{E-}4 = 10^{-4}$.

(b) The yield depends on the fuel's plutonium content (see Table 3).

The result of these factors is that one has a choice of dilution or temporary isolation, 10^2 to 10^4 yr, with perhaps some help from geochemistry. Storage may be limited by the stability of sociopolitical institutions or the immutability of the physical package. Beyond 10^2 yr the latter is probably the more reliable of the two. Dilution involves mixing ^{129}I with sufficient natural iodine, ^{127}I , such that the biological uptake of ^{129}I is reduced to a negligible level by the maximum total iodine permitted naturally. Both approaches seem reasonable. It must be recognized, however, that the distinction between them may vanish with time, i.e., storage over a period of several half-lives, or even less than a single half-life, may become dispersal--at the mercy of geological phenomena.^(a)

Iodine-129 does not produce enough heat or radiation to cause structural damage to waste forms or containers. Technology is available so that chemically and physically stable compounds can be prepared, fixed in a suitable matrix, such as cement, placed in a corrosion-resistant container, surrounded by additional barriers as an added option, and placed in a stable geological formation so that the probability of escape approaches zero for a period of 100 years or for 1000 years--but not for 10^8 yr. If this route is taken, the question is how far towards this zero release ideal, and for how long, is it necessary to go? If the dilution approach is taken, the questions are: How much dilution is required and can it be easily achieved? In what part of the environment should this dilution occur? Does dilution violate the intent of part (b) of the Environmental Protection Agency (EPA) guidelines, discussed later? For the present, we must assume the answer to the latter question to be "no."

Criteria for ^{129}I disposal must be designed to protect mankind from unacceptable risks for an extremely long time. Often disposal criteria are based on isolation for about 10 half-lives. However, this is not realistic

(a) An appreciation of the long decay time of ^{129}I may be gained by noting that its half-life is greater than the age (10 to 25 million yr) of the Cascade Mountains.

for ^{129}I with its half-life of 16 million years. It is more reasonable to compare ^{129}I with hazardous stable elements such as mercury or chromium and consider disposing of it in such a way that its concentration in air and in drinking water is safely below maximum permissible concentrations (MPC)^(a) or so that its isotopic ratio to stable ^{127}I is low enough to prevent unacceptable accumulation in living creatures.

The following sections will discuss the distribution and exchange rates of iodine and ^{129}I in various global compartments in relation to problems of disposal. Then the current derived standards will be discussed. With these considerations as background, the factors affecting criteria development will be discussed and related to various handling and disposal options.

DISTRIBUTION OF IODINE

Iodine occurs in small amounts in nearly all geologic formations, 0.1 ppm in basalt, 0.1 ppm in granite, 1.7 ppm in sandstone, and 2 ppm in shale (Krauskop 1979). Large concentrations occur in nitrate deposits. For example, Chile saltpeter contains 0.2%. It is also present in all old sea deposits, including oil well brines. The ocean is a large reservoir of iodine from which it is continuously deposited as sediment. Average sea water concentration is about 0.06 g/t. Iodine enters the atmosphere from the ocean surface through photochemically induced oxidation (Miyake 1963) and is removed by rainfall. The concentration in the atmosphere is uncertain. The total airborne is highly variable as is its distribution between aerosol form and molecular I_2 form and smaller amounts of unknown compounds. Estimates range from 10^{-9} to 10^{-7} in marine atmospheres (Duce 1963, Lininger 1966, Bruner 1963, Israel 1973). A minor but important fraction of the iodine is at steady state in the biosphere. The "biosphere" is not used in the general sense but rather will be defined as living terrestrial matter. Models can be devised that carry iodine in a cyclic pattern through various compartments of the globe, the sea, the atmosphere, the soil, etc.

(a) Since MPCs must be ultimately determined by radiation dose, changes in recommended doses imply that the MPCs should also be adjusted. Thus the values in Table 5 may be too high.

Iodine enters the cycle through volcanic action and through weathering of rocks.^(a) Transport back to the sea is by rainfall and river flow. It will be seen that residence times and mixing times are important. From tritium studies it was found that mixing time for ocean water above the thermocline (temperature barrier) is about 12 years (Michel 1975) while for the deep ocean the mixing time is about 100 times greater (Arnold 1959).

Consideration of the ^{129}I disposal must include this cycle of natural iodine. Miyake and Bergman both developed simple models for the cycle (Miyake 1963, Bergman 1971). A preliminary discussion with regard to iodine disposal was given earlier (Burger 1979) that included a brief summary of the distribution of iodine and water throughout the world. Since that time, Kocher has published a detailed model of the iodine cycle (Kocher 1979). It contains a number of refinements over the earlier models, such as three-compartment land and a two-compartment atmosphere. His model is shown in Figure 1. His evaluations of the inventories and transport rates are probably as good as can be done with the present analytical information. Two pathways, however, that we consider important are added; they are shown with dashed lines. No estimates have been made for the many transfer rates. There is considerable uncertainty in many of the rates, for example, the processes involving ocean sediments.

The important factors seem to be: which compartments are in direct interaction with man, what are their iodine inventories, and what is the residence time in each compartment? Residence time, τ , will be defined as the reciprocal of the rate constant for flow out by a given path. Any global model is complex; i.e., there is more than one cyclic path, some short and some long. Not included, for example, in Kocher's model, is the path returning iodine in sediments to the lithosphere, certain to be significant on a geologic time-scale. Table 1 is a simplified listing and shows a somewhat arbitrary division between long and short residence times. Not included is the iodine deep in the earth's crust, in ocean sediments, $\sim 1\text{E}16\text{--}1\text{E}18$ g, introduced from combustion of fossil

(a) A large amount of iodine apparently is introduced in burning fossil fuel, Miyake estimates $5\text{E}6$ kg/yr. This is ignored here since the time period involved, ~ 200 yr, is insignificant in geologic time.

FIGURE 1. Global Iodine Model (Kocher 1979). Pathways indicated by dashed lines added in the present report.

TABLE 1. Distribution of Natural Iodine(a)

Compartment	Volume, m ³	Concentration, g/m ³	Total I, g	Residence Time, yr	Notes
Atmosphere	3.8E18 (STP)	~4E-8	1.2E11	~0.06	(b)
Ocean, 100 m mixed layer	3.8E16	6E-2	2.2E15	20	(c)
Biosphere	1E12	0.1	1E11	20	(d)
Fresh water, lakes	5E14	3E-3	1.5E12	?, variable	(e)
Rivers	5E12	2E-3	1E10	<0.1, highly variable	
Deep ocean	1.3E18	6E-2	8E16	1E3-5E5	(f)
Soil, 1 m depth	1.5E14	10	1.5E15	1E4	(g)
Subsurface, 1 km depth	1.5E17	0.8	1.2E17	1E5-1E7	(h)

(a) Physical data (Krauskop 1979).

(b) Concentrations highly variable over land, much higher over oceans.

(c) Mixing time of the order of 10 yr (Suess 1969, Michel 1975).

(d) Assumes density, $\rho = 1$ at 0.1 ppm (Kocher 1979).

(e) About 1% in rivers, 99% in lakes, river flow is 3E19 g/yr, rainfall over land ~1E20 g/yr.

(f) Lower value is for transfer to ocean mixed layer. Higher one is for deposition in ocean sediments, not accurately known. Other models give longer times. Mixing time of the order of 1E3 yr (Arnold 1959).

(g) Assumes $\rho = 2$ at 5 ppm (Kocher 1979). Bruner's value for total iodine is a factor of 4 higher (Bruner 1963).

(h) Assumes $\rho = 2.5$ at 3 ppm.

fuels, ~5E9 g/yr (Miyake 1963) and in nitrate deposits. At 0.2% iodine (Mellor 1946) about 3E16 g exist in Chilean nitrate deposits.

There is some question as to what fraction of the subsurface iodine is in contact with the biosphere (through surface and subsurface water movement); perhaps this is no more than 1E15 g. Of the iodine that is in steady-state flow, about 2% then is in the soil, 2% in the subsoil, 2 to 3% in the mixed ocean layer, and the remainder in the deep ocean. This series of compartments with which man interacts will be termed the system. A different division that

considers the ocean sediments would lower the first three percentage values by an order of magnitude or more. Including a portion of the latter iodine in the system makes the estimated amount of iodine involved about $2 \pm 1 \times 10^{17}$ g. Figure 2 provides a graphic representation of natural iodine distribution.

Since iodine is transported largely through the water system, information on the water distribution is also needed. Table 2 shows the pertinent data.

DISTRIBUTION OF IODINE-129

Of the iodine isotopes 115 through 142, eleven have half-lives greater than 1 hr. These are listed in Table 3 along with the yields from thermal fission (Walker 1977). Iodine-129 is not produced by direct fission but grows in through the ^{129}I in chain. The decay rates are such that after a few years the fission product iodine is about 86% ^{129}I and the remainder ^{127}I . Virtually

FIGURE 2. Distribution of Natural Iodine

TABLE 2. Global Water (Franks 1972)

Distribution	
Location	Quantity, m ³
Ocean	1.3E18
Atmosphere	1.3E13 (stratosphere 5.0E6)
Soil	2.5E13
Lakes	1.2E14
Rivers	1.2E12
Lower ground water	4.1E15
Glaciers	2.1E14
Polar ice caps	2.7E16
Rate of Movement, m ³ /yr	
Turnover, global	3.5E14
Rivers, total	3.4E13
Rivers, U.S.	1.5E12
Rainfall, U.S.	5.9E12 (0.75 m/yr)

TABLE 3. Thermal Fission Yields of Iodine (Walker 1977)

Mass No.	Half-Life	Fission Yield, %	
		²³⁵ _U	²³⁹ _{Pu}
123	13.2 h	0.016	0.05
124	4.17 d	0.020	0.06
125	59.7 d	0.029	0.11
126	13 d	0.06	0.20
127	stable	0.12	0.5
129	1.59E7 yr	0.72	1.5
130	12.36 h	1.77	2.4
131	8.041 d	2.89	3.87
132	2.29 h	4.30	5.41
133	20.8 h	6.7	7.03
135	6.585 h	6.5	7.4

all of this is released from the fuel during reprocessing, where it is diluted with a small but unknown amount of ^{127}I occurring as impurity in process chemicals. It is at this point that releases to the environment may occur. Historically such releases have been predominantly into the atmosphere. At some plants, smaller but significant amounts have been released to streams or to the water table.

The natural sources of ^{129}I are primarily natural fission, and the interaction of high-energy particles in the upper atmosphere with xenon. Input to the mobile reservoir includes: atmospheric production, weathering of rocks, fission in the hydrosphere, volcanic action, and extraterrestrial matter. Anthropogenic sources are: nuclear power, nuclear weapons production and nuclear testing. For the natural sources Kohman and Edwards arrive at the values in Table 4 (Kohman).^(a) The inventory is given by the product of the rate and the mean lifetime,^(b) which for ^{129}I is $2.4\text{E}7$ yr.

If we use the inventory figure for ^{127}I just estimated, the $\text{I } 129/127$ ratio is $1\text{E}-12$. Allowing more ^{127}I into the reservoir by considering the time period of 24 million years for the average life of ^{129}I , i.e., inclusion

TABLE 4. ^{129}I Sources

Source	Rate, g/yr	Inventory, g
Upper atmosphere	$5.1\text{E}-3$	$1.2\text{E}5$
Volcanic action	$2.0\text{E}-3$	$4.8\text{E}4$
Rock weathering	$5.2\text{E}-4$	$7.6\text{E}3$
Meteorites	$7.8\text{E}-5$	$1.9\text{E}-3$
Ocean Fission	$2.1\text{E}-5$	$5.1\text{E}2$
Total		$2\text{E}5$ (34 Ci)

(a) No data were found for the ^{129}I contribution from combustion of fossil fuel. The content would presumably depend on the U-Th content since ^{129}I present in the original source would have decayed.

(b) The mean lifetime is defined as the reciprocal of the decay constant,

$$t_{\text{mean}} = \frac{1}{k} = \frac{1}{\ln 2} t_{1/2}$$

of some iodine from ocean sediment, might reduce this an order of magnitude. On the other hand, analyses of iodine reagents from the prenuclear era, salt-brines, pre-1936 thyroids, etc., give a ratio of about $1\text{E}-11$ to $3\text{E}-13$ (Brauer 1973b) in reasonable agreement with the first estimate. Soldat (1976) quotes references listing the natural abundance ratio in the range $1\text{E}-15$ to $1\text{E}-12$. An early prediction based on the ^{238}U content of sea water was $3.5\text{E}-14$ (Edwards 1962). Analysis of AgI deposits gave $(2.2-3.3\text{E}-15)$ (Srinivasan 1971). Further determinations are needed to fix more accurately present and past variations.

To obtain the total ^{129}I inventory, the contributions from nuclear power generation, plutonium production, and weapons testing must be added. Russell and Hahn in 1971 estimated that 10^4 Ci of ^{129}I would be produced from power generation by the year 2000 (Russell 1971). The yield, about 1.25 Ci/GWe-yr, varies with fuel and reactor type. To date, the contribution is small since reprocessing of power reactor fuel has been limited. The iodine in unreprocessed spent fuel is not included in this present system inventory. The actual contribution from nuclear power might have doubled that from the natural inventory. A small amount has been added by nuclear weapons testing. Haury estimated 12 Ci in 1973 (Haury 1973). The amount added through 1964 would be included in the estimate of Ehalt that was based on the ^{85}Kr atmospheric level. About 100 t of ^{235}U had been fissioned (Ehalt 1964). This would be equivalent to about 80 Ci of ^{129}I . However, the largest contribution is most certainly associated with worldwide military plutonium production and separations.

A present total inventory of 300 to 1000 Ci is not unlikely, $\sim(2-6)\text{E}6$ g ^{129}I . Thus, the averaged present $^{129}/^{127}$ ratio could be as high as $3\text{E}-11$ or possibly as low as $1\text{E}-12$.

The ratio as well as the inventory is difficult to fix because the iodine released to the atmosphere is still highly localized and the actual amount (ratio) observed is highly variable. The early studies of Edwards and the extensive studies of Brauer and coworkers indicate a much higher ^{129}I concentration than the above numbers (Edwards 1967), Brauer 1973a, 1973b, 1974, 1978). Disregarding samples from the immediate vicinity of nuclear processing

facilities, this ratio still covers an appreciable range; $\sim 2\text{E}-10$ for seaweed, $\sim 3\text{E}-9$ for silt, $\sim 1\text{E}-8$ for soils, and $\sim (1-2)\text{E}-7$ for air and fresh water. Air samples show roughly equal amounts of gaseous and particulate forms. Air samples from above the ocean are slightly lower, $\sim 1\text{E}-8$, and suggest that the mixed ocean layer may also be about $1\text{E}-8$. No deep ocean sampling has been reported.

These numbers are 10^2 to 10^4 times that calculated from the estimated 127 and 129 inventories. The data seem to indicate no appreciable change in the ratio over the past 10 to 20 years. More data are needed to determine the changes with time in the different compartments.

This large numerical factor does have significance with respect to the types of release of ^{129}I previously existing. Most of the ^{129}I produced in the U.S. in the last 35 years was probably released to the atmosphere, from which it then entered the iodine cycle. During the relatively short operating life of the Nuclear Fuels Services (NFS) plant a somewhat larger fraction was apparently introduced to the aqueous environment. The situation with respect to foreign plants is less certain. Although the doses resulting from the present level are not significant, the fact that the concentrations of ^{129}I remain high in the environment suggest that atmospheric disposal is not the prudent long-range method. Iodine-129 introduced to the land atmosphere rapidly enters the biosphere and the soil. In the latter the residence time is thousands of years. High concentrations found in the nonliving parts of plants and in the very top soil layers support this conclusion.

The final numbers we wish to compare in this section are MPC values and the concentrations listed in Table 1. Table 5 lists these data. There appears to be no problem accommodating the ^{129}I produced by the nuclear industry; it is a matter of distribution.

DERIVED STANDARDS

At present, criteria for fixation and storage or ultimate disposal of ^{129}I must be established on the basis of guidelines that establish radiation dose rates for individuals in the general public and release rates of ^{129}I to the environment.

TABLE 5. Comparison of Existing ^{129}I Concentrations
with MPC Values

Location	Concentration ^{127}I , g/m ³	Approximate ^{129}I Concentrations	
		Ambient, g/m ³	MPC, (a) g/m ³
Atmosphere	3E-8	3E-15	1.2E-7
Rivers	2E-3	2E-10	3.4E-4
Ocean	6E-2	6E-10	--

(a) Uncontrolled area MPCs for air and drinking water, respectively, given in Table II, Appendix B, 10 CFR 20 and converted to units of g/m³.

Recently the EPA recommended (40 CFR 190) radiation protection standards for limiting the exposure of the public from uranium fuel cycle operations. These standards are summarized below:

- (a) The annual dose equivalent to any member of the general public from effluents or radiation from uranium fuel cycle operations shall not exceed 75 millirem to the thyroid and 25 millirem to any other organ (excluding skin and cornea).
- (b) The total quantity of radioactive material discharged to the environment per gigawatt-year of electricity generated shall contain less than 50,000 curies of krypton-85, 5 millicuries of iodine-129, and 0.5 millicurie combined of plutonium-239 and other alpha-emitting transuranic radionuclides with half lives greater than one year.

The effective date for part (a) of these standards is December 1, 1979, for all operations except milling of uranium ore, for which the effective date is December 1980. Part (b) of these standards was effective December 1, 1979 except that the standard for release of ^{85}Kr and ^{129}I shall be effective January 1, 1983, for any such material generated by the fission process after these dates.

In addition to the EPA recommended release limits, we have the standards for radiation protection (10 CFR 20). The uncontrolled area MPC for air is

$2\text{E}-11 \text{ Ci/m}^3$ and for drinking water $6\text{E}-8 \text{ Ci/m}^3$. These numbers are not criteria but are examples of derived standards. A valid and useful set of criteria should lead to logical development of such standards. It seems reasonable to expect that a decontamination factor (DF) of 10^3 for ^{129}I can be reached at the fuel reprocessing plant (FRP). This DF is a factor of four better than is implied by the EPA release limit of $5\text{E}-3 \text{ Ci/GWe-yr}$.

The radiation dose standard (item a) can be met at an FRP of 1500 t/yr capacity with the projected DF of 10^3 for ^{129}I and the associated release rate of 0.06 Ci/yr . Dose calculations have been carried out in detail by several workers (e.g., Russell 1971, Palms 1975, Soldat 1976, Book 1977). The critical organ is the thyroid, which contains about 20% of the iodine in the human body. The amounts and concentrations vary with the age of the individual. These are shown in Table 6 along with the half-life for residence in the thyroid.

Soldat calculates that the maximum permissible dose rate to the public (individual) of 1.5 rem/yr would be reached with a concentration of $1.3\text{E}-9 \text{ Ci/g}$ thyroid. The 75 mrem/yr EPA recommended rate would correspond to $6.5\text{E}-11 \text{ Ci/g}$ thyroid. From the data of Table 6 it is seen that this latter dose would be produced by a $^{129}\text{I}/^{127}\text{I}$ ratio in the thyroid of $1\text{E}-6$. It will be recalled that the maximum ratios found in the environment (air and water) away from reprocessing facilities are about $1\text{E}-7$. The radiation dose rate to the thyroid of an infant residing 1.5 miles from a generic FRP in the predominant wind direction was calculated for an NRC study of fuel cycle facility siting (Harty 1976). Assuming a DF of only 20 for ^{129}I and 40 for ^{131}I and including the contribution from other radionuclides such as ^3H , the total infant

TABLE 6. Iodine Content of Thyroid(a)

Age, yr	Weight of Iodine, (a) mg	Mass of Thyroid, g	Effective Half-Life, days
1	0.18	2.0	20
4	0.90	5.0	20
14	4.2	15	50
Adult	7.0	20	100

thyroid dose rate was approximately 70 mrem/yr. Changing the DFs for both ^{129}I and ^{131}I to 10^3 would lower this total thyroid dose to 5 mrem/yr, well below the EPA dose guide level of 75 mrem/yr. A further improvement in the DF for ^{129}I would not significantly reduce the thyroid dose rate because of the contribution by ^3H (3 mrem/yr).

The restrictions on release rates (item b) were designed to limit the long-term collective radiation dose to the world's population. Compliance with this part of the standard can be accomplished by restricting the combined release rates from all the facilities associated with generation of nuclear power. For ^{129}I the only facilities of concern would be the fuel reprocessing plant, the high-level liquid waste solidification equipment, and the process for disposal of the ^{129}I collected during these two processes. Assuming a DF of 10^3 at the FRP leaves a maximum of about $3.8\text{E-}3$ Ci/GWe-yr that can be released to the environment during fixation and storage (or disposal). Optimistically, losses in the fixation process can be included as part of the 10^3 DF of the FRP. Storage for an arbitrary 10^3 yr then requires a fractional release rate of $3.1\text{E-}6$ fraction per year. It must be noted that this DF has not been demonstrated and will require careful process controls to minimize the iodine in the organic solvent, high-level waste, etc.

CRITERIA DEVELOPMENT

The significance of these residence times, distributions, and concentrations with respect to the factors involved in determining storage and disposal criteria must now be addressed. Following is a list of factors previously discussed. They are shown in expanded form as being essentially technological, economic, environmental, or sociopolitical. The factors are not necessarily complete. They could also be described in much greater detail--or condensed.

Most of the factors are general and have about the same importance regardless of the storage or disposal option. Items 1, 2, and 5, and the economic factors, 6, 7, and 8, fall in this class. The impact of these is obvious and needs no further comment here. Items 9 and 10 are basic to waste management and are important at all steps and with all options. The remaining factors

may require different degrees of consideration for different options. The following factors are involved in determining the criteria for ^{129}I disposal:

Technological

1. Acceptability of secondary wastes from process
2. Applicability to all parts of fuel cycle
3. Presence or absence of other radioactivity
4. Compatibility with environmental conditions encountered during processing, storage and transportation (including accidents)
 - a. Packaging
 - b. Thermal characteristics
 - c. Frangibility
 - d. Solubility
 - e. Interaction with other chemical species
5. Reliability, safety, and simplicity of process

Economic

6. Monetary cost
7. Material resources and energy expended
8. Manpower commitment

Environmental

9. Capacity of the environment and distribution of ^{129}I
10. Health impact resulting from control
11. Presence or absence of other hazardous materials

Sociopolitical

12. Adaptability to a changeable social and political environment
 - a. Stability of institutions
 - b. Public perception
 - c. Impact of future technology
13. International agreements
14. Radioactive half-life same order of magnitude as that of major geologic changes.

TEMPORARY STORAGE AND TRANSPORTATION

Temporary storage and transportation include the waste management separation step and preliminary fixation. Items 9 and 10 are fundamental, and existing derived or concentration limits (40 CFR 190) must be considered. These place special significance on item 4. Items 3 and 11 may also influence both storage and transportation. Item 12 is important and will be difficult to quantify. Item 13 must be considered, but it will be more significant in other steps in the disposal process. Item 14 is not applicable at this step.

Iodine-129 is a low-specific-activity radioactive material, and shipping regulations are currently limited to items such as proper labeling and shipping in sole-use vehicles. Transportation of large quantities should require additional consideration. The factors listed under item 4, including some detail as to temperature and time limits, would seem to be important.

DISPOSAL TO A DRY ENVIRONMENT

There is no way to predict the ultimate stability and hence the lifetime of a specific storage form over very long periods, e.g., $>10^5$ yr. For example, cuprous iodide is a naturally occurring mineral--but only by chance. The compound is very unstable in the presence of water and oxygen. At 10^6 yr or greater, the impact of major geologic changes must be considered. Thus application of any derived standards from items 9 and 10 will require a time limitation, perhaps 10^3 yr. This interacts with item 12. Beyond this period, item 14 becomes increasingly important, and it must be assumed that the material will be exposed to water and/or high temperatures. There is some reason, however, to think that secondary and tertiary barrier materials could have some effect on long-period geochemistry, and the details of item 4 should again be considered.

DISPOSAL TO A WET ENVIRONMENT, LAND

Disposal to wet environment may be accidental or deliberate. After the containment vessel is gone, the iodine-containing matrix, for example concrete, will lose iodine by erosion and diffusion. Studies with iodates of barium,

strontium, and lead have shown that leach rates of the order of $1\text{E-}4$ cm/day or possibly as low as $1\text{E-}6$ /day can be achieved (Clark 1977, Morgan 1978).

The higher rate applied to a cylinder, 0.3 m radius by 1 m would correspond to an iodine loss rate of $3\text{E-}3 \text{ yr}^{-1}$ or 0.3% per year. Even at a factor of 100 lower, the lifetime of the material is short for the time intervals in question and a factor of 10 faster than the arbitrary limit given on p. 15. However, other factors may reduce the loss rate. Leaching from the matrix may be greatly reduced by precipitation of films on the surface. Effective backfill chemicals may sufficiently reduce the iodine concentration that transport to the environment is orders of magnitude lower.^(a) Thus the details of item 4, which minimize ^{129}I content of the water or minimize the $^{129}\text{I}/^{127}\text{I}$ ratio, are again important.

The presence of other radioactivity, item 3, or toxic materials, item 11, complicate the problems. They must be treated in the same manner. Item 12 is significant. Item 13 must also be considered. Even though storage is within national boundaries, the half-life of ^{129}I and the potential larger dispersal area make disposal an international question.

Deliberate exposure to water could be made in such a way as to satisfy items 9 and 10. The data of Tables 1, 2, and 3, along with $^{129}/^{127}$ ratios observed in the environment, indicate the possibilities. An engineered leakage to a sizable river could even be preferable to an attempt at guaranteed zero release. The reasons, of course, are the large amount of ^{127}I present in water, and the rapidity of water turnover in the environment. The flow rate of many rivers is sufficiently adequate that disposal of large amounts of iodine to the sea via a solubility-controlled rate or a monitored release is conceivable.

For example, a 1500 t/yr processing plant would produce about 55 Ci or $3.4\text{E}5$ g/yr. If diluted with the average flow of the Columbia River, the ^{129}I concentration would be somewhat less than 1% of the MPC while the $^{129}/^{127}$ ratio

- (a) These data are supported in a PNL report in preparation by Wiemers and Burger.
(b) The usual assumption that iodine, being soluble, moves rapidly through all soils cannot be justified by the basic chemistry of iodine. In fact, its reactivity and multiple valence states suggest that rather slow movement might often be found.

would be about $1\text{E}-3$. As opposed to effects of atmospheric releases, there is no evidence that release to rivers produces a buildup in river sediments or plant life. However, the data are not very extensive. The $129/127$ ratio in the present example may be uncomfortably high. There is also the problem of adequate dispersion at the dilution site, and the possibility of reaction with other discharges to the river. On the favorable side is the short residence time, days, before further dilution in the ocean. In any case, items 3, 4, 11, 12, 13, and 14 are each important. The concept does appear to be in conflict with the present release limits if the latter are taken literally.^(a)

DISPOSAL TO A WET ENVIRONMENT, OCEAN

If the dilution option is taken, direct disposal to the ocean is preferable to the previous approach. The water volume and the iodine content guarantee a low ^{129}I concentration and a low $129/127$ ratio. The equivalent ocean volume would be about 100 m deep and the mixing time of this ocean layer is about 10 to 20 yr.^(b) However, on a local scale it is highly variable and lateral movement of water may range from 0.02 to 0.5 m/sec (Hollister 1977).

If ocean disposal is considered, then the data of Tables 1 and 2 indicate that the deep ocean is the obvious choice. In addition to the dilution factor, the residence time, 10^3 yr, is adequate to ensure dilution of the iodine before return to the environment. This is a very important point, and there appears to be no question of its validity regardless of the model used.

It was mentioned earlier that Kocher's model, Figure 1, does not show return of iodine to the lithosphere via ocean sediments. Actually, this process is significant in a time span of 10^6 to 10^7 yr, and sediments in oceanic trenches are constantly being overridden by continental plates. Much of the iodine so disposed of will decay before again reaching the environment. The ultimate disposal technique then is to place wastes in holes drilled in these areas (Macbeth 1978). For ^{129}I , however, the costly engineering work

(a) The 5 mCi/GWe-yr assumes release to the atmosphere. Releases to other compartments must be re-evaluated.

(b) Uniform dilution from a hypothetical point source requires a much longer time.

associated with drilling, filling, and capping does not seem necessary. Incorporation of iodine, preferably as the iodate, in the cement and deposition in ocean areas of reasonable sediment thickness would produce additional diffusional delay and guarantee adequate mixing of the fraction escaping to the ocean. (Sediments range from a few tens of meters in thickness to as much as 2 km.) The high cost normally associated with sea bed disposal can be avoided.

Even this degree of care may be unnecessary. Dilution of the 10^4 Ci (5.7×10^7 g) of ^{129}I that may be produced by the year 2000 (Russell 1971) by the deep ocean would produce a concentration of 8×10^{-15} Ci/m³ or 4.6×10^{-11} g/m³, roughly 7 orders of magnitude below the drinking water MPC. Adequate mixing is guaranteed by the residence time.

This option is influenced by items 12 and 14. In addition, criteria would need to be developed for items 4, 9, 10, and 13.

CONCLUSIONS

A set of general factors has been expanded into 14 basic items that can be used in setting criteria for storage and disposal of ^{129}I . The various disposal options have been considered with regard to the present distribution of iodine and its concentration in various compartments of a world model. The transfer rates and residence times and the accessibility to the biosphere indicate that the different disposal options will require different degrees of consideration of the 14 items in setting criteria. No attempt has been made to derive standards from these items.

Table 7 lists examples of measurements or evaluations which could be applied to the different factors. These are by no means complete and not necessarily the most important.

The fundamental issue is the control of radiation dose to the individual and to the world population. The capacity of the environment, or more exactly, the capacity of the various compartments of the world environment to accommodate the ^{129}I produced is basic to disposal decisions. There appears to be no problem in this regard. Dilution to acceptable limits with respect

TABLE 7. Examples of Measurements for Criteria

<u>Factor</u>	<u>Measurement</u>
<u>Technological</u>	
1. Secondary wastes	Secondary wastes should be capable of assimilation in normal plant processes
2. All parts of fuel cycle	Control complexity
3. Other radioactivity	Concentration or radiation limits Specific nuclides
4. Compatibility with environmental conditions	Physical characteristics of container, durability Time-temperature of package Solubility of fixation material
5. Reliability	Failure probability Type of maintenance required Capacity for substitution or bypass Technology demonstrated?
<u>Economic</u>	
6. Monetary cost	\$/GWe-yr, capital versus operating costs, space
7. Material resources and energy	Use of strategic materials, fraction required
8. Manpower	---
<u>Environmental</u>	
9. Capacity of environment	Concentration of ¹²⁹ I in global compartments ¹²⁹ I/ ¹²⁷ I ratio Residence time
10. Health impact	Dose calculations for different paths for individuals and populations Risk determinations for individuals and populations
11. Hazardous materials	MPCs and dilutions required for poisonous chemicals
<u>Sociopolitical</u>	
12. Changing social-political environment	Risk analysis of steps and for overall process compared to that for other common activities
13. International	---
14. $t_{1/2} \geq$ geologic periods	Probable geochemistry considered Release calculations limited to 10^4 yr

to both concentration of ^{129}I and the $^{129}/^{127}$ ratio can easily be achieved. Slow release to the environment of the ^{129}I produced by the nuclear industry can be permissible. For utilizing the dilution principle, it is preferable to package and dispose of ^{129}I separately from other fission products.

It is suggested that, based on the factors discussed here and perhaps others, a set of preliminary standards be developed to apply to 1) temporary storage and shipping, 2) disposal to a dry environment, with a time limitation on calculated behavior, and 3) disposal to the deep ocean, with complete release permitted in 10^3 yr. Of the two disposal options, ocean disposal is preferred as having a negligible impact on the environment.

Early quantification of some of the factors listed on p. 21 will permit better decisions on what research is essential for iodine removal, fixation, and disposal, and may well suggest that more attention should be directed to migration and to the soil chemistry of iodine.

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