

The Legacy of Radioactive Waste from the Weapons Laboratories:

Haw the Render the Radioactive Waste Harmless?

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1. Introduction

One of the unresolved problems after the end of the Cold War concerns the leftover of huge amounts of radioactive material and waste, mainly originating from the fabrication of weapon-grade material by the two super powers. In addition there is an ever increasing amount of waste coming from the civil nuclear sector. Apart from the need to address the question from a technical side, there remains the task to inform properly the public, and to get a general consensus what to do with this waste.

Politics consisted often in secrecy and covering up dangerous situations and procedures. Only slowly data are being released by the military/industrial complexes and nuclear power companies of East and West, mostly under constant pressure from environmental groups and non-governmental organisations. This reluctance by the responsible governmental branches prevented a timely investigation of the effects of radiation fallout, in particular the determination of an acceptable tolerance level for different kinds, their quantity, and duration of exposure. Health can be affected through physical contact, via the food chain, or by inhalation of radioactive material. When huge amounts of radioactivity were involved, this secrecy had detrimental consequences, costing the lives of innocent people, who were not warned or aware of dangers. Preventive or early medical care could have reduced the number of cancers. On the other end of the scale, billions of dollars were wasted in protection against minute radiation doses, which might even be beneficial for humans.

It is the purpose of this paper to try to assess the waste problem in a qualitative and quantitative way, to remove some of the unjustified fear of the public, but also clarify and find solutions for the remaining big problems. An attempt will be made to compare composition and amount of radioactive waste produced by the weapon complexes with the one originating from the civil nuclear sector. The general public is often worried about the impact of waste on the health of future generations. Therefore the *half life* of radioactive material has to be addressed together with technologies of transmutation of longer-lived isotopes. The majority of people is not familiar with the definition, that any isotope is called radioactive if its half-life is shorter than the age of the Earth.

Many technical efforts are being made for rendering nuclear waste harmless, both for population and environment, or at least reduce any danger. Some of them are promising and will be briefly described. All these efforts demand not only considerable funding, but also the involvement of qualified people, be it scientists or technicians. Preferably, personnel should be recruited from the weapon laboratories, since those people possess already a wide range of the necessary scientific background, which now should be profitably applied to a slightly different task. This could also solve the employment problem in *cities of science*, in particular in Russia, at a time when the demand for more nuclear weapons is somewhat dwindling.

2. What is waste?

By glancing superficially at this question everybody would seem to have a ready answer. However, thinking seriously about the subject, a definition of waste is not simple at all. When preparing this paper on radioactive waste a sentence came to mind, which I heard from a professor in my first physics course: "Dirt is matter in the wrong place". During the workshop I learned that this definition is attributed, in this or a similar formulation, to the great Russian scientist Mikhail Vasilyevich Lomonosov (1711 - 1765). Now, giving more thought to a refinement of this definition, numerous other, qualifying characteristics for dirt appear appropriate:

- (1) Dirt is matter in the wrong place if and when it shows up in **excessive quantities**.
- (2) Dirt is matter appearing at an **unwanted place** at an **inconvenient time**.
- (3) Dirt is matter **treated** (or analysed) **in the wrong way**.
- (4) Dirt is matter **stored** at a place **for the wrong purpose**.
- (5) Dirt is matter that will become increasingly **harmful with time**.
- (6) Dirt is matter in the wrong place **for a certain time only**, but loosing its detrimental characteristics by disintegration or being moved/washed away.
- (7) Dirt is matter in the wrong place **only when it is expensive or difficult to handle**.
- (8) Dirt is matter in the wrong place when it is **disposed deliberately** into a clean/different environment.
- (9) Dirt is matter which appears to be **useless**.
And using an antonym statement:
- (10) Matter **is not considered as dirt** according to the above definitions, when there is an option to **transform it into something useful**.

Other extensions of the above are possible. The presence of dirt may or may not require any **action**. A decision may be based upon a qualitative judgement: is dirt considered as something only **unpleasant**, is it **annoying, disturbing, detrimental**, or even **dangerous**, becoming a **threat** to health?

What happens if the word "*dirt*" is replaced in the definitions by "*radioactive material*" ? In most cases the information on the properties of material would be applicable again and not need to be modified.

Difficulties start when going a step further in replacing "*dirt*" by "*radioactive waste*". This imposes already an *a priori* quality on the matter under discussion. When the public at large is consulted on its opinion on radioactive waste, the reaction is expected to range from hesitant to aggressive. People are irritated, since they can not **feel, smell** or **taste** radioactivity.

They either do not remember or ever had learned what radioactivity is, or if they had, they may not have a quantitative conception of what a half-life of an isotope means. Scientific terms may have been explained incompletely or were taught not considering their limited background knowledge. We will get very diverging views on radioactivity and its possible impact from the interested, but not necessarily scientifically well-educated public, often depending on how the question is asked and in which terms the underlying facts are explained. A statement would/could be:

Radioactivity is so terrible, it has to be banned from Earth

A reaction to a question, posed to fifty high school students in Idaho Falls/USA, who had studied chemistry, a city where many of their parents work in the Idaho Nuclear Engineering Company, may illustrate the above statement:

"It's in Your Own Home And It's a Real Killer" [1]:

The chemical compound dihydrogen monoxide, or DHMO, has been implicated in the deaths of thousands of Americans every year, mainly through accidental ingestion. In gaseous form, it can cause severe burns. And according to a new report, *"the dangers of this chemical do not end there."*

The chemical is so caustic that it *"accelerates the corrosion and rusting of many metals ... is a major component in acid rain and ... has been found in excised tumours of terminal cancer patients."* Symptoms of ingestion include *"excessive sweating and urination,"* and *"for those who have developed a dependency on DHMO, complete withdrawal means certain death."*

Yet the presence of the chemical has been confirmed in every river, stream, lake and reservoir in America.

Judging from these facts, do you think dihydrogen monoxide should be banned?

40. percent of fourteen year old students voted to ban dihydrogen monoxide because it has caused too many deaths.

The reaction would worsen if one replaces hydrogen by one of its heavy isotopes, by deuterium or, even more worrisome, by radioactive tritium, which is actually the only dangerous of them. It would be a surprise if we get a much different answer from our adult politicians.

In order to treat adequately the existing problem of radioactive waste we have to rely, at least at present, on the judgement of scientists. But even with their help this can not be done in a comprehensive, completely satisfying way. There are still scientific disputes on the uncertainty of (i) the biologic impact of dose rate and the linear no-threshold theory (LNT) . There is (ii) the double use problem of scientific discoveries: fissionable or radioactive material for peaceful purposes (nuclear power reactors, medicine, material testing, etc.) on one hand, and of material for the stewardship of nuclear weapons and their improvements on the other. And there are (iii) various economical interests at play. These are just a few of the controversial aspects which enter any discussion. Therefore, the aim of the following chapters is to bring some coherence into the evaluation, provide information for convergence of ideas, and to reduce the many definitions to hopefully only a few.

3. Radioactivity in nature, radioactive waste from civil and military sector: A semi-quantitative assessment

Radioactivity is distributed unevenly over the crust of the Earth and in the atmosphere of our planet. Our species, all animals and plants have learnt to adapt, to live and to develop with it. Radioactivity becomes only a threat to humans (animals, plants), if it is too highly concentrated locally through human activities. We are all exposed to natural radioactivity, which varies with location on Earth (soil) and altitude (atmosphere). There is $\geq 5'000$ Becquerel inside each adult, and ~ 3 cosmic rays / cm^2 traverse our body every minute, and every day at least a billion particles of natural radiation enter our bodies. There is a natural heat flux of $0.06 \text{ W/cm}^2 = 30 \text{ TW} \approx 0.02\%$ of radiation from our sun (use of charcoal, oil is 0.01% of Sun). And there is heat coming from the inside of the globe, originating partially from the decay of uranium and thorium, producing $25 \text{ J/m}^3/\text{y}$ and $80'000 \text{ Bq/m}^3$ in the rock. And there are *man-made radioactive isotopes* being used extensively during medical tests and cancer treatment, produced by particle beams or in nuclear reactors. Radioactive isotopes were produced during explosion of nuclear weapons in the atmosphere and remain still there.

3.1 Classification of waste

There are several possibilities to classify human produced/accumulated radioactivity, namely by

- (i) **State of aggregation:**
solid, liquid or gaseous states (solids and liquids will be further discussed in this article).
- (ii) **Source of production and subsequent use:**
military or civilian. However, here the dividing line is not too well defined.

Two chemical elements play a major role:

(a) Uranium:

Facilities to produce reactor-grade enrichment of U^{235} to $3 \div 4$ per cent for **civil power reactors**.

Facilities for higher enrichment ($20 \div 98$ per cent) for **Weapon production** or use in **submarine reactors**.

Separation plants for highly-enriched uranium are very large and can therefore be easily identified by surveillance satellites or other detection systems.

(b) **Plutonium** is still being categorised into *reactor-grade* ($58\% \text{ Pu}^{239}$, $24\% \text{ Pu}^{240}$, plus other isotopes) and *weapon-grade* (more than 94% of Pu^{239} , $6\% \text{ Pu}^{240}$). Weapon-grade is the preferred material for military use and exclusively used by the nuclear weapon states (NWS). There is now almost general agreement between specialists that reactor-grade plutonium could also be used for manufacturing of nuclear weapons,

its disadvantages being that it produces three times more neutrons and generates five times more heat than the weapon-grade plutonium, making handling and maintenance of warheads more laborious. Furthermore, more fissile material (about twice as much) is needed to have equivalent explosion yields.

Reactors for production of weapon-grade material and its isotope separation plants create large amounts of radioactive waste, that can be detected due to their size and release of specific isotopes like Kr^{85} .

(iii) Handling and danger for humans :

High-level waste (HLW): The highly radioactive waste that results from the reprocessing of spent fuel from nuclear reactors and from the processes in the production of nuclear weapons. HLW contains highly radioactive, short-lived fission products, hazardous chemicals and toxic heavy metals. HLW includes liquid waste produced directly in reprocessing and any solid waste derived from the liquid.

Intermediate-level waste (ILW): A category of radioactive waste used in several countries, but without a consistent definition. For example, it may or may not include transuranic waste.

Low-level waste (LLW): A catchall term for any radioactive waste that is not spent fuel, high-level waste, or transuranic waste.

Spent nuclear fuel (SNF): Fuel elements and targets that have been irradiated in a nuclear reactor. It is sometimes called high-level waste.

Transuranic waste (TRU): Waste containing alpha-emitting transuranic elements having half-lives of more than 20 years, in concentrations of more than $100\text{--}10^{-9}$ Ci per gram of waste.

SNF and TRU waste can all be quantitatively described in units of

- (a) **volume**
- (b) **decayed radioactivity.**

(iv) Responsible organisation for handling of waste

(a) In the U.S. mainly the **Department of Energy (DOE)**, or Commercial.

(b) In the Former **Soviet Union (FSU)** mainly the **Ministry of Atomic Energy (MINATOM)**, but also by the **Ministry of Defence**, **Ministry of Transport**, **State Comity of Defence Industry**, and **Ministry of Construction Industry**.

Definitions for HLW and LLW categories differ slightly between U.S. and Russia.

Table 1. Radioactive Waste Classifications in the Former Soviet Union [2]
(for definition of dimensions related to radioactivity see ref. [3])

Type	Activity Level		
Liquid			
Low -Level Intermediate-Level High-Level	$<1 \cdot 10^{-5}$ Ci/litre $\geq 1 \cdot 10^{-5} \geq 1$ Ci/litre ≥ 1 Ci/litre		
Solid Wastes Based on Dose Rate, 10 cm from Surface			
Low-Level Medium-Level Intermediate-Level High-Level	≥ 30 mr/h $30 \geq 300$ mr/h $0.3 \geq 1$ r/h ≥ 1 r/h		
Solid Waste Based on Activity			
	Group 1	Group 2	Group 3
Beta Activity, Ci/litre Alpha Activity, Ci/litre	$2 \cdot 10^{-6}$ - $1 \cdot 10^{-4}$ $2 \cdot 10^{-7}$ - $1 \cdot 10^{-5}$	$1 \cdot 10^{-4}$ - $1 \cdot 10^{-1}$ $1 \cdot 10^{-5}$ - $1 \cdot 10^{-2}$	$> 1 \cdot 10^{-1}$ $> 1 \cdot 10^{-2}$
Gaseous			
Low-Level Intermediate-Level High-Level	$\leq 3.7 \cdot 10^{-3}$ Bq/litre $> 3.7 \cdot 10^{-3} \leq 370$ Bq/litre > 370 Bq/litre	$(1 \cdot 10^{-13}$ Ci/litre) $(1 \cdot 10^{-13} \leq 1 \cdot 10^{-8}$ Ci/litre) $(\geq 1 \cdot 10^{-8}$ Ci/litre)	

When interpreting numbers given in this article these ambiguities and quantitative uncertainties should be kept in mind.

The total volume of **Commercial** and **Department of Energy (DOE)** wastes and spent fuel, given in **volume** [cubic metres], through 1992 are shown in fig. 1 [4]. Almost 30 percent is low level waste, comes from the commercial sector, including **Light Water Reactors (LWR)** Spent Fuel (SF), that is permanently discharged, together with spacing between fuel assembly cycle. The problem is the future treatment of the three major components of almost the entire quantity of waste, 70 percent being under the responsibility of DOE. Fig. 2 [4] shows the situation as far as the **total radioactivity** of Commercial and DOE wastes till 1992 is concerned. The lion's share of 96 percent is in the permanently discharged spent fuel, and only the remainder in high level waste, ~4 percent under DOE ($1.05 \cdot 10^9$ Ci), and ~1 percent Commercial. DOE has already released into the environment $2.6 \cdot 10^6$ Ci, i.e. 2.5 per mill of its waste (to be discussed in detail in the next paragraph). Volume wise the spent fuel in form of heavy metals, with by far the giant part of the radioactivity, is small and could therefore be relatively easily disposed in underground caverns. Problematic are the transuranic elements with 6 percent of volume, relatively small radioactivity, but generally isotopes with rather long half lives (table 2; for details see ref. [5]).

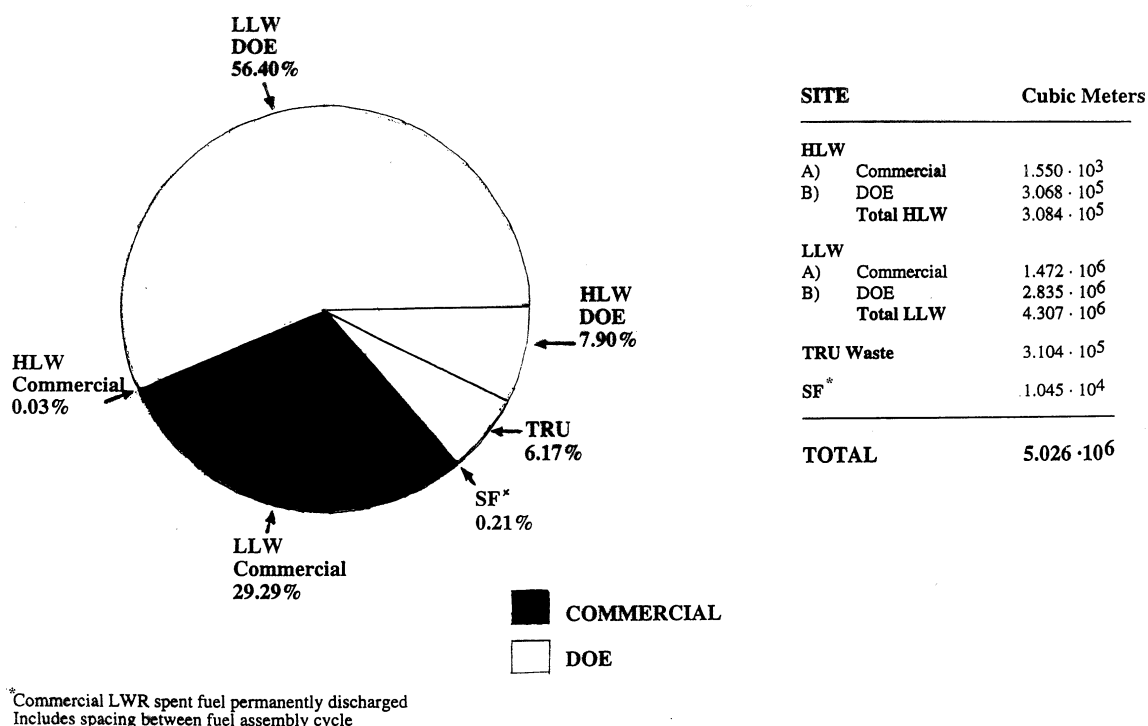
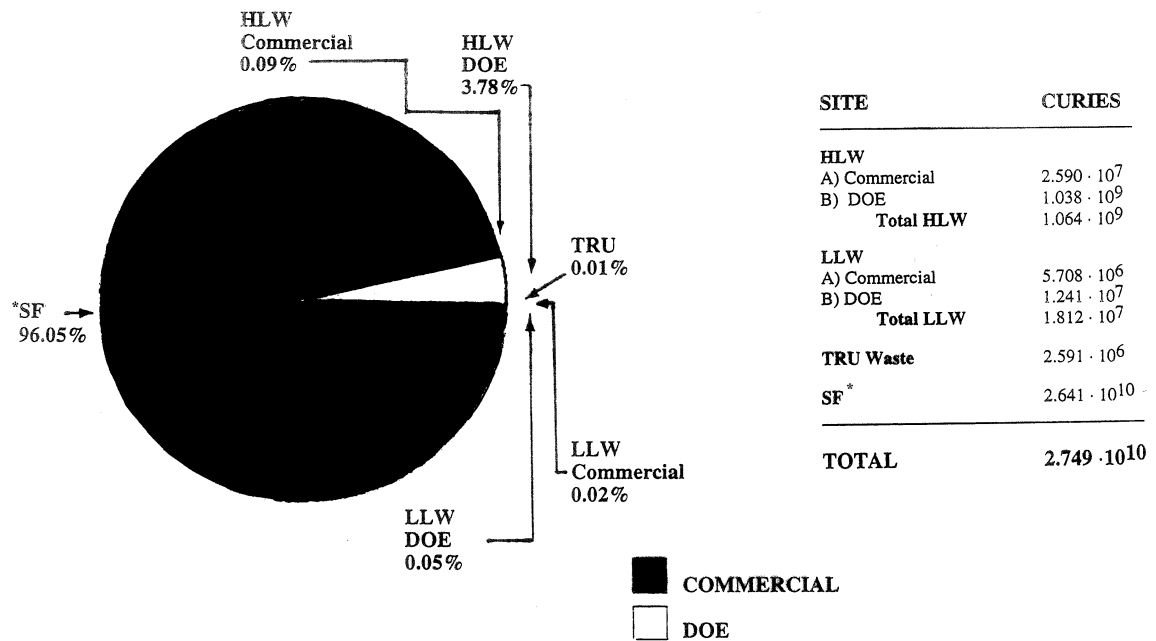


Fig.1. Total volumes of commercial and DOE wastes and spent fuel through 1992.



*Commercial LWR spent fuel permanently discharged

Fig.2. Total radioactivities of commercial and DOE wastes and spent fuel through 1992.

Table 2. Transuranic waste (retrievably stored) [5]

Site	Contact handled		Remote handled (>200 mrem)	
	Volume (m ³)	Radioactivity (10 ⁴ Ci)	Volume (m ³)	Radioactivity (10 ⁴ Ci)
Hanford	$1.15 \cdot 10^4$	15.5	273	3.15
Idaho	$3.93 \cdot 10^4$	35.0	200	0.72
Los Alamos	$1.12 \cdot 10^4$	20.2	94.1	0.05
Oak Ridge	$1.33 \cdot 10^3$	5.5	1842	9.65
Savannah River	$6.98 \cdot 10^3$	56.4	-	-
West Valley	$4.32 \cdot 10^1$	<0.01	529	0.01
Rocky Flats	$1.98 \cdot 10^3$	116.	-	-

Spent fuel increased in 1996 to $3.2 \cdot 10^4$ m³ and is expected to double by the year 2020. However, the radioactivity will increase during the next 25 years by only 10 percent, when its decay is taken into account (table 3) [5].

Table 3. Commercial spent nuclear fuel: projected amounts [5]
MTHM: Metric tons of heavy metal

Year	Mass (MTHM)	Radioactivity (Million Ci)
138.	32'200	30'200
139.	42'300	32'600
140.	61'800	39'800
141.	77'100	34'700

All waste in the U.S. combined (as of 1996) is $5.5 \cdot 10^6$ cubic meters, representing $31 \cdot 10^9$ Ci. The spent fuel under DOE as of July 1995 is 2'640 metric tons of heavy metals. Sixty percent (by volume) is high-level waste (HLW) from the weapons production, and as much as $670 \cdot 10^3$ m³ of it is in liquid form [6]. About 400'000 m³ HLW are left from the production of plutonium and HEU for nuclear weapons and naval propulsion reactors [7].

The a huge amount of **depleted uranium** is seldom mentioned as waste. There is a 560'000-metric-tonne stockpile of DOE in the U.S. in form of metal or oxide, and 199'900-metric-tonnes stored at Cogema in France. This depleted uranium consists of ~0.3% U²³⁵, with a half-life of $\tau_{1/2} = 7.13 \cdot 10^8$ years (77% α -decay, 23% spontaneous fission), and ~99.7% U²³⁸, $\tau_{1/2} = 4.51 \cdot 10^9$ years (6.7 α -decay, 93.3% spontaneous fission). It belongs into the category of LLW. Its radioactivity – when the metal is in its solid form – is for all practical, radiological considerations to be treated like the one of natural uranium ($U^{235} / U^{238} = 7 \cdot 10^{-3}$). Since its commercial value is low, it is nowadays used due to its high specific weight in penetrating anti-tank weapons (30 mm cannons). However, when pulverised upon impact it ignites by itself forming uranium oxide, which is a very poisonous chemical. It can be distributed over large areas in form of aerosols [8, 9]. The use of 55'000 grenades in the First Gulf War showed serious medical repercussions [10]. It was again used in the Kosovo War [11].

In 1999, global stocks of **plutonium** were estimated by the U.S. government [12] to be (242.3 ÷ 267.4) tons weapon-grade, (802.4 ÷ 1'037.4) tons commercial-grade, of which about 203.5 tons were separated. These data agree fairly well with those from the Nuclear Control Institute, the Institute for Science and International Security, and the Natural Resources Defence Council. Of the military total, there are roughly (140 ÷ 162) tons in Russia and the FSU, 85 tons in the U.S., and smaller amounts in France (6 ÷ 7) tons, China (1.7 ÷ 2.8) tons, the UK 7.6 tons, and in the other nuclear weapon states (less than one ton thought to be covered by IAEA safeguards). Inside weapons are 71 tons and outside 157 tons [13]. The total plutonium stock in 1994 was estimated to be 1'160 tons [14], including up to 250 tons in military stockpiles (228 tons according to [15]), 790 tons in spent fuel, and 120 tons in separated civil stocks. These figures give an indication on the rate of increase over a period of 5 years.

3.2 Quantitative overall assessment

The situation can be illustrated by two examples:

- (1) The amount of radio nuclides present in the World's Oceans is $\sim 440 \cdot 10^9$ Ci, mainly K⁴⁰ and Rb⁸⁷, and $\sim 1.7 \cdot 10^9$ Ci was added by the weapons complexes ($1'710 \cdot 10^6$ Ci from

- Russia, $2.6 \cdot 10^6$ Ci from the United States) [16]. Since this radioactive material was initially mainly released into rivers, it caused at that time an enormous local contamination, however, in a long run, when mixed with the ocean water it adds only 0.4 percent to the inventory, probably without significance for the aquatic population.
- (2) Since the Chernobyl nuclear reactor accident, the radioactivity released from it into the environment ($\sim 50 \cdot 10^6$ Ci in 1986 according to Russian statements, elsewhere are given $81 \cdot 10^6$ Ci ($\pm 50\%$) [17]) represents for the public a certain '*standard unit*', with which often justifiable or not, knowing the level of general scientific literacy – other nuclear activities or accidents are compared:

Table 4. Comparison of radioactivity released from Chernobyl with other sources in 1996 [2, 18, 19]

(Chernobyl) / (Production Reactor & Reprocessing Sites in the Former Soviet Union) = 1 575 000 Ci / 1 702 538 000 Ci = 0.093 per cent $\approx$ 1 per mill		
(Chernobyl: Cs ¹³⁷) / (World-wide atmospheric nuclear weapon tests: Cs ¹³⁷) = 795 200 Ci / 16 200 000 Ci = 0.5 per cent	$\tau_{1/2} = 30.5$ y β^- , 0.52 MeV	
(Chernobyl: Sr ⁹⁰) / (World-wide atmospheric nuclear weapon tests: Sr ⁹⁰) = 172 900 Ci / 10 500 000 Ci = 1.6 per cent	$\tau_{1/2} = 27.7$ y β^- , 0.54 MeV	
(Chernobyl: I ¹³¹) / (U.S. atmospheric nuclear weapon tests: I ¹³¹) = 7 300 000 Ci / ? Ci *) $\approx$ 10 per cent	$\tau_{1/2} = 8.05$ d β^- , 0.61 MeV	
(Chernobyl) / (U.S. Commercial spent nuclear fuel, until mid 1997) = 1 575 000 Ci / 31 700 000 000 Ci = 0.000 5 per cent = 5 per mill		

*) Exact quantity not known

These comparisons show that Chernobyl's accident - as deplorable it is on the level of human suffering, whose radiation claimed 28 lives [20], caused $\sim 1'200$ thyroid cancers over one decade, and a larger number of expected deaths by cancer (20'000 - 30'000 over a period of 50 years [21]) is not as catastrophic as the media and some opponents of nuclear energy want to convince us. It could have been avoided by following the reactor operating instructions.

In order to get a better perspective of overexposure by radiation on the population, any quantitative assessment of reactor accidents should include a comparison with other risks in life.

4. What went wrong in the past?

Public information policy on nuclear policy was insufficient and partially misleading. Blame for it can be put primarily on the military-industrial complex, but also on the civil nuclear industry, and on governments.

Our discussion will be restricted to technical and quantitative aspects related to contamination of the environment. Table 4 gave an indication where major releases of radioactivity into the environment occurred. These are identified as the production and reprocessing sites of nuclear fuel for military and for civil applications, and the atmospheric (and underground) nuclear weapons test explosions. The latter will not be further evaluated, since the atmospheric tests stopped in 1962 (comparative data on radiation release to the atmosphere are given in [22]), and values of radioactivity and their concentration at underground sites are either not well documented, or not released into the public domain.

The Manhattan project in the U.S. followed two different tracks: uranium enrichment for the cannon-type weapon and plutonium production for the implosion-type weapon, with the aim to make sure that at least one the bombs would work when needed. This weapon program was pursued with lack of attention to the budget (costs were estimated in 1941 at \$133 million, actual cost at end of World War II \$2 billion) [23] and, what is more disturbing, with little or no consideration of its environmental impact. When the Soviet Union entered the race, the neglect of these factors, now also combined with a substantial disregard of the health of people working on the bomb, was at least on the same level of negligence if not worse [24]. We know little in this respect about the programs in China, France, Great Britain, Israel, India and Pakistan. It would be a major surprise if there had not been similar indifference by the authorities and industry.

Data are slowly becoming available about radioactive waste in the U.S., published in physics journals [5, 6, 7, 25, 26, 27, 28], and for the FSU, summarised in an excellent, comprehensive study [6]. The following data are extracted essentially from these sources.

4.1 The case of U.S.

The high level waste (DOE) amounts to 100 million gallons or about 385'000 m³ (1 gallon = 3.7853 litres), corresponding to 10'000 tanker trucks. Over the years, nine plutonium-production reactors were built at Hanford/Washington. The extraction of 1 kilogram of plutonium at the plutonium and uranium extraction plant (PUREX) produces 1'300 litres of liquid high-level radioactive wastes, more than 210'000 litres of low-to-intermediate-level radioactive wastes and over 9.5 million litres of cooling water [6]. The U.S. has produced about 10⁵ kg plutonium for military purposes, corresponding to some 130'000 m³ HLW.

67 of the 177 liquid waste storage tanks at Hanford are leaking. Traces are now reported to be showing up in ground water [29]. The concern is that it could eventually reach the Columbia River. About one billion cubic metres of soil has been already contaminated by tens of millions gallon, more than 3'000 m³ originating from leaks in the tanks. 149 tanks are single shell, built between 1944 and 1966 and designed for a life-time of 25 years, containing 157·10⁶ Ci, and 28 double shell tanks, containing 111·10⁶ Ci. For comparison, about (50 ÷ 80)·10⁶ Ci are thought to have been released into the environment in May 1986 by the Chernobyl accident, including short-lived isotopes [17].

Danger of explosion of tanks

"The Hanford tanks contain a potent brew of chemicals: At the reprocessing plant the spent reactor fuel was dissolved in nitric acid, and after the processing waste to neutralise the acid and prevent corrosion of the storage tanks. ... Injected ferrocyanide and sodium titanate to precipitate cesium-137 and strontium-90, in which constant transformations occur through both chemical and radiological means, elevated temperatures provided by the decay heat. ... The threat of explosions comes from two different sources. One is the hydrogen that is accumulating in some tanks. The hydrogen appears to be generated by the radiolytic decomposition of water and some organic compounds. ... There is a viscous slurry of the bottom of one tank. Gas bubbles generate enough pressure within the slurry to raise its level. ... Suddenly the hydrogen is released in one large 'burp'. ... Pumping of liquid from single to double shell tanks, now most stuff in single shell is solid" [6].

4.2 The case of FSU

The tank problems at Hanford invite comparison with a catastrophic 1957 accident at a Soviet reprocessing plant in Kyshtym (Mayak) in the Ural: The explosion of an underground high-level waste storage tank, that contained $20 \cdot 10^6$ Ci, ejected $2 \cdot 10^6$ Ci up to 1'000 metres into the atmosphere, and the remainder was released in the vicinity of the tank. $2 \cdot 10^6$ Ci of radiation were distributed over an area of about 23'000 square kilometres [6, 30]. The remaining activity in 1997 is 44'300 Ci, almost all Sr^{90} .

In an explosion of a submarine reactor at Chazhma Bay near Vladivostock on August 10, 1985, released $7'027 \cdot 10^3$ Ci of radio nuclides, including short-lived isotopes [31]. This number can be compared with Chernobyl's major long-lived nuclides (May 1986), amounting to $5'862 \cdot 10^3$ Ci. The data after correction for decay for the two accidents amount in 1996 to $\sim 150 \cdot 10^3$ Ci, and $1'575 \cdot 10^3$ Ci, respectively.

Deliberate waste releases and those from the Mayak accident are given in table 5.

Table 5. Waste releases to the environment in USSR. Remaining activity in 1997 [32]

Tomsk-7 waste injection	~1 000 000 000 Ci
Krasnoyarsk-26 waste injection	450 000 000 Ci
Tomsk-7 reservoirs	130 000 000 Ci
Mayak reservoirs, lakes, Techa River	122 200 000 Ci
Mayak production reactors coolant water	132 000 Ci
Krasnoyarsk-26 production reactors coolant water	106 000 Ci
Tomsk-7 production reactor coolant water	37 000 Ci
Mayak - 1957 HLW tank explosion	44 300 Ci
Krasnoyarsk-26 reservoirs	>19 000 Ci
Mayak - 1967 release from Lake Karachai	~500 Ci
TOTAL	~1,702,538,800 Ci

Table 4 shows that the better known Mayak accident contributed little to the deliberate contamination. Fig. 3 displays the distribution of Russian environmental contamination with spent fuel reprocessing wastes [33].

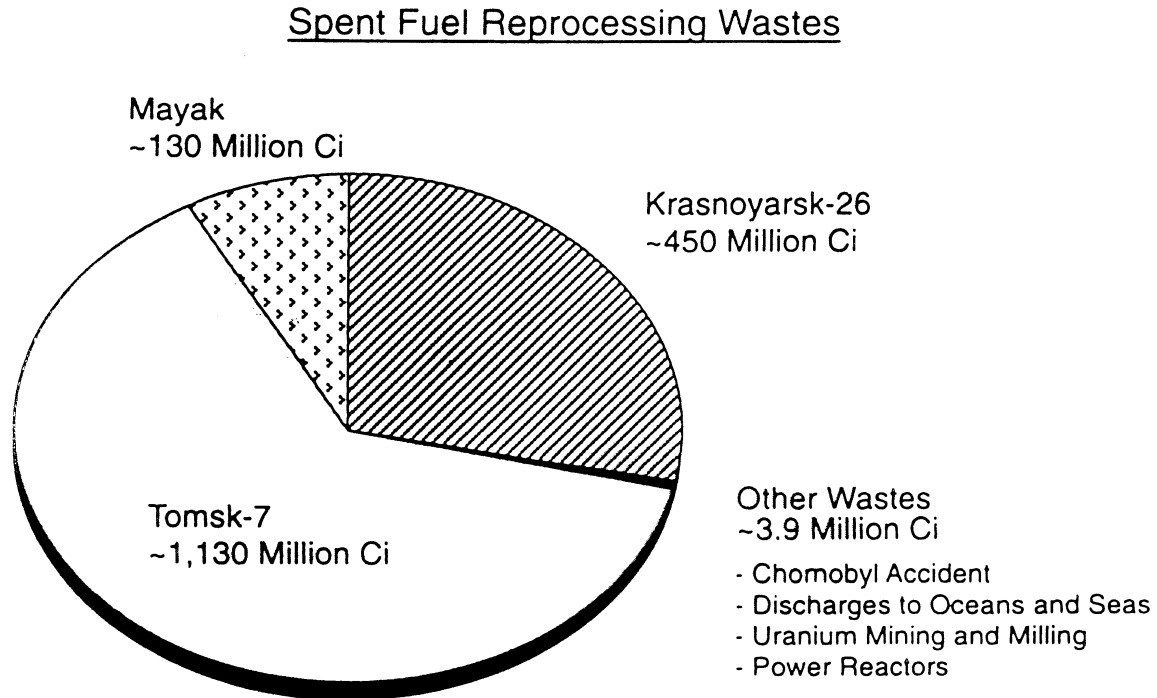


Fig.3. Spent Fuel Reprocessing Wastes: Distribution of Russian Environmental Contamination.

5. Where is the (real) danger for the future?

There will be no unbiased answer to this question. If the nuclear arsenal with its fissile material is considered part of total radioactive waste – and it should be considered as that in the author's view, since there is no direct *peaceful* use of the weapons – then it has to be put on the top of the list of potential dangers. If plutonium, produced in civil power reactors, is not under International Atomic Energy Agency (IAEA) safe-guarding, it presents a danger for proliferation of nuclear weapons, and should be put high on the list. There are technical solutions – which will not be discussed in this article – to render plutonium waste inaccessible for would-be nuclear weapon states or terrorist organisations.

Next positions are occupied by two substantial waste areas, where urgent action should be taken: these are the leftovers from construction of nuclear weapons and decommissioned nuclear-powered submarines.

In the U.S. remedies and restoration activities at Hanford/Washington should get highest priority, followed by the clean-up of the other weapon production centres. Their cost alone is estimated to be between \$189 billion and \$265 billion [26]. In comparison the building of the bomb has cost \$378 billion. In 1991 Hanford has spent \$783 million on managing high level

waste, out of a total annual budget for the site of \$1.4 billion [6]. Each sampling taken out of a tank to determine the chemical composition of the liquid costs up to \$1 million, not including the analysis itself. The danger of explosion and leakage of liquid-waste tanks has been discussed above.

The FSU had 244 nuclear submarines. Nearly 180 have now been decommissioned. Over 110 of the decommissioned subs still have operating nuclear reactors, usually two per vessel, waiting urgently to be dismantled [34]. Otherwise there is the risk of explosion of the unsufficiently or non-attended reactors. One such submarine exploded already in 1985 (see above). If all blow up, then the released radioactivity would be equivalent to ~15 *Chernobyl*.

A total of nine submarines, five diesel-powered and four nuclear-powered, sank following accidents. Five of the vessels were later recovered, four diesel-powered and one nuclear-powered, and four are still on the ocean floor. These lost subs contained five nuclear reactors and an estimated 43 nuclear warheads. At the time of sinking, the five reactors were estimated to contain 650'000 Ci of activity, and the 43 warheads 6'030 Ci [34].

6. What has been, can and should be done about radioactive waste to render it harmless?

The choice may be made between disposal in suitable, deep geologic repositories, vitrification, transmutation and separation technologies. In this chapter an attempt is being made to put some (chronological and objective) order into future activities in the field of radioactive waste treatment.

6.1 Identification of main dangers and risk assessment

One of the primary tasks in evaluation of radioactive material/waste should follow a scheme, similar to that sketched in chapter 2 for the classification of *dirt*. Then a crude comparison of the result should be made with other risks in life, what attention they have received in the past. What is the price to be paid of doing nothing? When the necessity of action on waste is established the fastest, most economical, and safest way for the elimination of the problem should be attempted.

6.2 Classification of waste according to magnitude and danger, with and without remedy

(a) A huge amount of **liquid waste** was **already released** to the environment by pumping it underground, mainly by the former USSR, amounting to 1 billion Ci. The volumes of radioactive waste that have been injected are 2.5 million m³ of LLW into a depth of 190 to 225 metres, and 2 million m³ of ILW and HLW at a depth of 380 to 475 metres [18]. The hope is that this waste will not migrate over significant distances. The practice of injection continues still in Russia. For comparison, at Hanford 1.3 billion m³ of liquids were discharged into the ground before 1970, however with considerably lower radioactivity than at Russian centres. In the U.S. between about 100'000 and 200'000 kg of mercury, used during isotope separation, a highly poisonous element due to its volatility and low vapour pressure, were released into the ground [25]. It is at least as dangerous for health as radioactivity. There is close to nothing that can be done to further contain or to neutralise the injected waste.

(b) There are radioactive products moved to the Earth's surface (and into the atmosphere) by mining uranium. Among the most prominent quantities are the **mill tailings**.

As of 1990 the FSU had accumulated approximately 5 billion tons of uranium mill tailings, with an annual production of 6 to 7 million tons coming from uranium mining operations. The total contaminated area from uranium mining and milling operations in the FSU is estimated at 600 km². The total activity of waste from uranium milling and mining operations is 600·10³ Ci [18]. By the year 2000, Russia is expected to cover its mill waste with 1.5-m-thick clay covered with grass, returning most of the land back to the "economy" [18], confirmed during a meeting at MINATOM in 1997 [36]. In the U.S. there are 120·10⁶ m³ mill tailings with small radioactivity levels, the principal hazard are the α -emitting Radon isotopes. There is the Department of Energy (DOE) *Uranium Mill Tailings Remedial Action Program* for the total contaminated area of ~4 km² [5, 18].

Are there means to cover mill tailings in a way, that radioactivity won't come at all into contact with large areas of the environment, humans, animals, plants? What is the required low dose level for this condition? These questions should be seen in context with studies, which were started already more than 100 years ago, that low-level radiation is not only harmless but actually beneficial. This stimulating and protective effect of small doses of *radiation* is known as radiation *hormesis*, also termed *adaptive response* [20, 37].

Global **stocks of plutonium** (1'160 tons), increasing by up to about 80 tons each year, can be judged from two points of view: stocks are huge, as far as their weapon usability is concerned, however, they are small as far as volume is considered. The largest weapon-grade quantities are either inside the weapons, or at the weapons laboratories of the **Nuclear Weapon States (NWSs)**. The civil plutonium can be found at or near almost all power stations, and at the few reprocessing plants.

Every ton of spent fuel contains enough plutonium for one or two nuclear weapons. End of 1998 there were about 130·10³ metric tons of it, almost a third of it American. Most of it (~90·10³ tons) remains at the 236 nuclear power stations (which together have 433 reactors) in 36 different countries. The total plutonium in spent fuel is ~1.1·10³ tons, or about four times the plutonium in nuclear arsenals of the U.S. and FSU at the peak of the arms race. Additional 10·10³ tons are equivalent to 100 tons plutonium [38].

In the reprocessing scheme with closed fuel cycle of Light-Water Reactors 1'000 tons spent fuel will be accumulated before the end of a 40-year operating limit. 20 to 27 tons are removed each year. Only one third had been reprocessed. The 10·10³ tons annually generated is roughly three times greater than the world's total maximum rated reprocessing capacity of about 3.2·10³ tons. Till now 180 tons of plutonium were reprocessed, three-fourth of them by French and British companies. Reprocessing releases (on a regular base or by accidents) a certain amount of radio-activity into the environment. The amount of activity in 1996, released by the Sellafield site (UK) until 1986, is 1'250·10³ Ci, the one by the La Hague site until 1985 (France) 380·10³ Ci [39] (estimates of key radionuclides, combined for the two sites, are 800'000 Ci of Cs¹³⁷, 135'000 Ci of Sr⁹⁰, 40'000 Ci of Tc⁹⁹, and 16'000 Ci of Pu^{239,240}). In comparison Russian environmental contamination by spent fuel reprocessing wastes amounts to ~1'130·10⁶ Ci at Tomsk-7, ~450·10⁶ Ci at Krasnoyarsk-26, and ~130·10⁶ Ci at Mayak [32].

6.3 Classification of plutonium and (highly-)enriched uranium as waste or asset

This classification is mainly a political and economical one, and depends heavily on the intended use and trust in safe-guarding. If proliferation risks can be disregarded then all three materials can be considered assets, provided they are handled in a safe way. Plutonium may be a valuable asset for future electric energy production after uranium resources are exhausted. Low enriched uranium will still serve for long time as fuel of power reactors, and highly enriched uranium in a variety of research reactors.

6.4 Danger of long-lived radioactive materials compared with short-lived

Categorising isotopes according to half-life makes only sense if simultaneously their quantity is expressed in number of atoms **and** their biological/health effect is taken into account. The public shows a tendency to look only at life-times, forgetting the other parameters, in particular the relation of accumulation in the body by ingestion and inhalation during the life span of humans.

The classification as waste or as asset may also change with scientific development and newly detected applications. An example is americium, first bedevilled, now hailed by some as a possible power supply for space craft in the exploration of the universe.

Scientists are just beginning to investigate benefits of transuranic elements (minor actinides), such as neptunium, americium and curium, which represent roughly 10 percent of that of plutonium.

6.5 Safe storage of some waste products

Radioactive material has been dug out of the Earth's crust and can be returned to it safely. Nobody worries for instance about the natural nuclear reactor of Oklo in Gabon, which burned for several million years and released 15'000 Megawatt-years of energy. Migration of radionuclides (actinides) of Oklo amounts only to two tens of meters in 1.7 billion years [37, 40].

A tremendous task ahead is to convince the population that there are many valuable procedures for safe storage of radioactive waste products. Many places have been identified to be appropriate. Until a decision is taken by the government almost all of the waste in the U.S. is currently being stored at the sites where it was created.

The best technologies aim for solidification and packaging the material in containers. The vitrification is mainly envisaged for liquid waste. The *Defence Waste Processing Facility at Savannah River* was brought on line in 1996 after 18 years of planning and \$2 billion in construction cost [26]. After chemical treatment for the concentration of long-lived radionuclides it is mixed with finely ground glass and heated and melted at about 1'150°C. It is then poured into stainless steel canisters, 0.6 m in diameter and 4.5 m long. The planning is for producing about $12 \cdot 10^3$ such canisters of glass during the first quarter of next century.

Deep seabed disposal is presently banned by international law.

More details on waste management ideas can be found in the proceedings of a Pugwash workshop on the Prospects of Nuclear Energy [41], where also the disposition and disposal of plutonium is being discussed [42, 43, 44].

6.6 Transmutation, a way to eliminate long-lived radioactive waste?

The *Committee on Nuclear Wastes of the National Research Council* in the U.S. has reviewed the three principal transmutation concepts for commercial reactor spent fuel: *Separations Technology and Transmutation Systems* (STATS) [45]. These concepts use a (i) light-water reactor (LWR), an (ii) advanced liquid-metal reactor (ALMR), or an (iii) accelerator-driven subcritical nuclear reactor concept for producing neutrons, called the accelerator transmutation of waste (ATW). The committee found no evidence that applications of advanced Separation & Transmutation have sufficient benefit for the U.S. high level waste (HLW) program to delay the development of the first permanent repository for commercial spent fuel. The committee concluded that the once-through LWR fuel cycle should not be abandoned.

The difficulties with transmutation of long-lived radionuclides into short-lived or even stable forms are both technical and economic. From the technical side it is the requirement to enrich the long-lived isotopes to acceptable levels prior to transmutation. On the economic side, commercial-scale transmutation would require a large reprocessing and reactor infrastructure. It seems that decades of additional work is required to find out if transmutation can be made effective for today's volumes of commercial spent fuel and defence waste and for the expected future quantities. Basic research on accelerator technologies should be continued in view of dwindling fossil resources, but will in what follows only briefly be discussed.

6.7 The energy amplifier, an option to satisfy future electricity demand and to eliminate long-lived radioactive isotopes?

As soon as a renewal of nuclear energy will take place, fast neutron reactors will again come to the forefront, because of the remarkable capability of breeding fuel. The whole of the fission energy contained can be made available. Fast neutron reactors can also be made the most efficient burners of fissile nuclei. Larger number of neutrons are available when fission chain reactions are kept going. One may design the reactor core to the purpose in mind, and direct neutrons in excess either for producing a maximum of new fissile nuclei through their capture by uranium isotope 238 or thorium isotope 232, or for consuming actinides by means of fission reactions, or for destroying by transmutation a number of unwanted nuclei.

The possibility to make strong neutron sources by means of spallation nuclear reactions, caused by letting high energy protons hit heavy nuclei, is not a new idea. Early in the 50s linear accelerators (MARK I and MARK II), working in a pulsed mode, were built in Livermore, producing currents between 50 and 500 milliamperes. To combine a particle accelerator and a nuclear fission reactor into one integrated hybrid-system was proposed about twenty years ago. The idea goes back to Los Alamos studies in the early seventies and has been picked up by Carlo Rubbia at CERN in the early nineties [46, 47], calling it now an "energy amplifier" (EA). Such a system could in the view of their proponents also be used to produce practical, large scale amounts of nuclear transmutations, in order to

- (1) eliminate the unwanted long-lived, radio-active waste from existing nuclear reactors,
- (2) produce vast energy in "cleaner" conditions, comparable to the promise of fusion,
- (3) produce short lived isotopes for medical applications, as a local substitute to reactors.

The new concept of energy amplification is to extract nuclear energy with the help of accelerator induced nuclear cascades. The energy is produced from a nuclear fuel material disposed in a moderator medium through a process of breeding of a fissile element from a fertile

element of the fuel material. Thorium as breeding fuel has considerable advantages when compared with uranium, in contrast with the full breeding based classic reactors for which thorium presents serious difficulties. Thorium is more abundant than uranium (may be no longer true, when the vast amount of uranium contained in ocean waters can be extracted at reasonable cost [42, 48]), it generates much less transuranic actinides among the radioactive waste and the risk of nuclear proliferation is negligible. According to the proponents of the project, the accelerator-driven reactor has inherent safety, since it can work sub-critical, can be stopped any time by cutting off the beam.

The schematic layout of Rubbia's EA (Fig. 4) combines a proton accelerator (1 GeV, 30 mA or so, not shown in the diagram) to a fast neutron subcritical reactor of 1'500 MW thermal power, cooled by convection driven molten lead.

There are arguments against building such a hybrid-system from technical, safety and economical points of view [49, 50].

Starting with item (2), the production of vast energy under cleaner and safer conditions, several technical difficulties have to be discussed. They arise primarily with the so-called beam entrance window, which separates the accelerator vacuum pipe from the liquid lead. It has to be thin enough to let the protons pass without much energy loss, thick enough to withstand the pressure of the molten lead, and resistant to the intense beam induced nuclear reactions, which may weaken its structure. An exchange of the window in regular intervals should be possible without major interruption of the operation of the reactor.

The second major technical point is to guarantee the convection of liquid lead due to gravity alone, not requiring any pumps. Some experience with this kind of cooling has been gained from six nuclear power driven Russian Alpha-class submarines, which are taken all out of service for reasons not made public. According to information from MINATOM [35] there are no further plans to built reactors for submarines based on this technology.

The third problem concerns the proper spacial distribution of the very fast neutrons. They are strongly anisotropic in the direction of the proton beam, causing a problem in respect to the efficiency of the heat transfer system.

It is argued that a slightly subcritical fast neutron reactor is **safe enough**. However, a hybrid-system will have to be operated with a criticality value k_{eff} close to 1 in order to be economical interesting and may need bar control after all. There remains also the problem that decay heat removal after shutdown for all fast neutron reactors will be the same in both schemes. If a reactor is critical or subcritical does not matter in view of the amount of fission products produced. They depend only on the thermal energy released.

Finally, since the proposed whole system is more complex than a normal fast neutron critical reactor, it appears to be less economic. The investment required for developing the whole system at once is estimated by some experts in the Euratom Scientific and Technical Committee in August 1997 to run into tens of billions of ECU (quoted in [49]).

During the various stages of the project the emphasis of the group appears to have shifted away from energy production now more towards items (1) and (3), namely waste incineration and reduced production of long-lived isotopes, and production of short-lived isotopes for medical application. However, it remains debatable if the production of isotopes for hospitals is a strong supporting argument for building the EA.

It is argued by one critic [49], that there is no difference concerning the production of waste in the hybrid system and in fast neutron reactors. However, various fission products seem to be readily transmuted. In a fast neutron environment long-living minor actinides become

fissile and can thus be burnt. It is claimed [45, 46] that the EA would be able to transmute the wastes from the current generation of nuclear reactors, which must be an enormous task for many installations of this type considering the huge quantities to be dealt with.

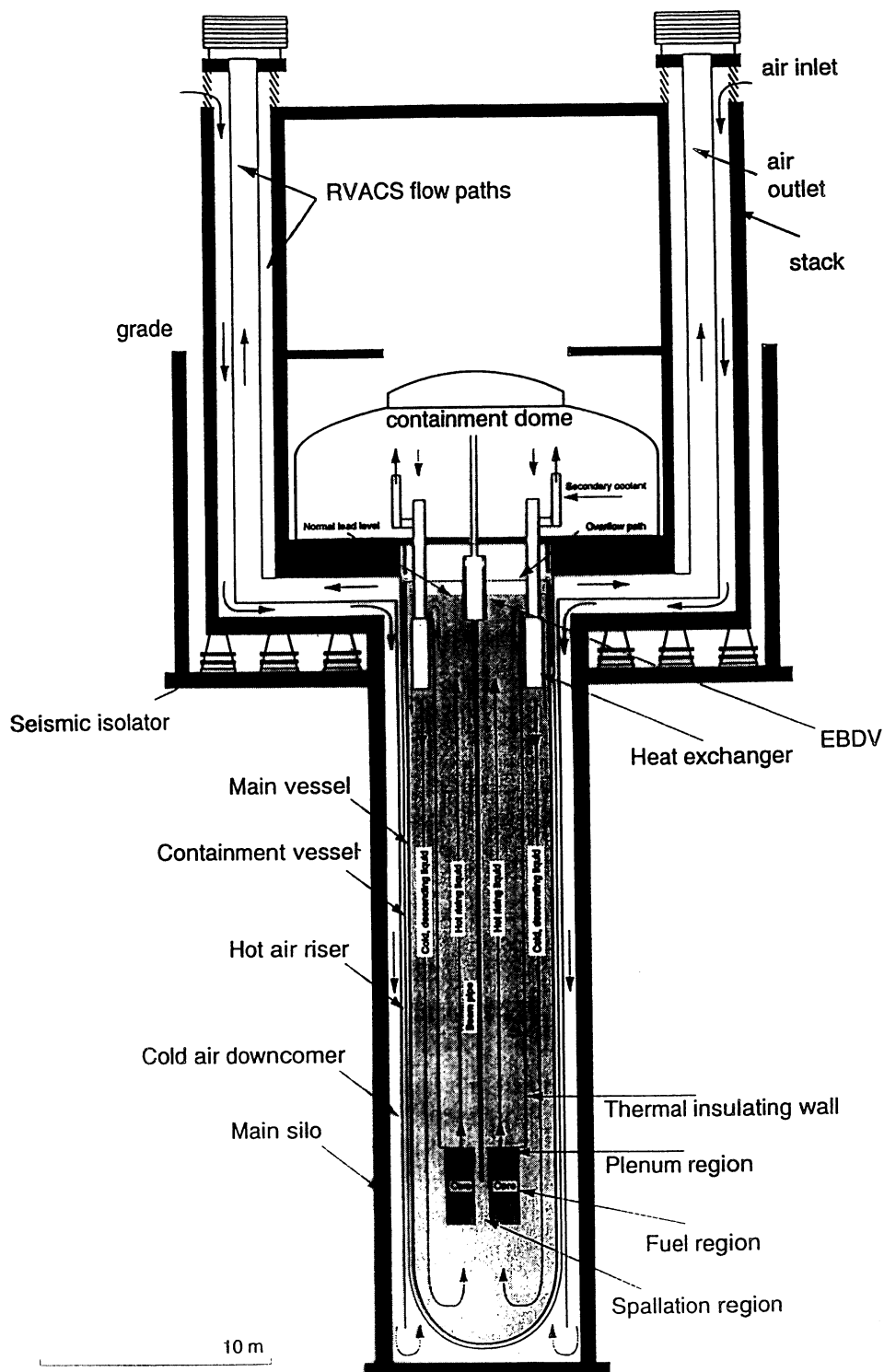


Fig4. General layout of the Energy Amplifier/Beam Dump.

7. Conclusions

During the Cold War the two superpowers produced huge amounts of (weapon-grade) plutonium and highly enriched uranium (HEU). During this process they left behind a tremendous amount of radioactive material. Both countries do not yet know how to deal with this legacy in a responsible way. Is this material of any further use for humanity? Apart from a straightforward definition:

Radioactive waste is unneeded material that contains unstable elements that decay by emitting alpha, beta or gamma emissions,

some other definitions are offered, which might/should be combined into a single one:

- (1) *Material is defined as radioactive waste if it cannot be used for beneficial purposes in any foreseeable future and produces harmful effects on humans and the environment.*
- (2) *Material is defined as radioactive waste if it consists of isotopes of any half-life, when it can get into humans either by ingestion, inhalation, or physical contact with the body, and when its gamma-rays penetrate the body in such a quantity that it can cause cancer (a probability still to be evaluated).*
- (3) *Material should be defined as radioactive waste if it can be transformed easily into matter for building nuclear weapons.*

In the U.S. there is a 2: 1 ratio in volume between DOE and Commercial LLW [9]. High level liquid waste is concentrated in storage tanks at weapons laboratories, mainly in Hanford/Washington.

In the FSU, a considerable amount of the radioactivity had already been released to the environment. A remedy by covering the contaminated area with earth can be only partially satisfying. A major remaining problem in Russia is the power reactors on the ~150 decommissioned submarines, which can be solved provided enough money can be made available (\$7 to \$10 million per submarine, and including fuel and waste storage a total estimated cost of \$2.2 billion [34]). Other problems in the FSU and former East-block countries are related to potential accidents at outdated nuclear reactors.

Nuclear waste from civil power reactors should get appropriate attention, but does not present a serious or immediate danger. Dry storage on surface should be reconsidered in view of the non-proliferation aspect. Plutonium stocks will increase as long as the civil power reactors of the present type are operated. Therefore, proliferation risks will continue to exist. A nuclear-weapon-free-world must be the ultimate goal of all politics. It is imperative that the public is informed correctly on risks and remedies in order to come to valuable solutions in a democratic way.

The responsibility of scientists is challenged to solve the problem of radioactive waste, to develop the technical means for it, and to inform and educate better the non-scientific community.

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1 [Becquerel, Bq] is equal to one decay per second
1 [rad] is equal to an absorbed dose of 0.01 J/kg deposited in any material by any kind of ionising radiation
1 [rem] is the unit of equivalent dose that equals the dose in rads multiplied by appropriate weighting factors to account for the dependence of biological effects on the type of ionising radiation.
1 [roentgen] occurs when the sum of electrical charges on all ions of one sign produced by X- or γ -rays in 1 kg of air is 0.000258 Coulomb.
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