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Nuclear High-level Waste Tank Explosions: Potential Causes and Impacts of a Hypothetical Accident at India's Kalpakkam Reprocessing Plant

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ABSTRACT

Tanks holding liquid high level waste from reprocessing spent fuel have large inventories of highly radioactive materials. These tanks could potentially be damaged by a variety of chemical explosions, leading to the dispersion of a significant fraction of their radioactive contents. This article describes some of the different chemical explosions that could occur and examines how such explosions could occur at the Kalpakkam Reprocessing Plant in India, which likely stores a large volume of high level liquid waste because vitrification of that waste did not begin until more than 15 years after the plant began operating in 1998. The atmospheric dispersion of the hypothetical radioactive release is modeled using the Hybrid Single-Particle Lagrangian Integrated Trajectory Model developed by the Air Research Laboratory of the U.S. National Oceanic and Atmospheric Administration. The results suggest that the modeled accident scenario would lead to nearly 97,000 cancers, with roughly 47,000 of these being fatal. Larger radioactive releases are possible and would lead to proportionately higher incidence of cancer and cancer-caused mortality.

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Introduction

Reprocessing operations are susceptible to many types of accidents (see Table 1).[1] Reportedly, the historical frequency of accidents at reprocessing plants is greater than the frequency of accidents at nuclear reactors.[2] There have been at least 16 serious accidents at 10 reprocessing plants worldwide,[3] including 2 criticality accidents, 3 fires, 8 chemical explosions, and 2 loss of coolant accidents.[4]

Among the accident risks posed at reprocessing plants and associated facilities, the category of accidents that may have the greatest potential consequences to public health and the environment are explosions at liquid high level waste (HLW) tanks, including explosions that occur as a result of the tank drying out.[5] These tanks contain highly concentrated radioactive fission products found in spent fuel. Therefore,

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Table 1. Types, releases, locations and year of accidents at reprocessing plants and associated nuclear facilities.

Type of Accident	Liquid Releases	Gaseous Releases	Occurrence
Criticality in dissolver tank	X	X	Windscale, 1973
Fire		X	La Hague, 1981 Karlsruhe, 1985 Tokai, 1997
Explosion		X	Savannah River, 1953 Kyshtym, 1957 Oak Ridge, 1959 La Hague, 1970 Savannah River, 1975 Tomsk-7, 1993 Tokai, 1997 Hanford, 1997
Leak in a discharge pipe; tank breach	X		La Hague, 1979–80 Sellafield, 1983
Loss of coolant		X	Savannah River, 1965 La Hague, 1980

Source: Adapted from "Background, Status, and Issues Related to the Regulation of Advanced Spent Nuclear Fuel Recycle Facilities," Advisory Committee on Nuclear Waste and Materials White Paper (Rockville, MD: Nuclear Regulatory Commission, June 2008), p. 35.

a tank explosion could lead to the dispersion of large quantities of radioactivity into the atmosphere.

The quantities of radionuclides that could potentially be released are comparable to the 1986 Chernobyl and 2011 Fukushima Daiichi accidents. In both cases, the primary long-term detrimental effect resulted from cesium-137 contamination. The radioactive element cesium-137 emits penetrating gamma rays as it decays with a half-life of 30 years.[6] The Chernobyl accident released about 85 PBq (1 PBq $= 10^{15}$ Bq) of cesium-137 to the atmosphere.[7] Estimates of the release from the multiple accidents at the Fukushima Daiichi power plant are varied, with a range from 10 PBq,[8] to 18–19 PBq,[9] to 35.8 (23.3–50.1) PBq.[10] In comparison, a single tank of HLW could contain hundreds of PBq of cesium-137. Thus, the dispersion of even a small fraction of this inventory could result in severe consequences.

This is a special concern with the reprocessing plant in Kalpakkam, India (Kalpakkam Reprocessing Plant or KARP, henceforth) because the associated plant for vitrifying the HLW produced in KARP did not start operating till 15 years after KARP began reprocessing spent fuel.[11] This implies that there must be a considerable amount of accumulated HLW in storage tanks.[12]

In this article, we focus on the consequences of a hypothetical accident at a HLW tank at KARP. After a brief overview of reprocessing and waste treatment, we discuss three kinds of chemical explosions that have occurred at reprocessing and associated facilities around the world and the conditions necessary for such explosions to occur. We then discuss some of the specific features of the HLW tanks at KARP, including some reasons for concern. Finally, we discuss the results of a simulation of atmospheric dispersion of a radioactive release from the HLW tank at Kalpakkam, including a calculation of the potential radiation dose to the exposed population.

Overview of reprocessing

Spent fuel refers to fuel that has been irradiated in nuclear reactors, and consists mainly of uranium that has not undergone fission, plutonium and, other transuranic elements such as neptunium, and fission products. Spent fuel generates large amounts of heat and therefore it is initially cooled in pools filled with water. Once cooled, there are two primary options for the disposition of spent fuel: direct disposal and reprocessing. In direct disposal, the spent fuel is stored in an interim storage facility or in the pool itself with the intent of eventual disposal in a geological repository if and when such facilities become available. Reprocessing and reprocessing facilies are the focus of this article.

Purex

Reprocessing can be done using a variety of chemical processes. The conventional method is called the Plutonium Uranium Redox EXtraction process (PUREX), which was originally developed to separate out plutonium from spent fuel for use in nuclear weapons. Spent fuel is reprocessed in batches (rather than continuously) and the reprocessing of each batch of spent fuel is called a campaign. The first step in the PUREX process is to remove the covering, or cladding, of the spent fuel rods (see Figure 1). This is usually done by chopping the rods into small pieces (about 5 cm long) and placing them in a hot (about 100°C) nitric acid solution.[13] The nitric acid dissolves the contents of the fuel rods, which are then mixed into the solution as nitrates. Most of the cladding does not dissolve and is disposed separately. Both chopping and dissolution release radioactive gases.

In a PUREX process plutonium and uranium are separated from the nitric acid solution (also known as the aqueous solution) based on their relative solubility into two different immiscible liquids, the aqueous acid solution and an organic solution. In the PUREX process, the organic solvent is tributyl phosphate (TBP) diluted in kerosene.[14] When the aqueous and organic solutions are mixed together, the plutonium and uranium present in the aqueous phase are transferred to the organic phase, whereas the fission products and other elements remain in the aqueous phase. When the mixture is allowed to settle, the two phases separate, thereby separating the uranium and plutonium from the fission products. This process of mixing and settling is repeated several times to extract most of the uranium and plutonium from the acidic solution.

Uranium and plutonium can be separated from one another using the same principle of differential solubility once more and converted into oxides for ease of storage and transportation.

High level waste

In addition to plutonium and uranium, reprocessing produces liquid waste streams. In India, liquid waste streams are classified as low, intermediate (or medium), and

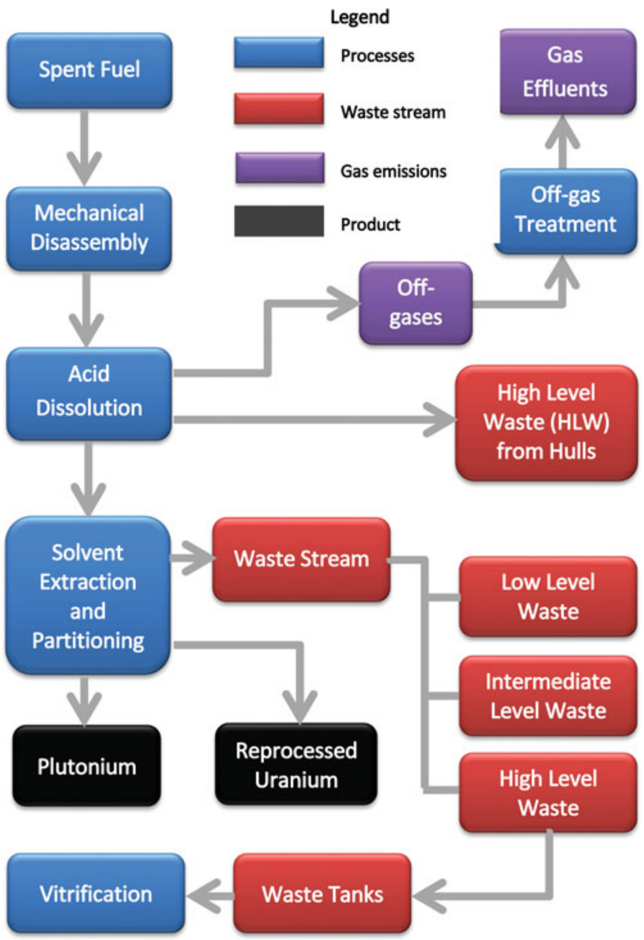

Figure 1. Schematic view of spent fuel reprocessing and radioactive waste treatment.

HLW depending on the radioactivity level or concentration. The largest component by volume is low level waste containing over 80% by volume of the waste stream, but only about 0.1% of the total radioactivity from the spent fuel.[15] HLW, on the other hand, constitutes less than 2% of the volume but contains nearly 99% of the total radioactivity in the spent fuel.

The composition of HLW depends on several factors, including the type of fuel, its burnup, and reprocessing chemistry.[16] It contains both long and short lived radioisotopes, including unrecovered plutonium and uranium, processing chemicals such as nitric acid and sodium nitrate, dissolved cladding materials such as zirconium, and the corrosion products of storage tanks and piping. Some dissolved TBP is also found in HLW. High level waste is acidic with a typical molar concentration (molarity) of 1–3 M of nitric acid (or 63–189 grams of HNO_3 per cubic decimeter).[17]

HLW is concentrated and its volume reduced by evaporation. The amount of reduction depends on the burnup and on the cooling time before reprocessing. Depending on "the length of the cooling period, either the heat generation of the resulting HLW or its content of dissolved solids including fission products and process chemicals are the factors limiting the degree of concentration."[18] The concentration of the HLW will also increase once stored in tanks through further evaporation. The resulting volume of HLW after concentration ranges from less than 0.1 to more than 1 m^3 for each ton of heavy metal reprocessed.[19]

Because of the heat constantly generated by the HLW, the storage tanks have to be cooled continuously. To reduce the hazards associated with the possibility of cooling failure, HLW is vitrified, which converts it into a solid form.[20] Vitrification involves treating the HLW by first concentrating it in evaporators to reduce its volume and then subsequently mixing it with additives such as SiO_2, B_2O_3, $A1_2O_3$, P_2O_5, Na_2O, and CaO at high temperatures.[21] When cooled and solidified, the mixture forms a glass or a near-glassy solid. However, vitrification is a complicated technology that is very sensitive to the chemical nature of the radioactive mixture that is being vitrified.[22] In this article, we focus on potential accidents at the liquid HLW stored in tanks and do not consider hazards involved with the vitrification process or vitrified waste.

Accidents

Given the complex nature of reprocessing plants, there are many types of accidents that could result in a radioactive release. Such accidents could be initiated by equipment failures, human errors, external natural events and malevolent acts. Radioactivity could be released through one or more mechanisms, including leakage and atmospheric dispersion. Analysts have highlighted the potential for cascading incidents, wherein initial events might create "high radiation fields, or other phenomena that hinder mitigating actions" thereby rendering further accidents more likely.[23]

There have been a number of probabilistic safety assessments (PSA) or probabilistic risk assessments (PRA) of reprocessing facilities around the world.[24] However, most of these assessments are not comprehensive.[25] Further, there are reasons to expect that PRAs underestimate the risk of accidents because they inevitably overlook some accident initiators;[26] they are also unable to comprehensively evaluate all possible human errors and malevolent actions.

HLW tanks typically incorporate multiple levels of safety, including cooling coils to remove decay heat, methods to remove hydrogen,[27] and equipment to stir the liquid to reduce the accumulation of sludge at the bottom of the tank.[28] At KARP, HLW tanks are emplaced in underground vaults, with thick walls and coverings.[29]

Despite these measures, accidents have occurred at several reprocessing plants and their associated facilities. Because of the large amount of radioactive materials present in reprocessing plants and HLW tanks, when safety mechanisms fail, the consequences can be extremely serious. The purpose of this article is to analyze and explore the consequences of chemical explosions which result in the release of radioactive material rather than determining the likelihood of such an accident.

Chemical explosions

An explosion is a fast release of a large amount of energy. In reprocessing plants and associated facilities, an explosion may lead, directly or indirectly, to the dispersion of radioactive material both inside and outside the facility through damage to the containment.[30] Explosions can occur due to the accumulation of flammable substances or runaway exothermic chemical reactions. Hydrogen, various salts, and organic materials are common causes of chemical explosions.[31]

Many explosions have occurred at reprocessing plants and associated facilities around the world. Below we summarize an example of each type of chemical explosion and what is known about each type. The likelihood and consequences of each type of explosion depends, in part, on the characteristics of the HLW solution.

Salt explosions

The most prominent example of a salt explosion occurred in September 1957 at the Mayak facility in the Soviet Union. Estimates of the explosive power of the accident range from 25—29 tons of TNT equivalent,[32] to 70—100 tons of TNT equivalent.[33] The accident was retrospectively classified as level 6 on the International Nuclear Event Scale (INES). About 70—80 tons of highly radioactive waste with a total radioactivity content of 740 PBq was released into the atmosphere.[34] According to subsequent estimates about 90% of the released radioactivity settled within about 5 km of the site.[35]

About 4 PBq of strontium-90 and yttrium-90 and about 18 PBq of zirconium-95 and niobium-95 were released.[36] A predetermined contamination density level of 74 kBq/m^2 of strontium-90 triggered the evacuation of twenty-two villages with a total population of about 10,000.[37] The area with a radioactive contamination density of 3.7 kBq/m^2 or higher was about 20,000 km^2.[38]

The explosion occurred in a stainless steel tank storing radioactive wastes resulting from the separation of plutonium. The tank had an outside diameter of 8 m and height of 6 m.[39] It was placed in a canyon of reinforced concrete, and covered with a cylindrical plate of reinforced concrete about 0.8 m thick and a layer of soil 1—1.5 m thick. The intensity of the explosion was large enough to rupture these barriers.

The tank held liquid radioactive wastes that required decay heat cooling to remove about 79 kWt and salts, primarily sodium nitrate and acetate. When the cooling system failed, the temperature in the tank rose to 350°C, the water evaporated, and there was a thermal explosion of the dried salt residue that breached the thick protective structures.[40]

Red oil explosions

Red oil explosions have long been considered a potential hazard at reprocessing plants.[41] Red oil is "defined as a substance of varying composition formed when organic constituents react with nitric acid."[42] The organic materials referred to here are TBP and various organic chemicals such as dibutyl phosphate and monobutyl phosphate that are formed when TBP is exposed to radiation.

The primary concern with red oil is its rapid decomposition, which is a high energy exothermic reaction, and "the associated over pressurization through a self-heating runaway reaction known as a Red Oil Excursion (ROE)."[43] The rate of the decomposition reaction, k, is usually described by an Arrhenius type relationship

$$k = A * exp\left(\frac{-E}{RT}\right)$$

where E is the energy of activation, T is the temperature at which the reaction is occurring, R is the universal gas constant, and A is called the frequency factor, which sets the scale of the reactions. In other words, the reaction rate increases rapidly with increasing temperature above a certain threshold. Since the reaction is exothermic, the heat accelerates the reaction rate and, unless the heat is transferred, it produces a runaway reaction. The onset temperature, which initiates a runaway reaction, and the activation energy depend on a number of factors, including the concentration of nitric acid and whether the system is closed or open.

An early estimate of these quantities under closed conditions found an onset temperature of 120°C and derived a value of 112 kJ/mole for the activation energy.[44] The frequency factor A depends on the amount of nitric acid, and was estimated as 4.3×10^{10} per second for 6–11 moles of nitric acid in the aqueous phase.

Other studies report different figures, including significantly lower onset temperatures. One study found an onset temperature for exothermic activity as low as 75°C, albeit at high acid concentrations.[45] The amount of exothermic energy that is released appears to depend on whether the heating occurs in an open or sealed tank.[46] Heating in sealed vessels is more likely to lead to explosions.

In addition to the rapid generation of heat, red oil reactions can produce large quantities of flammable gas.[47] If the heat is not removed and the gases are not vented fast enough, the vessel may rupture from high gas pressures.

Red oil accidents have occurred in many countries. In the United States, red oil accidents occurred at the Hanford Site in 1953, and at the Savannah River Site in 1953 and 1975.[48] In both explosions at Savannah River, solutions of uranyl nitrate, which, unbeknownst to the operators, contained TBP and its organic diluent, were heated and resulted in damaging explosions.[49]

Tomsk-7, Seversk, Russia

Perhaps the most severe red oil explosion occurred in 1993 at the Tomsk-7 site at Seversk, Russia. Categorized as level 3 on the INES scale,[50] the accident occurred in a stainless steel vessel with a volume of 34 m^3 used to prepare the solution prior to reprocessing;[51] the vessel was located in a cell below ground level with 2 m thick concrete walls and a concrete roof. [52] At the time of the accident, the vessel is reported to have "contained a total of 449 g of plutonium and 8757 kg of uranium which, assuming specific activities of 2.3 TBq/kg and 12.4 MBq/kg for the two elements, corresponds to total activities of 1.0 TBq and 0.11 TBq, respectively."[53]

The accident started with the addition of 1.5 m^3 of nitric acid to an organic mixture (volume of 23.5 m^3) at 10.30 PM on 6 April 1993.[54] It is estimated that the mixture contained about 0.5 m^3 of TBP.[55] Either because of equipment failure or operator error, compressed air, which was normally introduced into the tank to "ensure a uniform temperature and concentration of nitric acid," was not added to the solution.[56] This resulted in a layer of the organic solvent (density of 1400 kg/m^3) being sandwiched between a hot layer of uranyl nitrate solution (with a density of between 1480 and 1590 kg/m^3) below and a layer of nitric acid (density of 1390 kg/m^3) above. The solvent layer became hot and an exothermic reaction leading to the formation and decomposition of red oil ensued.[57] There is some evidence that the onset of the reaction occurred when the temperature of the organic compounds was as low as 90°C.[58]

Around midnight, monitoring instruments showed rising pressure. Despite attempts to depressurize the vessel, the pressure continued to increase and reached 5.0 atm (0.51 MPa) by 12.55 AM on 7 April. The pressure continued to rise rapidly and within a few minutes the vessel ruptured, followed by an explosion several seconds later. The building housing the vessel and its neighboring industrial structures were partially destroyed. A shock wave propagated through the corridor of a neighboring building and destroyed a glass-block wall.[59]

The strength of the explosion has been roughly estimated at 100 kg of TNT equivalent or 418.4 MJ. The maximum pressure that caused the rupture has been estimated at 3.03 MPa.[60] The damage to the building is believed to have been a result of the deflagration of flammable gases, most likely butene (C_4H_8) and CO, released by the initial explosion.[61] Radioactive material was released to the environment through large holes in the side walls and roof of the building.

The fraction of the fission products that were released during the accident are uncertain, but significant. According to one estimate, a total of 4.3 TBq of long living fission and activated nuclides may have been released from a total of 20.7 TBq.[62]

Nuclear fuel complex at Hyderabad

Tomsk was not the last site of a red oil explosion. During the early hours of 17 November 2002, an explosion occurred in the evaporator section of the solvent extraction plant of New Uranium Oxide Fuel Plant (NUOFP), at the Nuclear Fuel Complex at Hyderabad, India.[63] The process used at the complex involves dissolving the yellow cake, which is produced by processing uranium ore, in nitric acid and using solvent extraction to obtain uranyl nitrate, which is then futher processed to produce uranium dioxide.[64] Like the PUREX method, this process uses TBP.[65] Following the accident, a special investigation committee was appointed by the Atomic Energy Regulatory Board (AERB), which observed that conditions which could produce a red oil explosion, including the presence of nitrate, organic liquids, and high temperatures, were present at the time of the accident.[66] According to the AERB, "the carry over of organic solvent into the evaporator along with the use of steam higher than set pressure resulted in rise in the temperature of organic nitrate complex above 130°C, taking the reaction into a run away mode and thereby leading to red oil explosion"[67]

The accident prompted research on red oil conducted by multiple institutions associated with the Indian nuclear establishment and academia, presumably funded by the Department of Atomic Energy.[68] One set of studies conducted at the Indira Gandhi Centre for Atomic Research synthesized red oil and established that "presence of metallic nitrates was not essential for red oil formation as thought earlier and that red oil formation occurred at acid concentrations that were considerably lower than what had been assumed thus far."[69] Another study "strongly" advocated "for a revisit of safety limits set in the fuel reprocessing plants to prevent red oil formation" because it showed that that the onset temperature for runaway red oil decomposition could be as low as 75°C, much lower than previously assumed.[70] Finally, one review of red oil excursions cautioned that "radiolysis of the organic phase and accumulation of the radiolytic degradation products in the aqueous phase may have an accelerating effect on red oil formation and also on the decomposition behavior."[71]

Hydrogen explosions

An important explosion risk is created by the accumulation of hydrogen. Hydrogen can be produced from a variety of chemical reactions, or through the disintegration of chemicals, including water, by high levels of radiation, i.e., radiolysis. Hydrogen is flammable if the gas is present in air at between 4 and 74% by volume and poses an explosion hazard if the concentration is above about 13% by volume.[72] Hydrogen is formed radiolytically during PUREX reprocessing in the dissolver, feed adjustment, and solvent-extraction units, and in high level liquid waste storage tanks.[73] Accumulation of radiolytic hydrogen is of particular concern in the waste tanks of reprocessing plants.[74]

HLW tanks continuously produce hydrogen due to the radiolysis of water and other chemicals in the waste. The rate of hydrogen production is characterized by the g-value, the number of H_2 molecules produced per 100 eV of radiolytic energy absorbed by the waste. The g-value for H_2 production is usually taken as 0.45 in the case of beta and gamma radiation and 1.30 for alpha radiation.[75]

Most reprocessing plants are designed to reduce the risk of the hazards of hydrogen production.[76] Nevertheless, there is a risk that hydrogen concentrations reach dangerous levels, for example, when a ventilation system malfunctions.[77] At the Hanford facility in the United States, the phenomenon of hydrogen being trapped under the surface of the waste has been observed. This leads to an increase in pressure followed by a sudden release, during which the concentration of hydrogen could reach hazardous levels.[78]

One accident resulting from the build-up of hydrogen produced by chemical reactions occurred in Dounreay in May 1977. In that case, hydrogen formed in a vertical shaft sunk in the 1950s, was ignited by a spark from a water pump, and exploded.[79] There is disagreement over whether or not the explosion resulted in local radioactive contamination.[80] In January 1970, a hydrogen explosion reportedly occurred at France's La Hague facility, but this has not been independently confirmed.[81]

Though there have been few hydrogen explosions in reprocessing plants or waste tanks, the Oak Ridge National Laboratory considered a hydrogen explosion as its "maximum theoretical accident."[82] Oak Ridge's study made "illustrative (but not the worst possible)" assumptions; the most relevant of those assumptions are that the tank is made of stainless steel (0.5 inches thick) and contains HLW that generates 11 kWt/m^3 of heat. The tank was assumed to be "housed in a 3-ft-thick concrete vault buried under 10 ft of earth." Given this configuration, Oak Ridge's accident scenario involved the tank being sealed ["loss of purge air to the tank"] and the concentration of hydrogen increasing to 30% by volume, which would take about 24 hours, followed by ignition. In this scenario, the Oak Ridge calculation found that the resultant explosion would liberate approximately 1.2 GJ (251 kg of TNT equivalent) and generate sufficient pressure to "elevate the concrete roof and earth cover by several feet …and the tank would be directly vented to the atmosphere."[83]

Necessary conditions

One or more of three preconditions must be met for a red oil, salt, or hydrogen explosion to occur. The first precondition is closed vents. An open vent allows hydrogen to escape and evaporative cooling to occur. In the absence of venting, increasing temperatures lead to increased pressures as the vapor accumulates. For the temperature to reach 130°C or more, the level at which a red oil reaction can be safely assumed to become runaway, the pressure must exceed 275 kPa.[84] In comparison, the rupture pressure of the various process tanks at Savannah River in the United States is reportedly greater than 1,379 kPa.[85] In the case of red oil and salt explosions that require higher temperatures, a second precondition is the loss of cooling. For red oil explosions, a third precondition is the cessation of internal stirring. If there is no stirring, sludge can settle at the bottom allowing the fomation of distinct layers of organic and aqueous phases.

All of these conditions may not have to simultaneously occur for a runaway reaction to take place. For example, in an unstirred enclosed vessel, available heat transfer mechanisms are limited. If a red oil reaction produced chemical heat, it would occur in a floating organic phase. Given the typical quantities of organic material in a waste tank and the dimensions of the tank itself, this phase will be in the form of a disc, with its radius being much greater than its thickness. Because organic solutions are poor conductors of heat, only a small region of the organic phase can be affected by cooling at the vessel walls even under the best of circumstances. Thus, the geometry could prevent large-scale convection processes within the organic phase.[86]

The pre-conditions could also result from a single cause. For example, an earthquake could disable venting, cooling, and stirring. Another cause could be the physical and chemical phenomena that occur in conjunction with red oil formation and its runaway reactions. For example, as the self-heating accelerates, the organic phase is prone to the formation of foam because of the gases produced.[87] Foaming provides a mechanism for plugging vents and, in effect, confining the event, as has happened in past accidents.[88] In addition, the foam can be thermally isolated from the rest of the system because of its good insulating qualities.

Because design details are not available in public, there is no way to reliably evaluate the liklihood of an explosive accident. Nonetheless, given the muiltiple failure possibliities of complex reprocessing plants, the large volumes of radioactive wastes present at these plants, and the fact that accidents have occurred in the past, accidicent scenarios are worth examining.

The Kalpakkam reprocessing plant

India's Department of Atomic Energy carries out reprocessing of spent fuel from power reactors at two locations: Tarapur on the western coast and Kalpakkam in the southern coast. There are two plants at Tarapur (commissioned in 1977 and 2011) and one at Kalpakkam (commissioned in 1998), each with 100 tons of spent fuel per year capacity. Both plants have only reprocessed spent fuel from pressurized heavy water reactors (PHWRs). Existing capacity to reprocess power reactor spent fuel at KARP is being increased.[89] However, the older Tarapur reprocessing plant is reportedly used to "carry out aged Pu purification work," which presumably means that it is no longer used for reprocessing but rather to separate out americium-241 that builds up in plutonium.[90] KARP also has a smaller facility (sometimes called a hot cell or a Lead Mini cell) for reprocessing spent fuel from the Fast Breeder Test Reactor. Spent fuel from the Protype Fast Breeder Reactor under construction will be reprocessed at the Fast Reactor Fuel Cycle Facility that is also to be constructed in the Kalpakkam complex.

Operation of KARP was originally planned for 1991,[91] but was delayed due to quality control problems in piping and other equipment,[92] and was finally commissioned in 1998.[93] The intermediate level waste (ILW) and HLW are stored in an underground facility in Kalpakkam called the Waste Tank Farm (WTF). The WTF at Kalpakkam has five stainless steel (type 304L) tanks for HLW and nine carbon steel tanks for ILW. Each of the HLW tanks has a capacity of 300 m^3.[94]

As elsewhere, the HLW at KARP is to be sent to an adjoining Waste Immobilisation Plant (WIP) to be processed in a vitrification plant. Although it was assumed that vitrification operations would start concurrently with the reprocessing,[95] this did not happen in KARP and WIP was finally commissioned in November 2013, fifteen years after KARP started operating.[96]

Because of the many years that KARP has operated without an operational vitrification facility, a considerable amount of HLW must have accumulated in liquid form in tank which can be estimated.

The amount of HLW produced depends on the amount of spent fuel processed at KARP. Operational records of KARP are not available in the public domain.

However, there is some evidence that the plant has operated poorly.[97] For this exercise, therefore, we assume that KARP has operated at an average of about 30% of its full capacity over its lifetime. At the time of this writing, KARP has operated for about 18 years. Therefore, at the assumed capacity factor, it would have processed about 540 tons of spent fuel.

The volume of HLW generated per ton of spent fuel processed is reported to be 0.6 m^3.[98] Thus, assuming 540 tons of spent fuel has been reprocessed at KARP, there should be 324 m^3 of HLW accumulated in the waste tanks. Although the volume of each tank is 300 m^3, we assume that the tanks are initially filled up to 90% of their full volume, or about 270 m^3.[99] Therefore, storing the HLW will require two tanks, one of which should be filled to the maximum allowed capacity of 270 m^3 (containing the HLW generated by reprocessing 450 tons of spent fuel). Even assuming that some of the HLW has already been vitrified by WIP, it is unlikely that the accumulated inventory of HLW will occupy less than one tank.

There are other processes to take into account. Because of the ambient temperature of the region as well as the heat generated by radioactivity, HLW in tanks would evaporate and there would be a gradual reduction in volume of HLW stored in each of these tanks over a period of time. We therefore assume that the volume has come down to 240 m^3, or about 80% of the full volume of the tank, leading to a corresponding increase in concentration of radioactivity.[100]

The radioactive content of HLW from Indian reprocessing plants is reportedly up to 7.4 PBq/m^3.[101] What is of greater interest is the amount of cesium-137 in each tank, since that is expected to be the leading contributor to the radiation dose in the event of an accident, in part because it has a half-life of 30 years.[102] For a burnup of 6,600 MWd/tU for the spent fuel from PHWRs, the quantity of cesium-137 produced in each ton of spent fuel is 0.74 PBq after a cooling period of three years and 0.71 PBq after a cooling period of 5 years.[103] Assuming that the HLW contains an average of about 7.5 years of cooling, the concentration would be about 0.67 PBq per ton of spent fuel reprocessed. Therefore, each waste tank would contain about 300 PBq of cesium-137; since we assumed that the HLW eventually occupies only 240 m^3 of volume, the concentration of cesium-137 would be about 1.3 PBq/m^3.

Elsewhere, the concentration of cesium in "simulated" HLW (used for experiments) is reported to be 543 g/m^3 and that would translate to 1.7 PBq/m^3.[104] Even after accounting for the inclusion of cesium-134, this is higher than the value we have assumed, and hence our calculations of radioactivity contamination levels and cancer casualties would likely underestimate the actual values.

There is some basis for concern that all three types of chemical explosions described earlier could take place at KARP and other reprocessing plants in India. Even the DAE stated in its 2011 annual report: "Red-oil formation with nitric acid-tri-butyl phosphate (TBP) runaway reaction is a safety concern in [any] reprocessing plant using PUREX process."[105] Such a reaction could breach the waste tank and the building containing it.

One quantitative estimate carried out by the Westinghouse Corporation for the Savannah River facility in the United States found that "in order for a runaway red oil reaction of sufficient magnitude to compromise the F-Canyon containment to occur, it must involve at least 3,000 pounds of TBP."[106] At room temperature, the density of TBP is about 0.97 g/ml.[107] Thus, the volume of TBP needed for a red oil reaction capable of breaching the containment at the F-Canyon at Savannah River would be about 1.4 m^3. Assuming that the containment at KARP's waste tanks has the same strength as the F-Canyon, the amount of TBP needed to cause an explosion that breaches the containment will be less than 1% of the volume of an HLW tank at KARP. High level waste does indeed include small amounts of organic process chemicals and something in the range of 1% cannot be ruled out.[108]

This quantitative estimate suggests that, however unlikely, it is certainly possible that a red oil excursion could develop that is capable of breaching a waste tank.[109] Although a red oil explosion may be powerful enough to cause a breach of the tank, the event may not be energetic enough to vaporize a significant portion of the HLW and the release might only have local radiological significance.

We now turn to the possibility of a hydrogen explosion. The DAE estimates that hydrogen is produced in a HLW tank at the rate of 0.149 liters/second or 0.536 m^3/hour (at standard temperature and pressure).[110] Under normal circumstances, the hydrogen will be vented. However, in case the vents are blocked, the hydrogen will start accumulating within the tank. Since we assumed that the tank, after evaporation, is filled to 80% of its volume, the remaining volume that can hold the released hydrogen is 60 m^3. The minimum concentration needed for an explosion is 13% by volume. This implies that the amount of hydrogen needed for an explosion is 7.8 m^3, requiring about 14.6 hours to accumulate. In order for the hydrogen to reach the concentration level of 30% assumed in the Oak Ridge study, it would take nearly 34 hours. It is certainly possible that a blockage of vents might not be noticed for this period of time.

At a concentration of 30%, the hydrogen would weigh about 24 kg. A very crude estimate of the effect of such a hydrogen explosion can be produced by calculating the equivalent mass of TNT that would have the same explosive power using the formula

$$m_{TNT} = \frac{f_H C_H m_H}{C_{TNT}}$$

where m_H is the mass of hydrogen that explodes, C_H and C_{TNT} are the heats of combustion of hydrogen gas and TNT respectively, and f_H is a parameter that captures the fact that not all of the heat generated by combustion goes into the shock wave.[111] Assuming a value of 0.1 for f_H,[112] 4.8 MJ/kg for C_{TNT} and 120 MJ/kg for C_H, the TNT equivalent for 24 kg of hydrogen is 60 kg of TNT.[113] Figure 2 below shows the overpressure corresponding to this TNT equivalent value as a function of distance from the point of explosion.

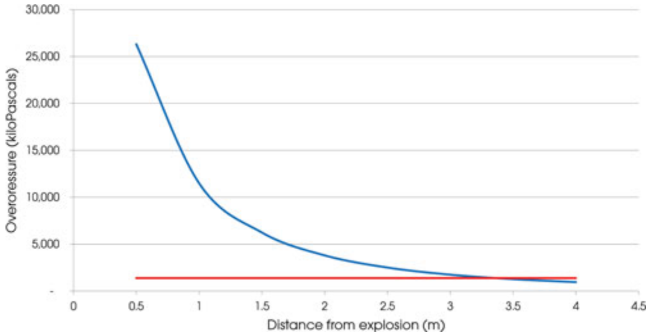

Figure 2. The upper darker curve shows the overpressure from the explosion of 24 kg of hydrogen as a function of distance. For comparison, the rupture pressure of steel tanks at the Savannah River Plant is estimated to be 1379 kPa and this is shown in the lower flat line.

The rupture pressure of the steel tanks at Savannah River is estimated to be about 1,379 kPa.[114] This will be easily exceeded in the scenario we have considered since the HLW tanks at KARP are about 4 m tall; thus, if 20% of the height is replaced with hydrogen, the center of explosion cannot be more than 0.8 m away from the top of the tank. A comparison with the overpressure from an explosion as a function of distance as shown in Figure 2 shows that the overpressure from a hydrogen explosion will likely rupture the tank. As in the case of a red oil explosions, even a hydrogen explosion that is powerful enough to cause a breach of the tank is unlikely to be energetic enough to vaporize a significant portion of the HLW. The radionuclides released from the tank by such an explosion might only have local radiological significance.

There is also the possiblity of a salt explosion because HLW tanks are reported to have compositional variations (see Figure 3),[115] and some layers might have large quantities of explosive salts that could, in the absence of cooling, experience high enough temperatures so as to set off a runoff chemical reaction. We are not aware of any quantitative estimates of salt formation, but, as discussed earlier, salt formation and drying out did lead to a major explosion at Mayak. There is not enough public information to allow us to know whether the waste tanks in KARP have accumulated similar layers of sludge and salt.

There is, however, the possibility that an explosion involving salts might occur through the accumulation of prior failures and cascading events.[116] The cooling system and monitoring sensors could fail, resulting in an increase in the temperature, evaporation of liquids and drying out of the HLW. The solid residue of the remaining radioactive material would continue to increase in temperature and could explode, in a similar fashion to the Mayak explosion described earlier. A hydrogen or red oil exposion could lead to both a tank breach as well as the creation of a radiation barrier that prevents plant personnel from being able to carry out repairs or other actions. Under these conditions, the continued evaporation and drying out of liquids might be followed by an energetic explosion of the solid residues that disperses a significant portion of the inventory of the HLW tank.

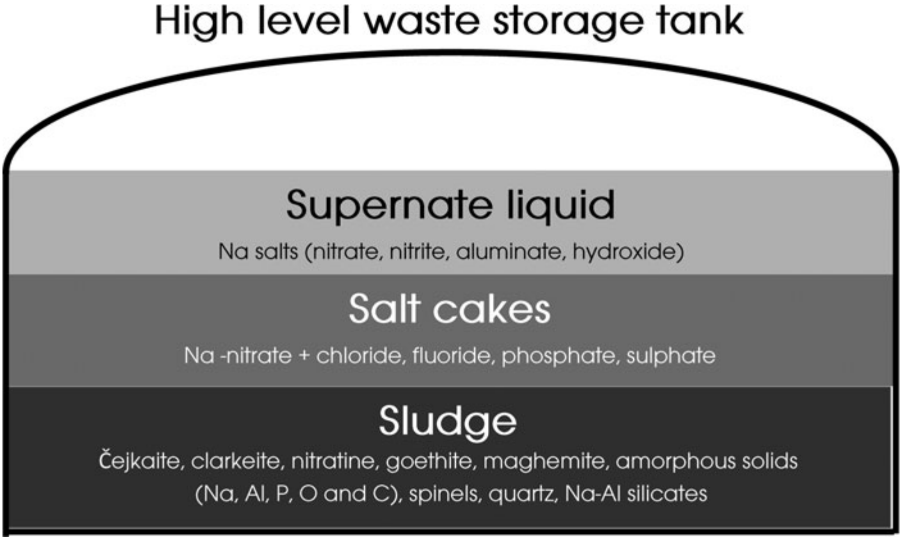

Figure 3. Schematic representation of layer formation in a high level waste storage tank. Some layers with different compositions in HLW tanks might have large quantities of salt that could lead to an explosion in the absence of cooling (adapted from image in: Sengupta, Pranesh. "A Review on Immobilization of Phosphate Containing High Level Nuclear Wastes Within Glass Matrix—Present Status and Future Challenges." *Journal of Hazardous Materials, 235–236* [2012]: 17–28.)

To summarize, both historical precedent and technical plausibility leads us to be concerned about chemical explosions at HLW tanks. Although the likelihood of a cascade of events along the lines described earlier is admittedly low, it is possible that one or more explosions could rupture the tank and the other physical barriers, and disperse a substantial fraction of the fission products in the HLW into the atmosphere. The next part of the article considers how these fission products might be dispersed over southern India.

Dispersion and deposition

To calculate atmospheric dispersion of radioactivity from an explosion in a reprocessing waste tank, we use the Hybrid Single-Particle Lagrangian Integrated Trajectory model (version HYSPLIT4) developed by the Air Research Laboratory (ARL) of the National Oceanic and Atmospheric Administration (NOAA).[117] The software is in the public domain.[118]

The software simulates trajectories, dispersion, and deposition of atmospheric emissions from a source, based on meteorological data. For the following simulations we use the Global Data Assimilation System (GDAS) meteorological data provided by the National Center for Environmental Prediction (NCEP). The model tracks both advection and diffusion of virtual particles through grid cells, and concentrations are evaluated as sums of particles. The results of dispersion and deposition are displayed graphically as equi-concentration contours on a map.

The simulation assumes that an explosion takes place in a storage tank containing HLW from the reprocessing activity at KARP, expelling 10% of the radioactive contents of the tank in the form of micron size aerosols to a height of 100 m. To reflect the explosive nature of the release we assume a vertical line source from 10 to 100 m above ground level, from which the virtual particles are evenly released into the atmosphere.The release assumes about 30 PBq of cesium-137, based on an assumed 300 PBq of cesium-137 in the HLW tank.

The release takes the shape of a plume, the particles of which diffuse in the atmosphere, and advect under the prevailing wind conditions. To estimate a suitable simulation time of the plume to let most of the cesium-137 deposit, we adopted the standard assumption that the particles get mixed vertically to a height of 1000 m, what is termed the mixing layer. Since in the absence of precipitation a micron-size particle acquires a downward terminal velocity of 0.002 m/s,[119] all of the particles thrown up to the top of the mixing layer would take, on average, about 140 hours to reach the ground. Monitoring the plume for this period of time (140 hours), therefore, allows for most of the released radioactivity to deposit on the ground.[120]

The Hysplit model calculates dry and wet deposition based on meteorological data given for a specific time and location. The simulation calculates the total ground deposition for every grid point of the output data. Model results are given in kBq/m^2 based on the accumulated cesium-137 deposition.

KARP is located on the coast and meteorological conditions vary widely, leading to variations in quantities such as wind direction and velocity, vertical movement, and precipitation. This variability is evident in Figure 4, which shows the trajectories of releases from KARP for the first day of each month in 2015. As shown, about half of the trajectories have significant pathways over land, where deposition of radioactive matter could affect areas that are inhabited by people.[121]

For our detailed analysis of the impact of a release on the population, we chose the date of the release to be 1 November 2015. The total area where all the radionuclides are deposited and the area of ground deposition are shown in Figures 5 and 6 respectively. According to the simulations only about 5% of the released radioactivity is deposited on land (Figure 6). This is important because an exercise of this sort might be construed as engaging in a worst case analysis, whereas our choice of date is relatively arbitrary and leads to the vast majority of radionuclides to be deposited over the Bay of Bengal. Indeed, our simulations for other dates (1 April 2013, for example) show the plume going primarily over land in some cases, with more dire consequences.

Although the deposition pattern is sensitive to the assumptions made and the specific atmospheric dispersion model used, the total dose to, and the consequent health impact on, the population should be a relatively robust prediction. The health impact is dominated by many individuals accumulating relatively low doses of radiation. This implies that as long as the same quantity of radionuclides is dispersed over areas with similar populaton densities, the total population dose will not be sensitive to variables in the atmopheric dispersion. Of course, the health impacts to any individual or residents of an individual town or village will depend on the details of the dispersion, but the purpose of our modeling is to calculate the total

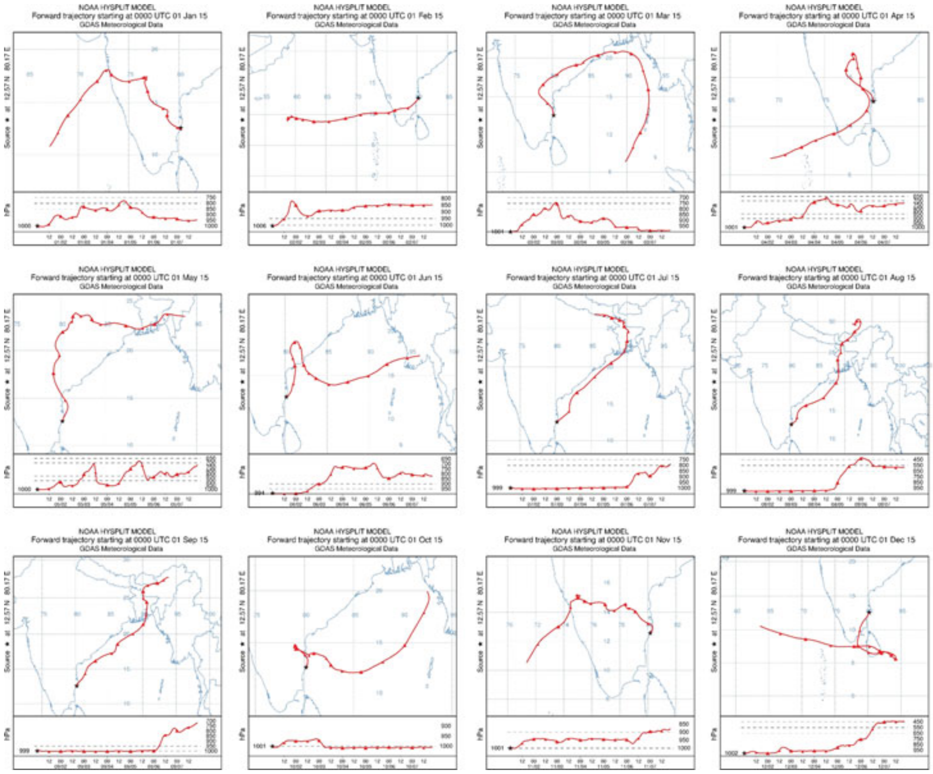

Figure 4. Ensemble of trajectory simulations for releases from Kalpakkam. For the first week of each month of 2015 the main trajectory was simulated. In more than half of the cases a significant part of the trajectory is over land. The graphs below each map show the vertical movement over time.

population exposure, not to calculate doses to specific individuals or locations. It is therefore sufficient to use average population density figures.

The radiation dose is calculated from the radioactivity level deposited on the ground (ground shine), integrated over the next 50 years. This in turn is used to estimate the excess cancer risk. Since the cloud shine and inhalation doses are very small compared to the long-term dose from ground contamination, we ignore the former contributions in our calculations.

Whole body dose from ground shine

The dose to a person living in the contaminated area is the product of the dose coefficient for cesium-137, as reduced due to the shielding from houses and other structures in the area, and the gradual weathering and submersion of the radionuclides in the soil. The last factor has been empirically fitted to a two-exponential decay depicting a rapid and then a relatively slow submersion as well as the radioactive decay of cesium-137:[122]

$$r(t) = 0.57 * e^{-\left(\frac{ln2}{2.4}+\frac{ln2}{30}\right)t} + 0.58 * e^{-\left(\frac{ln2}{37}+\frac{ln2}{30}\right)t}$$

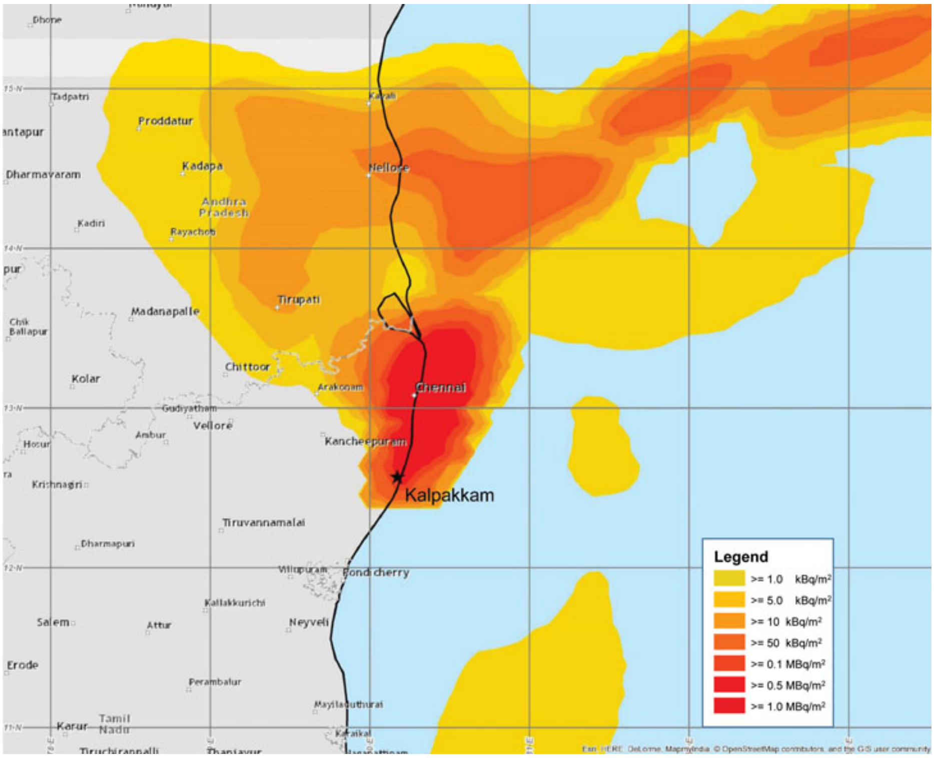

Figure 5. Deposition due to a radioactive release of 30 PBq of cesium-137 from a HLW tank at Kalpakkam.

which integrates over 50 years, after rounding, to 14 years. In the formula, the numbers 2.4 and 37 are empirically determined half-lives due to weathering and the 30 represents the radioactive half-life of cesium-137. All these half-lives are measured in years. The dose coefficient for cesium-137 is 17.35 (mSv/y)/(MBq/m^2).[123] Shielding affects this number significantly. Values for shielding are strongly dependent on the environment (rural or urban), and estimates in the literature vary widely.[124] We choose shielding factors reported in the literature specific to India. These suggest shielding factors of 0.3 for brick/concrete houses, 0.5 for brick/tile houses, 0.8 for mud/leaf houses, and 1 for leaf/leaf houses.[125] There are no published numbers for shielding in urban areas in India, but urban doses are expected to be lower. Therefore, we assume an average shielding factor of 0.7 for rural areas, which are defined here as areas with less than 500 people per square kilometer, and an average shielding factor of 0.4 for urban areas with more than 500 people per square kilometer. Note that these are average figures meant to account for variations in the kinds of houses and workplaces occupied by people with different lifestyles and schedules. With these assumed shielding factors, the fifty year doses per unit of cesium-137 contamination to people in rural and urban areas will be 170 mSv/(MBq/m^2) and 97 mSv/(MBq/m^2) respectively.

In the scenario chosen for elaboration, the highest contamination levels are found over Chennai, which is also densely populated with a total of about 9 million people

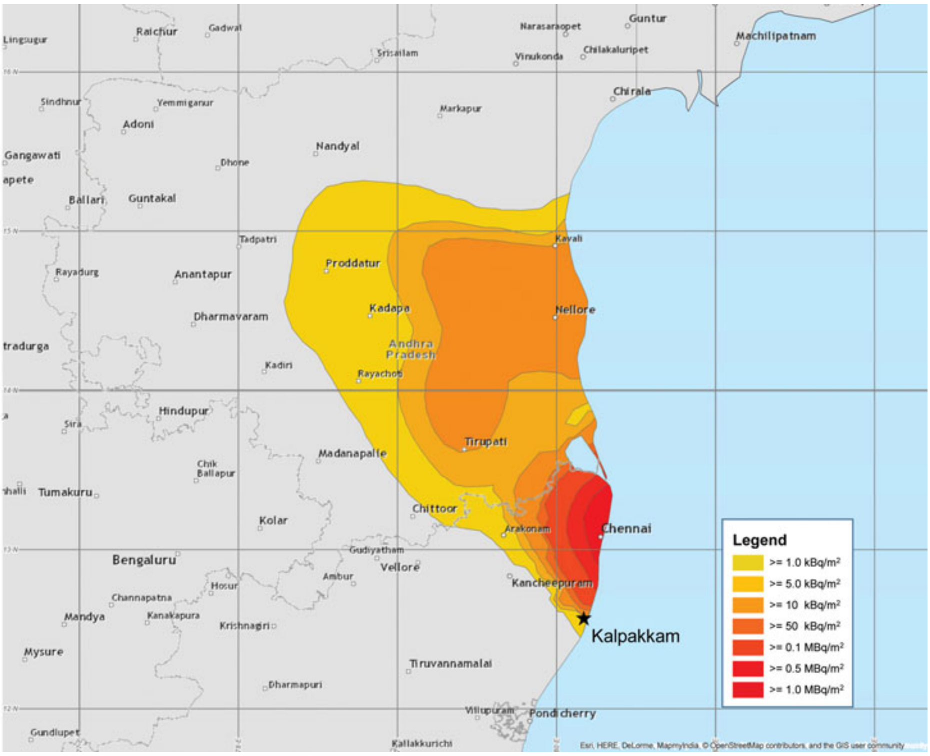

Figure 6. Map of deposition due to a radioactive release of 30 PBq of cesium-137 from a HLW tank at Kalpakkam.

in its greater metropolitan area. Following a 30 PBq release on 1 November 2015, nearly 4.2 million people would be living in areas with high contamination levels of more than 1 MBq/m^2, which is roughly the evacuation criterion the Japanese government used to permanently relocate residents following the Fukushima accident.[126] Chennai's closeness to KARP makes it a likely fallout zone when the wind is blowing north. Since KARP is only 70 km away from Chennai, it would take a plume from KARP 3 to 5 hours with average wind speeds of only 4–6 m/s[127] to reach the city, hardly enough time to evacuate a significant fraction of the population. Should some fraction of this population be evacuated, there would be major societal and economical costs. For the purposes of this analysis, we assume that such large-scale evacuation is not attempted and calculate the total radiation dose to the population and the resulting cancer mortality figures.

Based on the factors listed above and the results from Hysplit of the quantities of cesium-137 deposited over South India, we estimate that a 30 PBq release at KARP on 1 November 2015 would lead to a total population radiation dose of 920,000 person-Sv. The main contributor to this value is the population dose from the urban area of Chennai with 900,000 person-Sv. Taking into account the trajectories shown in Figure 4, other scenarios are conceivable where the major population dose comes from exposure to inhabitants of rural areas.

Table 2. Biological Effects of Ionizing Radiation (BEIR) Committee estimate of number of cancer cases and deaths expected to result from 100,000 persons (with an age distribution similar to the U.S. population) exposed to 100 mSv. The estimates are accompanied by 95% subjective confidence intervals shown in parentheses that reflect the most important uncertainty sources to a U.S. population.

	All Solid Cancers		Leukemia	
	Males	Females	Males	Females
Excess cancer cases	800 (400–1600)	1300 (690–2500)	100 (30–300)	70 (20–250)
Excess cancer deaths	410 (200–830)	610 (300–1200)	70 (20–220)	50 (10–190)

The overall dose is nearly twenty times the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) estimate of 48,000 person-Sv for the collective dose to the population of Japan from the multiple accidents at the Fukushima Daiichi nuclear reactors.[128]

Table 2 lists the estimates of cancer incidence and deaths resulting from exposure to 10^4 person-Sv according to the Biological Effects of Ionizing Radiation (BEIR) VII study carried out by the U.S. National Academy of Sciences.[129] The number of deaths that would result from cancer incidence, of course, depends on the quality of health care, and must be viewed as less definitive when compared to the estimate of cancer incidence. In the accident scenario that we have sketched out here, the radioactive release of 30 PBq of cesium-137 from a HLW tank at KARP under the metereological conditions assumed could give rise to a collective population dose of 920,000 person-Sv and this could result in nearly 97,000 excess cancers that would develop over decades in the exposed population. Of these, nearly 47,000 cases might be fatal. The population dose is linearly dependent on the assumed release.

As shown in Table 2 there are considerable uncertainties in the estimates of cancer incidence and mortality from radiation exposure. These uncertainties add to the earlier-mentioned uncertainties concerning deposition patterns and shielding. The result is that there are significant uncertainties in the number of people who would develop and die from cancer. However, by and large, we have erred on the side of choosing parameters that underestimate rather than overestimate the number of deaths.

Over the last decade, there is also increasing evidence for other, non-cancer, health impacts, fatal in many cases, that result from exposure to even low levels of radiation. In particular, recent reviews suggest that the excess mortality from all circulatory diseases linked to exposure from low and moderate doses of radiation could be of the same order of magnitude as from cancers.[130]

Conclusion

Reprocessing plants and associated facilities have large and concentrated radioactive inventories that can be accidentally dispersed into the atmosphere. This article focuses on HLW storage tanks that are susceptible to a variety of chemical explosions, resulting in possible tank rupture and the release of radioactive material.

Even though HLW tanks are protected by a variety of safety systems, it is conceivable that all the safety systems could fail under some circumstances. Further, tanks, as opposed to reactors, are usually not located inside containment structures, making it likely that any escape of radioactivity will lead to dispersal. Exploring the impact of such an accident might be considered a low probability, worst-case scenario.[131] But, as sociologist Lee Clarke argues, there is value in engaging in constructing such scenarios because disasters, even worst-case ones, are a part of life.[132]

We modeled an accident scenario at KARP, located 65 km south of the city of Chennai (Madras). A considerable amount of HLW has likely accumulated in the KARP storage tanks due to the fifteen year delay in the commissioning of the associated vitrification plant. KARP is also of concern because of management issues. In 2003, there was a valve failure at KARP which led to what its management termed the "worst ever radiation exposure incident" in the history of nuclear power in India.[133] Analyzing that incident in more detail shows that the DAE had violated many of the recommendations offered by most safety theorists, including lack of redundancy, a relatively low level of importance offered to safety, and the absence of a healthy relationship between management and workers.[134]

Modeling the atmospheric dispersion of a radioactive release, assumed to be 10% of the inventory of just one HLW tank at KARP, shows that such an accident could have serious consequences for public health, with nearly 47,000 cancer related deaths and the contamination of large areas for decades. Roughly an equal number of people would develop non-fatal cancers.

These estimates of radiation exposure and contamination result solely from an exposure to cesium-137 from long-term ground contamination. In an actual release there would be additional exposure to radiation from other radionuclides deposited on the ground, from inhalation when the radioactive cloud passes over the population as well as from exposure to gamma radiation from the radioactive cloud. While the dose from ground contamination due to cesium-137 is expected to be the leading term, the doses from other radionuclides are likely to be significant as well. We ignore these exposures to produce conservative (closer to a lower bound) estimates of the number of cancers and deaths.

After Chernobyl, the area that was defined as contaminated and agriculture banned, because it contained cesium-137 in excess of 37 kBq/m^2, extended to more than 146,000 km^2 in Belarus, Ukraine, and the Russian Federation alone.[135] The corresponding areas in the scenario described above is about 3,500 km^2. The large difference is likely due to the larger release of cesium-137 (85 PBq) from Chernobyl compared to the 30 PBq in the scenario we have assumed and the nature of the dispersion on the day chosen for our scenario. The latter choice results in most of the cesium-137 in this scenario being deposited in the Bay of Bengal, and the fallout pattern that is relatively concerntrated in a relatively small area. However, this area is a center of financial activity and this deposition could lead to considerable economic losses. Other release dates may lead to, for example, a lower total population dose but larger contaminated areas.

Acknowledgments

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