CHAPTER 7: A SURVEY OF DECONTAMINATION METHODS

[redacted]

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1. Introduction

This chapter is an attempt to survey the published literature relevant to contamination problems and decontamination solutions proposed for dealing with them. Priority has been given to contained explosions for the stimulation or storage of gas and oil. Some consideration has also been given to the entirely different problems arising from uncontained explosions.

The amount of literature devoted to decontamination in PNE projects is small compared with that dealing with other aspects such as seismic effects and radiological measurements. However, adequate decontamination is imperative if effective use is to be made of PNE.

Projects which are or have been considered fall into three main categories from the point of view of decontamination:-

(a) Fully contained explosions for the release of natural resources (minerals, oil or gas) or for the creation of storage facilities.

(b) Partially vented "retarcs" for quarrying or overburden removal.

(c) Cratering explosions for constructional purposes (dams, harbours, canals, reservoirs and roads or railways).

It is extremely unlikely that projects of types (b) and (c) would be considered feasible in densely populated areas such as Western Europe. However, contained cavities might be created underground or, more probably, under the sea-bed in such densely populated areas. Therefore contained explosions are considered first in this study of decontamination problems.

2. Decontamination of Storage Cavities

Storage can be provided in a stable, roughly spherical cavity in a salt bed or in the collapsed chimney formed by a deeply buried nuclear explosion in an impermeable rock formation. To a limited extent, it may be possible to choose the site to minimise [redacted] contamination by selecting a rock formation with a low [redacted] content. [redacted] suggests that a rock free of solid carbon and relatively rich in carbonates would reduce the amount of [redacted] found in the hydrocarbons in a Gasbuggy-type explosion and this may also apply in a storage application.

The cavity or chimney would not be re-entered until several months after the explosion to allow for appreciable decay of the total radioactivity. The main nuclides left in the gaseous phase would be [redacted]. Unfortunately these are the most difficult to remove and might have to be vented to the atmosphere. There are already significant amounts of these gases in the atmosphere from weapons tests and nuclear power production. Decontamination, if a suitable method can be found, would be infinitely preferable. [redacted] recommends a [redacted] for a gas storage cavity presumably to minimise [redacted] but this would be at the expense of much greater [redacted] (see Chapter 1).

The cavity/chimney would probably be re-entered through the original emplacement hole and, for oil storage, a second hole would be drilled and whipstocked to the base of the cavity/chimney. This would provide a circuit for flushing out the storage space as well as for storage and recovery. Water or gas could be used to flush the storage space. Water flushing would remove much of the HTO and some soluble fission products; HT and [redacted] must be displaced either by the gas or the water. In the absence of a second entry to the storage space repeated venting, evacuation and re-pressurization through the re-entry hole would be required unless a concentric system of pipes was used.

Although the puddle material from explosions in granite is apparently resistant to leaching, that from an explosion in salt is not (see Chapter 4). Glasses formed in a carbonate (eg, limestone) region might also be susceptible to leaching since calcium and magnesium oxides react readily with water. As in a salt cavity, the radioactive contaminants might become mobile even if they were not actually dissolved. Oil storage will most probably be near the coast and sea-water will be used for flushing and as the driving medium to recover the stored oil. Therefore the glass puddle will be permanently in contact with water but the dissolved activity should remain in the water and not contaminate the oil. Monitoring of the sea-water as it passes out of the storage area will be essential and some decontamination may be necessary.

Because of uncertainties about puddle behaviour, storage in dry rock with non-aqueous purging may be preferred especially for gas storage. [redacted] has studied the creation of gas storage facilities in France. He proposes a [redacted] explosion in plastic clayey soil to give a stable cavity. He predicts that [redacted] would be formed, [redacted]. After 2 months only [redacted] and [redacted] would be significant. This would be removed by repeated evacuation and pressurisation with gas which would also remove the H_2, [redacted] formed in the explosion. The subsequently stored gas will be saturated with water vapour as it leaves the cavity. Dehydration in a standard plant should remove 98% of this water leaving [redacted] in the gas. A further [redacted] will be present from the residual gases after purging. These latter activities will be reduced by dilution as the storage facility is used.

Early access to the cavity can take advantage of the residual over-pressure from the explosion for venting but, overall, the short-lived activity present makes a delay of some months preferable. When the gas is released to the atmosphere, some plant treatment, such as that designed for Project Dribble, would be desirable ([redacted] This consisted of a remotely operated gas-scrubbing and filtering system with a provision for the storage of liquid wastes for subsequent disposal. [redacted] has suggested evacuation by pumping and [redacted] suggest displacement by a denser fluid for the removal of the residual gases.

Cycling of natural gas through the cavity with removal of tritiated water before re-injection would reduce the residual [redacted]. After some tens of cycles, pressurisation to the storage capacity would reduce the HTO concentration in the gas to a permissible level. Rotary pumps capable of handling gas volumes in excess of 2.8×10^4 cm^3/day at high vacuum are available and could be used to pump the cavity. As the pressure is lowered, HTO will be vaporised and the discharge from the pump would be dehydrated to remove the [redacted]. Residual free [redacted] (and other non-condensable tritiated molecules) could be oxidized to HTO by catalytic methods and removed in a second dehydration stage.

[redacted], using aircraft oxygen tanks and tracers, has shown that continuous purging is superior to pressure cycling and that a single access hole using concentric pipes can be employed.

3. Product Decontamination following Stimulation of Gas and Oil Wells

3.1 Gas

Only a limited amount of data is available on the actual decontamination procedures relating to nuclearly stimulated gas or oil wells. [redacted] dismisses the subject rather lightly but at the same conference [redacted] did offer some specific solutions in relation to the Rulison event. The first, dilution with uncontaminated gas, evades the issue and does not deal with the problem of the highly contaminated gas present at re-entry. It is undesirable and wasteful simply to flare this gas to the atmosphere. A second solution is the use of the gas for controlled industrial processes, eg, electricity generation, fuel or chemical production, with only occupational exposures to the gas. [redacted] carries this a step further; he suggests the synthesis of ammonia from the gas. The [redacted] would then be removed from the product during the normal processing required to remove Ar, CO_2 and residual methane. Some inactive [redacted] might be necessary as a carrier. The [redacted] would be removed from the gas by exchange into the wash water used, after the conversion of the gas to CO_2 and H_2, to remove the CO_2 and before the reaction with nitrogen.

[redacted] has suggested that the more highly contaminated gas is withdrawn quickly and stored in another geologic horizon, eg, a sub-surface aquifer, assuming that such is available. Experience at Rulison, Chapter 4, section 10, showed that the rapid removal of 2 chimney volumes reduced the radionuclide concentrations in the gas by a factor of [redacted] which may be sufficient.

[redacted] has considered the third possibility, true decontamination of gas, in some detail and discusses exchange reactions, with a suitable catalyst, for the removal of [redacted] etc) into a counter current of steam or water. Exchange reactions into ethylene glycol, which is used in gas drying, might also be effective. The effect of scrubbing the gas with ammonia or hydrogen sulphide could also be investigated. [redacted] found that passage over certain sandstones, clays and shales reduced the tritium content of water.

Several methods for the removal of [redacted] and xenon from reactors and fuel re-processing effluents and radon from uranium mines were discussed at a recent symposium on the noble gases at Las Vegas on 24 - 28 September 1973 (sponsored by the National Environmental Research Center). [redacted] presented data showing that xenon and radon could be removed from a gas stream very efficiently by reaction with the dioxengyl salt, O_2SbF_{11} or the fluoronitrogen salt, [redacted] One problem, the need for dry gas to prevent hydrolysis of the salt, would be already covered in gas stimulation by the need to remove [redacted] So far no effective compound for fixing [redacted] has been developed. However, a stable complex [redacted] is known. [redacted] suggested that AuF_6 might be sufficiently oxidising for the spontaneous fixation of krypton.

At this same symposium [redacted] discussed the installation and testing of a treatment system for a power station using zeolites and activated carbon to retain the noble gases. Several other authors discussed the adsorption characteristics of these gases, namely [redacted]. and [redacted] Cryogenic methods were considered by [redacted] suggested that the recovered [redacted] could be used for power independent light sources rather than simply stored.

Certainly methods for removing and concentrating [redacted] are available but whether they can be applied economically to gas from a nuclearly stimulated well has not been examined.

3.2 Oil

The Russians have some direct experience of oil stimulation (see Chapter 4) but no data on their decontamination methods is available. In the USA the main interest has been in "in situ" retorting from oil shale beds. So far only laboratory tests have been carried out by [redacted] Some difficulties may be experienced since [redacted] of the shale hydrocarbons seems to occur during retorting. Prior removal of the [redacted] by contacting with water vapour for considerable periods may be necessary. [redacted], etc, did not enter the oil and could be removed by filtration or distillation.

4. Surface Cratering

The amount of earth moved by the [redacted] Sedan event (1962) was estimated at [redacted] of which some [redacted] were distributed around the crater and up to some [redacted] from it. On this bulk ejecta was superimposed the missile ejecta and fallout which extended to a significant depth up to [redacted] from the crater. The areas involved are therefore potentially very large, up to [redacted]. So far only natural processes, radioactive decay and weathering have operated to reduce the activity in this earth.

There are three distinct hazards in the vicinity of a nuclear crater: the β-γ radiation field from any fission and activation products, the inhalation on ingestion of re-suspended contaminated soil (α-, β- or γ-radiation) and the special case of [redacted] absorption on transport through water in the area.

No real proposals for dealing with the first two hazards have been made. Device design and delay to permit decay have been used to reduce them. If earth moving operations were required to decontaminate or shield an area, they would be on a massive scale and would probably render the project uneconomic.

[redacted] has reported some decontamination associated with the Palaquin event but this only relates to standard procedures for personnel protection and wash down of equipment.

Many of the proposed applications required that the craters be filled with water which will inevitably be contaminated to some degree. Depending on the precise use, some form of dilution or chemical treatment might be required to deal with the fission products. Since most of these are readily retained by ion exchange on soil particles, some form of filter bed might be adequate. [redacted] water cannot be held up to any great extent in this way. Dilution, controlled pumping or controlled evapo-transpiration techniques might reduce the [redacted] to acceptable levels in the crater water but, as a whole, this [redacted] will be disposed of to the ecosphere. Therefore, true decontamination with subsequent controlled storage of the contaminant will not be attained.

5. Retarce

The depth of burial can be chosen so that, although no material is ejected, the rock overlying the crater is fractured and a mound is produced due to the increased volume of the broken rock. This rock can then be quarried for use in dams, road or rail beds, or for mineral recovery. Alternatively, by suitable siting of the explosion a dam may be created in situ.

The Sulky event did not crater and only a small dust cloud of relatively low activity was formed. This was followed by a continuous stream of more radioactive gas which flowed for some time. This outflow should reduce the amount of activity remaining on the broken rock considerably compared with a contained chimney as the main volatiles, [redacted], would be considerably reduced. [redacted] suggests that, after the usual delay (∿ 6 months) for the short-lived isotopes to decay, the aggregate could be quarried by some remotely controlled technique and transferred by conveyor belt through a washing stage or a leaching stage if mineral recovery was the object. Until some practical work has been performed on actual rubble, the residual levels of activity cannot be predicted.

For applications such as rock fill in dams or roads, there would be a considerable amount of self-shielding once the material was emplaced and any final dressing or surface sealing would provide additional shielding so decontamination is of less importance. Close control of the occupational workers handling the aggregate and their equipment would still be necessary.

6. Mineral Extraction

The main possibilities for PNE in this field have been reviewed by [redacted] and can be divided into three categories, two of which require a contained chimney. These are in situ leaching of a contained chimney which will probably not give as high a recovery as the alternative of block caving the chimney and heap leaching the recovered ore. The third method involves the removal of overburden or break-up of the ore by a nuclear explosion for open-cast mining.

In situ leaching has been studied principally in relation to copper recovery because favourable ore formations have already been located. [redacted] mention that [redacted] water in the leach liquor will be by far the most important contaminant [redacted]. Experimental work by these authors and [redacted] using rubble from two Nevada test site shots, has shown that ∿ 80% of [redacted] water can be displaced from a rubble bed in the first 0.4 bed volume of effluent. An initial washing of the chimney with water could remove a large proportion of the tritium without any loss of copper values. This wash water could then be disposed of separately by controlled evaporation, diluted or stored. All the other radionuclides were separated from the copper during processing, except Ru-106, which could be removed by electolytic purification of the copper. Certain precautions would have to be taken with regard to operating personnel and disposal of waste liquors but special shielding of processing tanks above ground are not envisaged.

Block caving and overburden removal present more severe problems since the radioactive rock and soil in the chimney or retarc will have to be moved. [redacted] based their approach to the removal of overburden on an unspecified period for decay, followed by remote controlled working using operators inside shielded air-conditioned vehicles. As with craters, no large scale decontamination seems feasible. Sufficiently careful control of occupational workers and subsequent decontamination during the processing of the product is the best that can be achieved.

GLOSSARY

ABSORBED DOSE (rad, rd). The energy imparted by ionizing radiation to a unit mass of material is the absorbed dose and is equal to $\Delta E_D/\Delta m$ where E_D is the energy imparted to the volume containing the mass Δm. E_D is equal to the sum of the energies of the radiation entering the volume minus the sum of the energies of the radiation leaving the volume and minus the energy equivalent to any increase in rest mass created by nuclear or elementary particle reactions occurring in the volume.

$$1 \text{ rad} = 10^{-2} \text{ J kg}^{-1} = 100 \text{ erg g}^{-1}.$$

Adsorbed dose rate is expressed in rad s^{-1} etc.

ABSORBED DOSE AND EXPOSURE (qv). The energy absorption per roentgen depends on the material and the energy of the radiation and is equal to

$$\frac{\text{total charge produced}}{\text{charge on an electron}} \times \text{energy required to produce an ion pair.}$$

This last is approximately constant at 5.4×10^{-18} J (33.7 eV). One roentgen produces the same energy absorption as 0.87 rad in air, 0.97 rad in water and 0.92 rad in bone for 1 MeV radiation. At 30 keV 1 roentgen is equivalent to 4.32 rad in bone.

For many radiological protection purposes it is sufficient to assume that 1 roentgen produces the same energy absorption in the body as 1 rad.

ACTIVITY (curie, Ci). The activity of a specified amount of radioactive material is the number of nuclear transformations that occur in that amount per unit time. The unit of activity is the curie.

$$1 \text{ Ci} = 3.7 \times 10^{10} \text{ nuclear transformations per second.}$$

ALLUVIUM. Compacted particles of rock, sand, clay, etc, deposited from water over long periods of time. Much of the earth's surface is covered with alluvium.

α-PARTICLES (α-rays). These consist of two protons and two neutrons (the helium nucleus) and are positively charged. They are emitted with a characteristic energy for any particular nuclide which is generally of a high atomic number. Their penetrating power is small (< 0.1 mm) but dense ionization occurs along this short track.

AQUIFER. An underground rock structure through which water moves.

β-PARTICLES (β-rays). A charged particle emitted from an atomic nucleus with a mass and charge equal in magnitude to that of an electron. Their range in tissue is usually less than 2 cm and the density of ionization along the track is less than that for α-particles. They are emitted in a continuous spectrum of energies up to a maximum energy which is characteristic of the parent nuclide. The average energy is approximately one-third of the maximum energy.

CAVITY. An expanding sphere of material vaporized by the shock wave and heat from an underground nuclear detonation; at this stage the cavity is known as the vaporization cavity. The stable cavity results when pressure in the cavity decreases and expansion stops. The radius of the stable cavity is about seven times that of the vaporization cavity.

CHIMNEY. A tall, roughly cylindrical volume of broken rock and rubble formed by the collapse of the overlying medium (over-burden) into the cavity produced by an underground explosion.

CLEAN NUCLEAR EXPLOSIVE. A nuclear device which produces minimum amounts of biologically significant isotopes for a particular application in the Plowshare programme.

CONTAINED EXPLOSION. An explosion at such a depth underground that there should be no significant escape of radioactive material to the atmosphere.

CRATER. A depression in the ground left by some underground nuclear explosions. A throwout crater results from the expulsion of ground material by the expanding gases in the explosion cavity. A subsidence crater may be formed if the collapse of the chimney material reaches the ground surface.

DOSE DISTRIBUTION FACTOR. This factor must be included in calculations of dose equivalent to account for such effects as the preferential deposition of strontium in bone or iodine in the thyroid gland. Other specific effects of certain nuclides could also be included in this factor.

DOSE EQUIVALENT (roentgen equivalent man, rem). This is equal to the absorbed dose multiplied by certain modifying factors which take account of the effectiveness of the radiation in inducing ionization and the non-uniform distribution of some internally deposited nuclides, etc.

Dose equivalent = dose × quality factor (qv) × dose distribution factor (qv).

EXPOSURE (roentgen, R). This applies to X- or γ-radiation in air and is equal to $\Delta Q/\Delta m$ where ΔQ is the sum of all the electrical charges of one sign produced when all the electrons liberated by the photons are completely stopped in a volume of air containing mass Δm.

$$1 \text{ roentgen} = 2.58 \times 10^{-4} \text{ C kg}^{-1} \ (\approx 2 \times 19^{9} \text{ ions cm}^{-3}).$$

Exposure to 1 roentgen of X- or γ-rays results in the absorption of 87 ergs g^{-1} of air. In substances of different atomic numbers and densities the energy absorbed per unit volume for the same quantity of radiation will be different. For soft tissue the energy absorbed per gram is 98 ergs.

Exposure rate is pressed in R s^{-1}, etc.

FALLOUT. The gradual return of particles to the ground from the clouds (main cloud and base surge) formed in a cratering explosion. Also the material itself. Local (or early) fallout occurs downwind from the explosion site within about 1 day after the detonation. Delayed fallout consists of very small particles which ascend to high altitudes and return to earth slowly over a very large area. Fallout sector is the area over which local fallout is predicted from wind speeds and directions from the surface to the top of the cloud.

FISSION. The splitting of the nucleus of a heavy atom into nuclei of two lighter atoms, with an accompanying decrease in mass. The loss of mass results in the release of a large amount of energy. Fission is usually caused by the absorption of a neutron by the nucleus. The important fissile nuclei are uranium-235 and plutonium-239.

FISSION PRODUCTS. A general term for the complex mixture of substances produced from nuclear fission. Approximately 80 different primary fission fragments result directly from some 40 different modes of fission. These radioactive fission fragments through decay produce additional (daughter) products; the resulting complex mixture of fission products contains about 200 different isotopes of more than 35 elements.

FRACTIONATION. A sample containing fission products is said to be fractionated when the relative proportions of the fission products differ from those expected from the fission process. The degree of fractionation is determined in relation to one particular fission product. In the past Mo-99 has generally been taken as the reference nuclide for underground explosions but Zr-95 or Nd-147 may also be used.

Degree of fractionation of nuclide X

$$= \frac{\text{atoms of X (sample)/atoms of Mo-99 (sample)}}{\text{expected ratio of atoms X/Mo-99}}.$$

Frequently the data may not lend itself to more than a qualitative estimation, in which case a particular nuclide may be classed as enriched or depleted with respect to the reference nuclide.

FUSION. The combining (or fusing) of two very light nuclei to form a nucleus of a heavier atom. There is a net loss of mass which results in the release of a large amount of energy. Deuterium and tritium, the isotopes of hydrogen, are commonly used in this process.

γ-RAYS. These are short wavelength electromagnetic radiations of nuclear origin. They are emitted with energies characteristic of the parent nuclide in the range 10 keV to 9 MeV. They penetrate much more deeply than α- or β-rays and their intensity decreases exponentially with the thickness of the absorbing material. Typically deep-seated organs in the body receive a dose from a X-ray source which is 60% of that received by the skin.

GROUNDWATER. A natural underground water system.

HALF-LIFE ($t_{\frac{1}{2}}$). The time for the radioactivity (the emission of α- or β-particles or γ-rays) of a quantity of an isotope to decay to half its original value. The effective half-life of an isotope is the time required for the radioactivity of that isotope in the body (or in an organ) to decrease to half its original value through both decay and biological elimination. The effective half-life is also applied in this survey to the radioactivity of iodine in cows' milk.

IONIC RETARDATION. Delay in the flow of dissolved ions due to absorption or exchange at a surface.

ISOTOPES. Forms of the same element having identical chemical properties but differing in their atomic masses (because of different numbers of neutrons but the same numbers of protons in their respective nuclei) and in their nuclear properties (eg, radioactivity and fission).

KILOTON (kton). One thousand tons; used to express energy released in a nuclear explosion in terms of the energy released originally by the corresponding weight of TNT and now defined as 10^{12} cal.

LITHOSTATIC PRESSURE. The pressure due to the weight of the earth's crust above any point within it.

MEGATON (Mton). One million tons or one thousand kilotons; used to express the energy released in a nuclear explosion.

NUCLIDE. A specific isotope of an element defined in the text by its chemical symbol, its mass number and its energy state, eg, Tc-99m, Sr-90, U-235. A radionuclide is a nuclide which undergoes spontaneous radioactive decay.

PLOWSHARE. A programme of the US Atomic Energy Commission for the research and development of peaceful uses of nuclear explosives. The name comes from the Bible: Isaiah 2.4 ".... and they shall beat their swords into plowshares."

QUALITY FACTOR (formerly relative biological effectiveness, RBE). This is defined as the ratio of the dose required by irradiation with 250 kV X-rays to produce a certain biological effect, to the dose required by irradiation with the radiation under consideration to produce the same biological effect. Some recommended values are:-

	QF
β-, γ-, and X-radiation	1
Thermal neutrons	2.5
Fast neutrons	10
Protons	10
Naturally occurring α-radiation	10
Heavy recoil nuclei	20

RADIOACTIVITY. The spontaneous emission of radiation, generally α- or β-particles, often accompanied by γ-rays, from the nuclei of an (unstable) isotope. As a result of this emission, the radioactive nucleus changes (decays) into a nucleus of an isotope of a different element, which may or may not be radioactive. Ultimately, as a result of decay, a stable (non-radioactive) end product is formed.

REFRACTORY FISSION PRODUCTS. All nuclides formed in the fission process decay by β/γ-emission with no change in atomic mass, only a change in atomic number, until a stable isotope is attained. Only a very small amount (if any) of the radionuclide finally measured is formed during fission and, depending on their half-lives, the chemical properties of its precursors in the mass chain will govern the apparent condensation behaviour and the final distribution of that nuclide in the post-shot debris. Refractory fission products are those measured radionuclides whose precursors have stable oxides at high temperatures which condense early and condense readily into the molten rock. Zr-95 is a typical example; its mass chain is as follows:-

[redacted]

Other examples are Ce-143, Ce-144 and Nd-147.

SCALED DEPTH OF BURIAL. The actual depth of burial (in feet) of an underground nuclear device divided by the cube root of the energy yield of the explosion (in kilotons TNT equivalent). It provides a comparison of the effective depths of burial for explosions of different yields. For a contained explosion, the scaled depth of burial must exceed a value appropriate to the yield.

STEMMING. The plugging of a hole or tunnel, in which a nuclear detonation device has been placed, to prevent the escape of radioactivity into the atmosphere.

TUFF. Various types of consolidated particulate material of volcanic origin.

VENTING. The prompt escape to the atmosphere of gases and solid residues from an underground explosion.

VESICULATE. Form blisters.

VOLATILE FISSION PRODUCTS. These are the measured fission products which have volatile or gaseous precursors (see refractory fission products) with half-lives comparable with the times involved in the condensation processes. Therefore they do not condense readily into the molten rock and are generally enhanced in the chimney rubble, crater ejecta and fallout. One of the most volatile fission products is Sr-89:-

[redacted]

Other examples are Sr-90, Cs-137 and Ba-140.

Intermediate behaviour is observed in radionuclides whose precursors are not particularly volatile but do not form stable oxides and so are less readily condensed. Ru-103, Ru-106 and Te-132 are typical of this group.

X-RAYS. Electromagnetic radiation resulting from the rearrangement of the electron configuration within the atom. The energy is characteristic of the atom in which it was produced (ie, the daughter nuclide in radioactive decay). X-rays are identical with γ-rays except in their mode of formation and are of a lower energy.

YIELD. The total effective energy release in a nuclear explosion. It is usually expressed in terms of the equivalent tonnage of TNT required to produce the same explosive energy release.

BIBLIOGRAPHY AND REFERENCES

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Reference B

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