Working Paper

The Danish Inventory of radioactive waste and the required repository type

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Ing.(grad.) Gerhard Schmidt

Öko-Institut e.V. / Oeko-Institut e.V.

Geschäftsstelle Freiburg / Freiburg Head Office
Postfach / P.O. Box 17 71
D-79017 Freiburg. Deutschland / Germany
Tel.: +49 761 45295-0
Fax: +49 761 45295-288

Büro Darmstadt / Darmstadt Office
Rheinstraße 95
D-64295 Darmstadt. Deutschland / Germany
Tel.: +49 6151 8191-0
Fax: +49 6151 8191-133

Büro Berlin / Berlin Office
Schicklerstraße 5-7
D-10179 Berlin. Deutschland / Germany
Tel.: +49 30 405085-0
Fax: +49 30 405085-388

info@oeko.de
www.oeko.de

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Ing.(grad.) Gerhard Schmidt
Division on Nuclear Engineering and Facility Safety

Öko-Institut
Rheinstrasse 95
D-64295 Darmstadt
Phone +49-6151-8191-107, mail g.schmidt (at) oeko.de

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Zusammenfassung

Dänemark verfügt über ein geringes Inventar an radioaktiven Abfällen. Da in Dänemark keine kommerziellen Kernkraftwerke errichtet und betrieben wurden, resultiert dieses Inventar aus verschiedenen Forschungsaktivitäten.

Um mit diesen Abfällen langfristig umzugehen, hat die dänische Regierung angeordnet, die Abfälle zu beschreiben und mögliche Optionen für deren Verbleib zu untersuchen. Basierend auf vagen Kriterien wurden die meisten Abfallarten als „kurzlebig“ und als geeignet für eine oberflächennahe Deponierung bezeichnet. Die Regierung hat dann den geologischen Dienst GEUS damit beauftragt, Dänemark nach potentiell geeigneten Standorten für ein oberflächennahes Endlager abzuscannen. Als „geeignet“ wurden dabei Standorte in 0 bis 100 m Tiefe definiert. Weder wurden Isolationseigenschaften oder andere Anforderungen an geologische Schichten gestellt noch wurden diese Eignungskriterien in einem erweiterten Kreis abgestimmt (Experten, Öffentlichkeit). GEUS hat eine größere Anzahl geeigneter Standorte identifiziert und daraus sechs als vielversprechend ausgewählt.

In diesem Papier wird die Grundentscheidung analysiert, für die meisten Abfallarten die oberflächennahe Deponierung zu wählen. Als zentrales Kriterium für die Eignung der Abfallarten für die oberflächennahe Deponierung wird deren radioaktiver Zerfall über die kommenden 300 Jahre unter heute geltende Freigabekriterien definiert. Die Ergebnisse zeigen, dass keine der dänischen Abfallarten dieses einfache Kriterium erfüllt. Alle liegen in diesem Zeitraum über der Freigabegrenze, die meisten von ihnen um mehrere Größenordnungen und über wesentlich längere Zeiträume wie 100.000 Jahre und länger.

Die grundlegende Annahme bei der durchgeführten Standortauswahl, es sei nach oberflächennahen Standorten für kurzlebige Abfälle zu suchen, erweist sich daher als fehlerhaft. Der gesamte Prozess sollte daher erneut durchgeführt werden und auf der Basis, dass der Langzeiteinschluss in undurchlässigen Schichten zu garantieren ist. Die Eignungskriterien sollten sich auf den Langzeiteinschluss konzentrieren und sollten im Vorhinein abgestimmt sein.

Abstract

Denmark has a relatively small inventory of radioactive wastes. As Denmark never built and operated nuclear power plants, the wastes resulted only from various research activities.

In order to manage those wastes, the Danish Government has ordered to describe those wastes and the available management options. Based on vague criteria, most of the waste types were termed as “short-lived” and as suitable for a surface-near disposal facility. The Government then ordered the Geological survey organization of Denmark, GEUS, to scan Denmark for suitable locations. “Suitable” depth was defined as 0 to 100 m below ground. Neither were isolation properties or other requirements for geological layers defined nor were those criteria agreed in a broader sense (with experts, with the public). GEUS identified a number of potentially suitable locations and selected six of those as the most promising.

In this paper the basic decision of preferring surface-near disposal for most of the waste types is analysed. As a central criterion for the suitability of the waste types for surface-near disposal is defined that those waste types decay within 300 years to below today’s clearance levels. The results show, that none of the Danish types of waste meets this simple requirement. All are above

that criterion, most of them by several orders of magnitude and over very much longer times such as 100.000 years or even longer.

The basic assumption of the performed site selection procedure, to search for near-surface locations for short-lived wastes, so proves to be invalid. The whole process should be re-done on the basis that the long-term isolation of those wastes in impermeable layers has to be guaranteed. The suitability criteria should focus on the long-term isolation of all wastes and should be agreed in advance.

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1. Preface and content

Denmark has a relatively small inventory of radioactive wastes in terms of waste masses. As Denmark never built and operated nuclear power plants, the wastes resulted only from various research activities at the Risø Laboratory or were collected there (e.g. radioactive sources). The research activities at Risø were manifold, from general nuclear research over operation of three research reactors to uranium ore leaching. This resulted in several waste types, such as

- spent fuel from different research reactors,
- decommissioning wastes from these research reactors,
- operating wastes from those,
- spent radiation sources,
- uranium and uranium tailings,

and a large variety of different waste items (e.g. irradiated graphite and contaminated aluminium as decommissioning wastes). In total, 22 different waste types have to be managed.

This paper evaluates the wastes in the Danish inventory and derives some basic conclusions on the Danish concept.

2. Steps chosen in Denmark for managing its radioactive wastes

The following steps have been performed in Denmark to manage its radioactive wastes:

1. In a feasibility study /DD 2011/ the existing waste inventory was described and analyzed to identify the possible management options for those wastes. But this was not based on any safety criteria but on the sole believe that a small inventory requires only a small facility on the surface or in a small depth of less than 100 m below ground. With this pre-decision in mind, the different waste types and their radioactive decay over time were calculated and short-lived and longer-lived wastes were identified from that, but again without clear criteria. For all wastes, that were either of a high activity (e.g. spent fuel) or extremely long-lived (such as uranium) alternative options were briefly discussed, but not elaborated in more detail. So the study finally came to the conclusion that an above-ground disposal facility is sufficient and all waste types that do not fit into that facility should be managed elsewhere or in a different way (without going into further detail with those alternative solutions).
2. Following that, the Danish Geological Survey GEUS was commissioned to scan Denmark's landmass for potential sites that fulfil this criterion (at surface or at a maximum depth of 100 m). Again, there were no additional suitability criteria given, such as preferred geological layer types, maximum hydraulic conductivity of layers, minimum layer thicknesses, minimum layer depth, etc. Without those typical safety-related criteria GEUS identified suitable areas in Denmark, collected existing knowledge (on surface structures, by generic geologic knowledge, and via information from past boreholes) and, from that knowledge base, condensed the areas down to only six potential host sites in five different municipalities.
3. On this basis, as the next step, it is planned to perform a Strategic Environmental Impact Assessment (SEIA) procedure.

This is already all-in-all an astounding procedure up to now:

- The basic decision that a surface-near facility is sufficient has obviously been drawn early in the process, without defining and applying any rational safety-related criteria.
- The decision to exclude all wastes from the inventory of the surface-near disposal facility and to find something else, whatsoever, for those seems already fixed[1].
- The site selection procedure was started neither with clear safety criteria nor with a consent on those criteria among experts nor with a procedure to achieve a consent on those in a broader sense. The up-most strategic issues were already decided upon, by a few experts that obviously were not very familiar with the basics of nuclear waste disposal.
- So what can be the outcome of a Strategic Assessment now? To just re-confirm those two basic decisions that have already been drawn long ago, to re-confirm the methodology and the results of GEUS's site selection procedure and to compel the unwilling five affected communities by a national decision to accept this procedure and national decision?

The following lays down some of the basics of nuclear waste management and disposal in brief and asks if these basic rules are followed in Denmark.

3. Safety requirements for nuclear waste management

3.1. Basic requirements of nuclear waste disposal

The basic task of disposing nuclear wastes is to prevent its radioactive constituents from entering the environment, from moving into the air (for gaseous constituents) or into rivers, lakes or the sea (for dissolved constituents). The enclosure of those radioactive constituents in un-permeable layers then has the effect that the radioactive constituents decay without causing any radioactive doses to man and the environment. This basic concept is called "Concentrate and Confine" (C&C)[2].

The time over which the confinement of wastes has to perform its safety function depends from the waste's constituents. Table 3-1 provides three basic categories of wastes and their required isolation time.

The waste type requiring only a single year of confinement and isolation are medical wastes from the application of radionuclides. Those radionuclides have extremely short half-life times and decay nearly completely within one year. Wastes consisting of such radionuclides can be stored, then measured and released from regulatory control. Those wastes are then managed like other non-radiological medical wastes (e.g. incinerated or disposed). But note that those wastes always show additional small concentrations of longer-lived nuclides, but their concentration is below any radiological concern (on this term see below).

The second waste type requiring up to 300 years confinement are waste types with high concentrations of tritium (half-life 12.32 years) or those contaminated with cesium-137 (30.17 years) or strontium (28.97 years). Those nuclides decay enough in 300 years so they can

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[1] But what if only two of the 22 wastes fit into a surface-near facility?

[2] The opposite is "Disperse and Dilute", D&D, spreading the radioactive content as fast as possible over as much as possible water and land mass. D&D is attributed to an accumulative dose increase (small doses add up over long exposure times to large populations) and the resulting collective health detriments.

be released from further regulatory control. Note that these wastes also contain longer-living radionuclides as constituents, but in small concentrations that are not of any regulatory concern[3].

A very few special wastes decay within roughly 50 years to below regulatory concern limits. These wastes result from decommissioning with high contaminations of cobalt-60 and small concentrations of longer-lived nuclides. But these are single items while the majority of the wastes are longer-lived.

Table 3-1: Time of isolation and means to achieve protection of man and environment from radioactive wastes

Time (years)	Waste types	Facility type	Safety guaranteed by …	Remarks
1	Medical waste from application of isotopes	Decay storage facility (at source)	Administrational means	Final release from regulatory control after storage, further management of the waste similar to other medical wastes
300	Specific operational wastes from NPPs	Above ground or near-surface disposal	Administrational means, active monitoring and inspection necessary	Only applicable for specific waste types, surface-near disposal facilities have a high probability of failures: vulnerable to loss-of-control cases, loss-of-memory, leaching, ageing, and several natural events
> 300	All waste types	Deep disposal in geologic formations	Stability and integrity of geologic host formation, low hydraulic conductivity and high sorption capability of the near-field	Requires proof of integrity and isolation properties beyond reasonable doubt, not vulnerable to future ice ages and geologic fracturing

Source::Own collection and description in accordance with international regulations

All other wastes are too long-lived to be suitable for surface disposal. They require isolation times that are

- beyond 300 years where any administrational control (protection of the site against re-use including direct contact with the disposed wastes, societal memory conserving the special protection status that the location requires) is unreliable and insufficient,
- beyond 1,000 years where massive climatic or societal destructive events are highly probable on the near-surface, or
- beyond 10,000 years where massive climatic changes are to be expected (ice ages, etc.).

3.2. Disposal in geologic formations

It is clear from this that for any wastes that cannot be exempted from regulatory control or cleared[4] within a few hundred years geologic disposal is the only viable option to guarantee their long-term

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[3] The term “not of regulatory concern” means that associated doses are below 10 µSv/a, one hundredth of the dose limit for emissions from nuclear facilities. The accepted health risk from that limit is in the order of 10^{-6} per year.

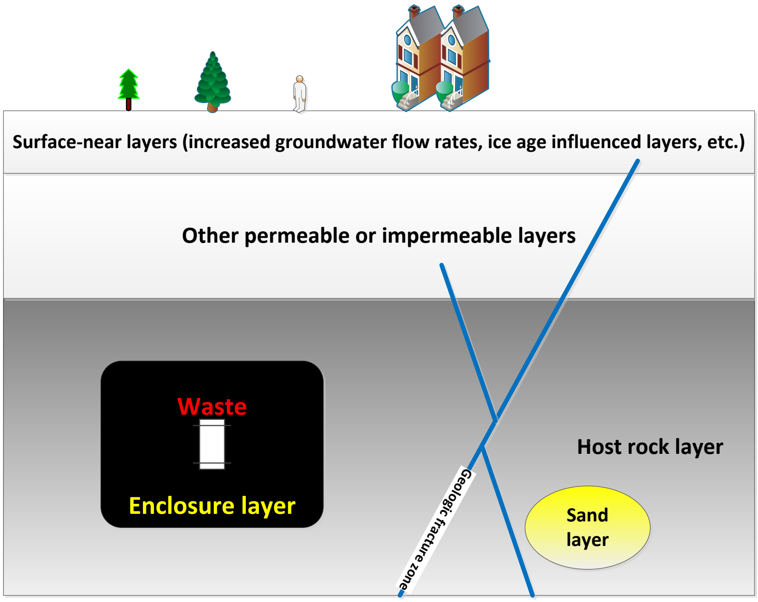
isolation from the environment. The probability that within a few thousand years from now an ice-age occurs is exactly 1.0, if long-term climate research understands past history correct. What happens in that case with a near-surface facility made of concrete, clay or HDPE liners? They are simply rubbled into dust and perform no safety function any more.

The basic concept of final disposal in geologic formations is therefore the reliably predictable isolation of wastes. The depth of repositories is not selected arbitrarily, as /DD 2011/ seems to assume. While 500 m distance in horizontal direction is not a large distance to travel (definitely not for a radionuclide dissolved in a creek or river water) the 500 m depth in vertical direction provides enough distance against such phenomena like rain, snow, storm, rapid groundwater movement, ice-ages and glaciers, civil wars and other incidents with probabilities higher than or equal to 1.0 over the next 1,000,000 years.

The basic safety concept of geologic disposal is

1. to isolate the waste from any possibility that an individual can come into direct contact with those wastes (physical protection). The necessary depth is a function of long-term continental up-lift, protection against the various surface-activities and climate changes (over geologic times).
2. to place the waste into an environment (geologic formation) where hydraulic water flow is either zero or near zero, so that neither leaching nor hydrologic driven water movement takes place (that is: water travel times from the emplacement horizon to the surface/biosphere should be longer than 100,000 years and in no case shorter than 50,000 years).
3. to close all openings to the isolating formation with a quality that does not compromise the stability and low water conductivity of the isolating geologic layer itself.

Figure 3-1: Repository in an impermeable geologic layer



Source: Own

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[4] See definition of those terms below.

Central for the enclosure function is the impermeable enclosure layer because this takes over nearly the complete function of isolation. All other layers around can be permeable or not, can have impurities such as sand layers (very common in natural clay layers), can be fractured (with increased water flow and fast transportation in those fractures), can be subject to long-term erosion, etc.. But not the enclosure layer: its basic isolation properties have to be constant, reliable, predictable and long-term stable. Any other layers can contribute to increase isolation (partial barrier function), but their contribution shall not be decisive.

Those geologic properties of the selected layers are not applicable in bedrock, if this is the only available isolation layer (Sweden, Finland) as in that case it is technically nearly impossible to identify large enough layers of completely water-tight and un-fractured bedrock and to proof its integrity in advance. Under these circumstances man-made technical barriers (such as high-performance copper canisters) have to guarantee enclosure of the waste and its constituents for the necessary times.

If geologic layers are available that are capable of taking over the complete isolation function (such as naturally compacted dry clay or dry layers in salt domes or rock-salt layers, it is not necessary to rely on questionable technical barriers[5]. In such dry environments it is impossible (salt) or a very long time is needed to dissolve waste fractions (dry clay), if the layers and locations for emplacement of the wastes are carefully selected and remain stable over very long times. A long time is further needed for solutions to travel through the formation (salt: no travelling; dry clay: very slow diffusive travelling). If a small fraction of only the most mobile radioactive substances so finally can reach the biosphere (less than 1/1000), they have majorly decayed. In clay only a few very mobile radionuclides (such as carbon-14, iodine-129 or chlorine-36) can spread into the isolation layer and any further by diffusion, not by pressure difference driven hydraulic flow. Even though a small portion of those nuclides finally can reach the biosphere (e.g. in a million years) the vast majority decays within its long travel time in geology.

Those basic safety functions of geologic disposal were neither recognized nor identified as rationale behind final repositories nor basically understood in /DD 2011/. Instead the study cares about an acceptable limit level for uranium in the biosphere, while uranium is completely insoluble in dry clay, travels only centimeters deep into dry clay formations and less than one millimeter deep into dry salt over a million years. Figure 3-2 symbols this: plutonium and uranium remain within the waste because they are insoluble and do not travel with water or by diffusion. The more mobile nuclides cesium and zirconium are trapped onto rust particles left from canister degradation and within the backfill of the disposal borehole and in the mine backfill (e. g. bentonite). Cesium and niobium are completely trapped by the first 50 cm thick layer of the geologic layer. The remaining geologic isolation layer is required to delay and decay the most mobile radionuclides iodine-129 and selenium-79. Discussing about uranium dispersing to the biosphere means that this has already hopped over four barriers to get there. Either the author does not understand barriers or does not want to apply the barrier concept in his safety planning of the repository. So: If uranium is allowed to travel to the biosphere in the Danish repository, there is something very basically wrong with that repository in that it does not have effective barriers. If it has no effective barriers for uranium, the more mobile radionuclides, such as iodine, cesium, radium, etc. will spread nearly completely to the biosphere.

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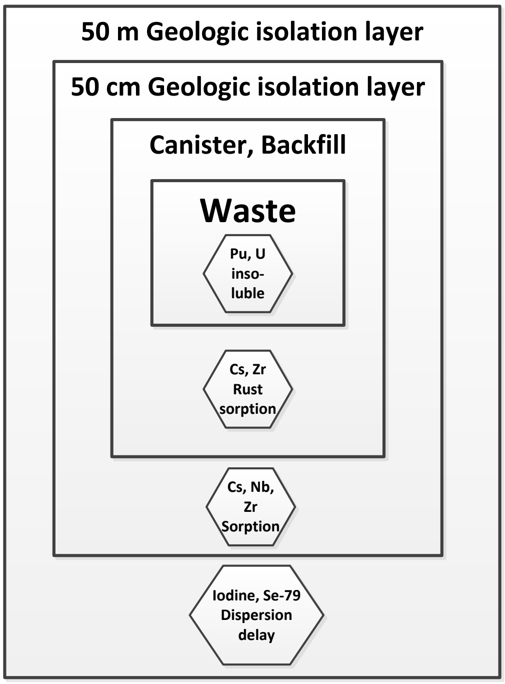
[5] Technical barriers are questionable, because the proof that they withstand all influences is the more complicated the longer the timescale is. So proofing corrosion resistance of technical materials over a million years can only utilize experiments over relatively short periods of a few years and always need to extrapolate. Experiments over a few years do not make sure that all mechanisms of degradation are well enough understood.

Figure 3-2: Barrier functions enclosing waste constituents



Source: Own

One of the astounding facts of the Danish site selection procedure is that GEUS only looked for surface-near clay layers. Surface-near clay layers are in most cases the end result of past ice-ages. It is therefore almost proven that those surface-near layers are also prone to future ice-ages, because they definitely were that in the past. The opposite would have been a sufficient criterion: if layers result from past ice-ages they are per se unsuitable for that purpose today.

Surface clay layers are very young, in geologic terms, and mostly are not consolidated. Unconsolidated clay is not very recommendable for a repository because of its adverse technical properties (instability of any openings, complicated stabilization necessary) and its high water (enables fast corrosion) and mobile salt content (increasing corrosion rates, pit corrosion mechanisms). Unconsolidated clay should only be approached if consolidated clay is definitely unavailable.

3.3. Long-term interim storage

The site selection, the planning procedure, the construction, the operation and the final closure of a repository requires very long times of several decades. During those the wastes have to be stored and managed safely in surface-near facilities.

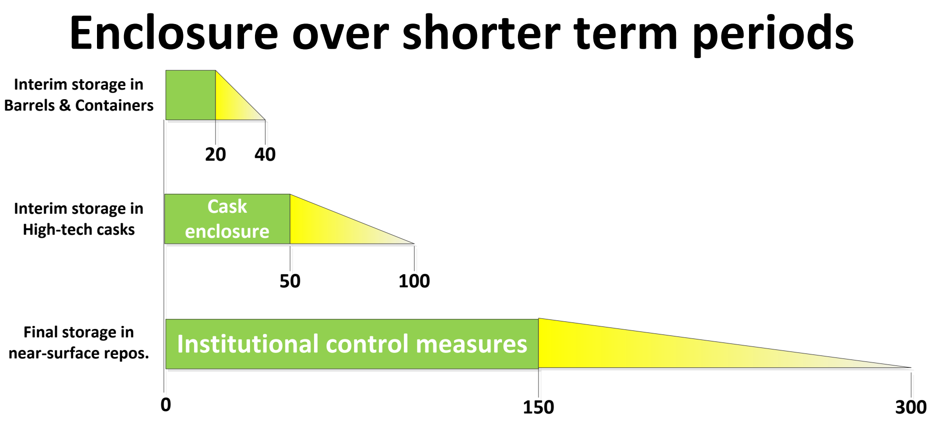
If storage times beyond 20 or 30 years are selected, this requires some further considerations. These times require long-term reliable package forms. Simple ISO containers do not withstand corrosion (from the outside through moisture, from the inside through the residual water content of the wastes) over such times. It is further in doubt that this is the case for much more sophisticated package forms, such as dry storage containers and their metal or plastic seals. The simpler the package form the more frequent will be necessary re-packaging, and the associated handling, of the wastes with resulting doses for workers, and the required preservation of practical, technical, engineering and theoretical knowledge, capabilities and experiences in handling/packaging those wastes. To perform this over many generations and over virtually several 100 years is nearly impossible, can in no case be evaluated as a responsible general position and cannot be termed a sustainable concept to cope with such long-term risks.

Considerably longer storage times such as 100 years cannot guarantee to be safe as the waste packages are subject to all phenomena on the surface that can finally compromise safety.

3.4. Time scales

The following provides an overview on time scales in nuclear waste management. The first two cases concern the interim storage of wastes, the third the near-surface disposal.

Figure 3-3: Enclosure over shorter term periods



Source: Own

Interim storage in barrels and containers is subject to rapid ageing processes, such as rust or cracking. With some care to be taken a reliable enclosure can be achieved over 20 years, accepting decreasing enclosure reliability or performing extra measures to increase package stability can extend this period to 40 years. Beyond that the storage requires re-packaging or other expensive measures.

Cask enclosure can be performed reliably over 50 years, if the cask is designed and built for those periods. Beyond that ageing phenomenon increase and much greater care must be taken. Storage times beyond 100 years are unreliable.

Final storage in near-surface repositories[6] requires several institutional control measures to be reliable over the whole time period. For example, the immission of radionuclides into ground- and surface-water requires monitoring. Any use of the site for purposes that includes surface degradation has to be reliably prevented. An overview of the most relevant impacts to be designed and planned for is given in Figure 3-4.

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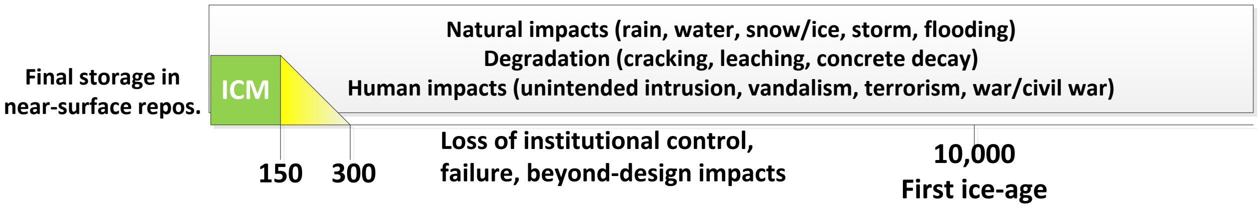
[6] The term “repository” is not exact for these facilities, because they are based on safety functions to be actively performed, such as strict re-use limitations and extensive monitoring.

Figure 3-4: Potential impacts that can compromise safety of near-surface facilities



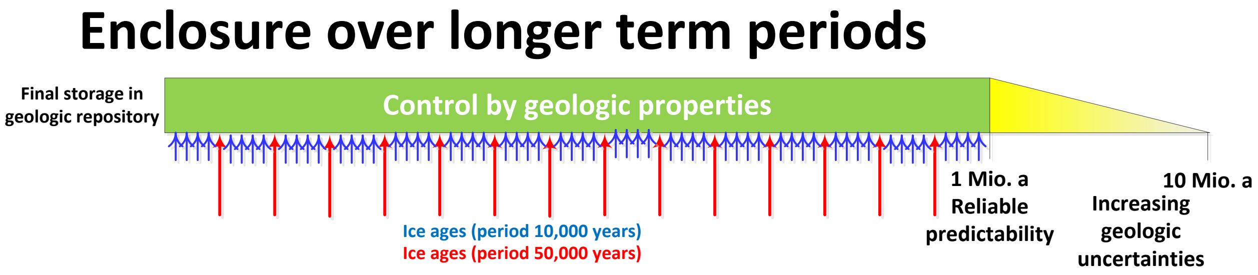
Source: Own

Over times beyond 300 years institutional control will fail with a high degree of certainty. If not, the listed impacts will in any case degrade such a facility.

To perform all institutional control over the 300 years a long-term ownership has to be setup and continued over the complete institutional control period. As those can fail over very long times it is in doubt that the necessary reliability can be kept up over the whole period of 300 years and is surely unreliable over longer time periods.

Figure 3-5 shows the characteristics for geologic repositories.

Figure 3-5: Enclosure over longer term periods



Source: Own

The enclosure of radioactive wastes over long times is completely to be controlled by the geologic properties of the selected isolating layer. The selection includes a layout and proof for its geologic stability, its inertness against all natural phenomena, e.g. shorter and longer term ice ages. For times beyond 1 million years a prediction can be made, but its reliability is the more limited the longer the time scale, so can have only indicative nature.

Predictability, based on geologic knowledge, is a basic requirement in this case. Any reasonable doubts on the long-term reliability of the isolation layer lead to unsuitability of the site for the purpose of geologic disposal. That makes clear that the site selection process requires a well-established knowledge on the site's geology. Selection processes that are not aware of the required quality of knowledge are useless.

3.5. Basic conclusions for the Danish waste management concept

The following basic conclusions can be drawn from that:

- Only wastes that decay within short periods of less than a year to below clearance criteria can be stored without the need of being managed as radioactive afterwards.

- Most other wastes, with only a few very specific exceptions, contain longer-living nuclides at a relevant level[7] that their decay does not lead to clearance of those wastes in manageable times. It is therefore recommended that those wastes, that do not decay to below current clearance levels in less than 300 years shall be finally disposed in suitable geologic formations.
- Denmark should dispose all its waste in such a carefully selected facility and should not take the risk that the disposal in a less reliably isolating manner will be considered unsafe later on in the process and that the decision may be revoked within a few decades later.

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[7] See chapter 4 for an evaluation of the Danish inventory applying this criterion.

4. Required isolation times for the Danish inventory

To determine which wastes in the Danish inventory are short-lived (equal to: suitable for a near-surface disposal facility) and which are longer-lived (equal to: require isolation in a geologic repository), a quantitative decision instrument is required. The method applied in /DD 2011/ to simply interpret decay curves is unsuitable and unreliable, as will be demonstrated in this chapter.

Chapter 3.1 describes how the longevity of radioactive wastes with a large number of different radionuclides can be determined. Chapters 3.2 to 3.5 analyses the Danish inventory applying this method. Finally chapter 3.6 demonstrates that the method used in /DD 2011/ is inappropriate.

4.1. The longevity based on current clearance criteria

4.1.1. The exemption or clearance concept in radiation protection

The basic criterion is simple: if the radioactive constituents of a waste material have decayed to activity levels to fulfil current exemption or clearance criteria, the waste can be termed not to be "harmful" any more. If the concentration of a single long-lived radionuclide (half-life time above 30 years) in the waste exceeds today's exemption or clearance levels, the waste cannot be termed short-lived. The waste then requires effective isolation and confinement in a geologic repository.

Exemption or clearance levels for radionuclides are well established in international and national radiation protection practices. The international basic safety standards (BSS) - jointly published by FAO, IAEA, ILO, OECD/NEA, PAHO and WHO -, the European Union – first in its Radiation Safety Standards of 1996 and later on in the recent update 2013 – and many EU member states – e.g. Germany –defined and implemented those exemption levels and apply those values in their practical radiation safety regulation – e.g. to determine if a certain application requires a license or not. In several countries wastes that fulfil the exemption or clearance criteria can be cleared, if this compliance has been proven (by measurement) and is confirmed by a regulatory decision (e.g. in Germany).

The clearance concept involves a dose calculation for relevant radionuclides and for a number of different scenarios. These scenarios reflect the potential uses that the cleared material or waste can take. The scenario with the largest dose effect is used to determine a radionuclide's clearance level. The broad variety of scenarios can be narrowed down if a material or waste is used only for certain purposes and if this can be guaranteed, e.g. for disposal together with large quantities of non-radioactive wastes or for steel recycling by melting. The resulting clearance levels for this "conditioned clearance" are in general by a factor of approximately 10 higher than for unrestricted clearance. Those "conditioned clearance" levels are unusual and are not applied here because they only make sense if the conditions can be guaranteed already today, but not in 10,000 years or later.

Denmark has implemented exemption or clearance levels only for naturally occurring radionuclides, because artificially generated nuclides do not play a practical role in radiation protection practice there. The following calculations use the clearance levels for unrestricted release from regulatory control established in Germany as a base. If other datasets would be used, single nuclides would differ to a small extent. But the major results and conclusions would not change.

4.1.2. Exemption or clearance levels for radionuclides

For a few selected radionuclides Table 4-1 provides practical values for exemption or clearance levels in use today. The table shows recent EU values as well as those established in Germany.

Table 4-1: Examples for nuclide-specific exemption and clearance levels, in Bq/g material

Nuclide	EU Table A	DE StrlSchV
H-3	100	1000
C-14	1	80
Cl-36	1	8
Ca-45	100	70
Fe-55	1000	200
Fe-59	1	1
Co-60	0,1	0,1
Ni-59	100	300
Ni-63	100	300
Se-79	1	3
Sr-90	1	0,6
Zr-93	10	100
Nb-93m	10	400
Nb-94	0,1	0,2
Nb-95	1	2
Mo-93	10	4
Tc-99	1	0,6
Ag-110m	0,1	0,1
I-129	0,01	0,06
Cs-135	100	20
Eu-152	0,1	0,2
Eu-154	0,1	0,2
Eu-155	1	30
U-238	1	0,6
Np-237	1	0,1
Pu-239	0,1	0,04
Pu-241	10	2
Am-241	0,1	0,05

Source: EU Table A: /EU 2013/, Table A, EU clearance level for large amounts of material; DE StrlSchV: /StrlSchV 2012/, Tabelle 1, clearance level for unrestricted release from regulatory control

As can be seen from that table the exemption and clearance levels between EU and DE differ by up to a factor of 10. The German values are less restrictive for fission and activation products, in average by roughly a factor of 6, and more restrictive for actinides, on average by a factor of roughly 3. For the purpose we use these levels here for, to determine the longevity of wastes, these differences are of minor influence.

But, as also can be seen, the different radionuclides are by a factor of up to five orders of magnitude different (EU: 0.01 to 1,000; DE: 0.04 to 1,000) in their radiological relevance. The doses that the different radionuclides can cause are by such factors different. This puts the simple summing up of activities of radionuclides whatsoever, as chosen in /DD 2011/, into perspective: those sums are simply nonsense as they say nearly nothing about the radiological relevance of the summed up values.

As all wastes (even the most short-lived ones) consist of a mixture of short- and longer-lived nuclides, the exemption or clearance practice uses the criterion of the sum of relations between the activity concentration of a nuclide and its clearance level. A material or waste can only be cleared if the sum of all those relations is less than 1.0.

Those clearance criteria (in the EU /EU 2013/ as well as in Germany /StrlSchV 2012/[8]) are defined on a risk base of roughly one in a million for an individual. That means that the risk for a health damage for individuals by that clearance practice is trivial (De minimis). The clearance levels reflect the radiotoxicity of the different nuclides in terms of decay modes (alpha, beta, gamma),

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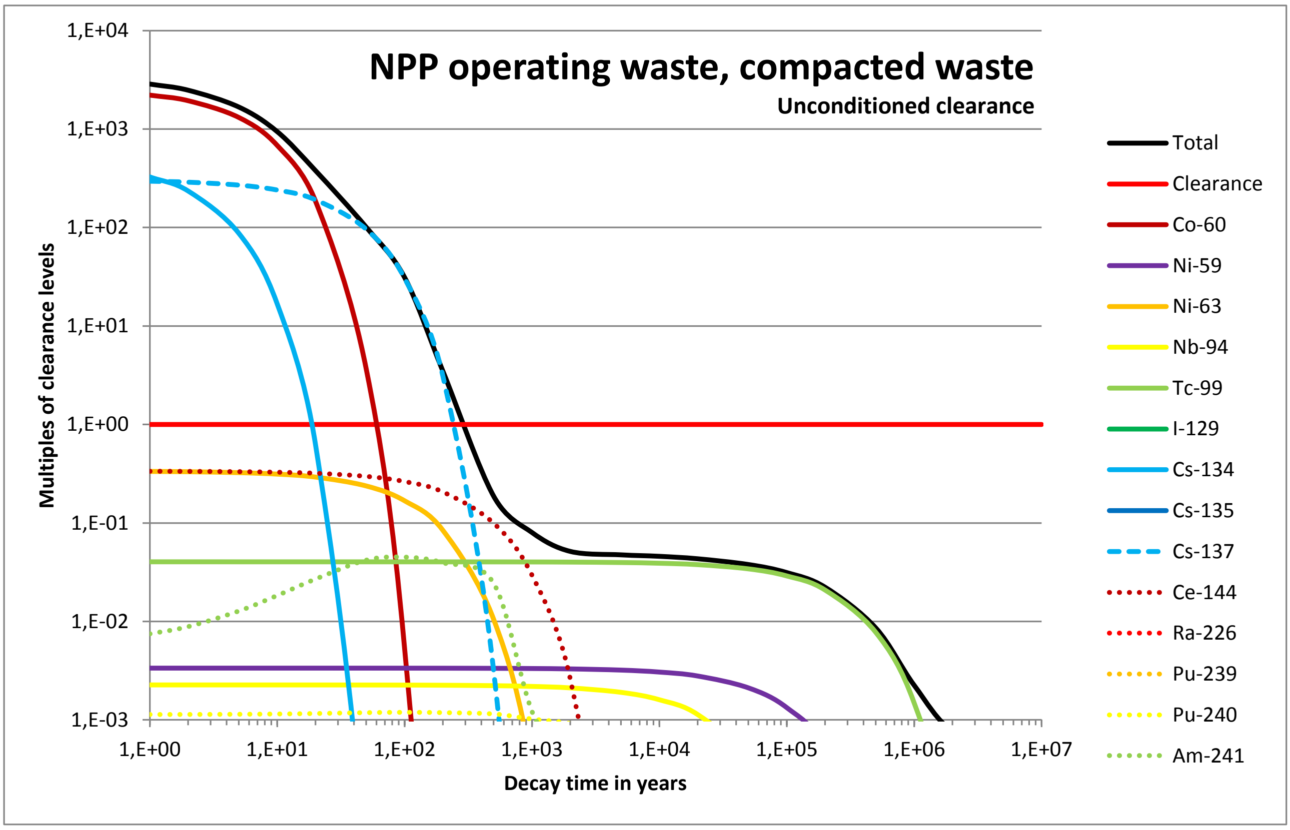
[8] The values used in this calculation were taken from /StrlSchV 2012/. Small differences between /EU 2013/ and the German regulation in /StrlSchV 2012/ are insignificant, as shown above.

ingestion and inhalation doses, doses from direct radiation, transfer of the nuclides from soil to plants and via different bio pathways, etc.

4.2. Typical short-lived wastes that fit into near-surface facilities

In order to demonstrate that the described method enables to determine which wastes fit into near-surface disposal sites with an active administrational control over 300 years a typical waste from nuclear power plant operation is shown in Figure 4-1.

Figure 4-1: Multiples of clearance levels for a typical short-lived waste



Sources: Waste composition: NAGRA data base; clearance levels: Germany; own calculation

Note that both axes in the diagram are logarithmic, so that each scale unit is ten times higher/lower than the next above resp. below (y-axis) or the next to the left resp. right (x-axis). Displayed are the multiples of the clearance levels for the individual nuclides in this waste and their sum.

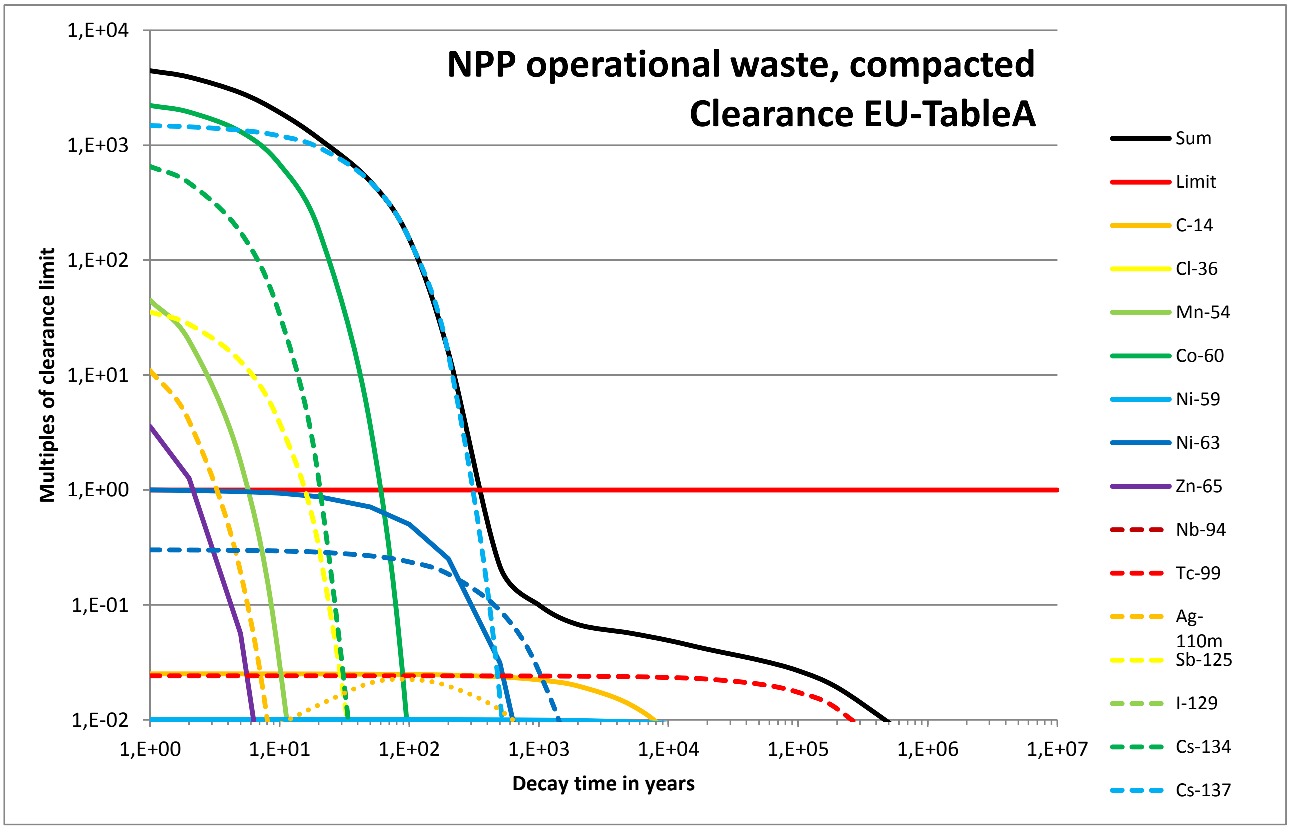
The sum criterion of 1.0, which means: all radionuclides in the waste together in combination are below clearance levels, is marked with a red line.

As can be seen, the waste currently exceeds the clearance levels (red line = 1.0) by more than a 1,000-fold, so cannot be cleared today. The main contributor to that exceedance today is Co-60 with a half-life time of roughly 5 years (brown line). A faster decaying radionuclide, Cs-134, contributes to the exceedance, but is not the limiting factor. With time the Co-60 content of the waste decays, the exceedance drops down to 100-fold after roughly 20 years. Now the radionuclide Cs-137 (light blue broken line) mainly causes the exceedance. Slightly after 300 years (exactly 425 years), this nuclide has decayed enough and the sum curve falls below the clearance level. The waste could be cleared after this enhanced decay period, he is – by definition - not dangerous any more.

As can also be seen from the other radionuclide curves, there are considerably longer-living radionuclides in this waste that would require longer times to decay, such as e.g. Ce-144, Ni-63 and Tc-99. But their concentration in Bq/g in the waste is so small that the clearance level is not exceeded by those nuclides but only by Co-60 and Cs-137.

Figure 4-2 provides the curves for the same waste, but uses the EU Table A clearance values.

Figure 4-2: The same waste, but different clearance values (EU Table A)



Source: Waste composition: NAGRA database; clearance criteria : /EU 2013/ Table A; calculation : own

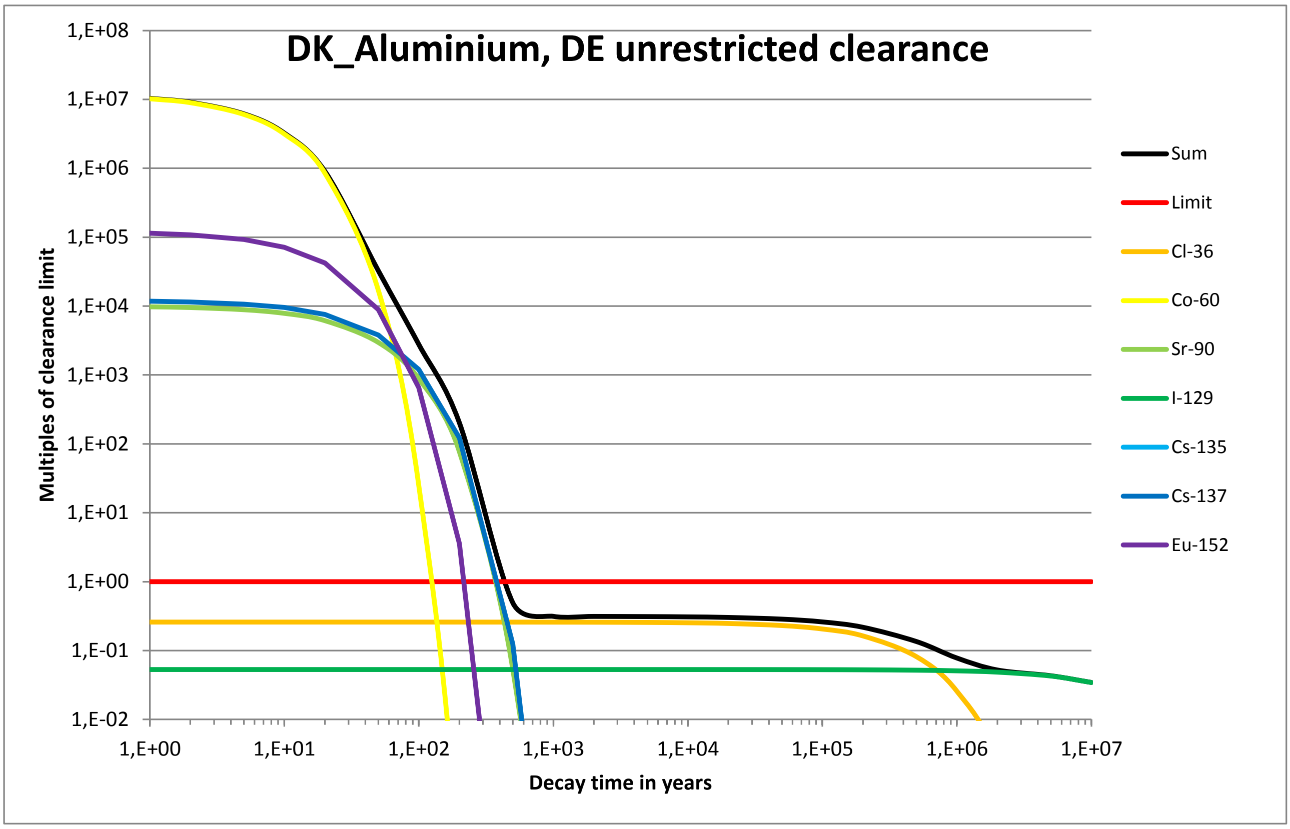
As can be seen the curves in this Figure are different from the previous one as more nuclides contribute to the exceedance in the beginning (due to smaller EU clearance values). But the result is nearly the same: the waste decays in roughly 300 years to below the clearance limit.

So the result of the calculation is robust and not relevantly depending from the selected set of clearance values (EU or Germany).

4.3. Application of clearance levels, first example: type 2 waste

To illustrate what the clearance levels say about the longevity of wastes, the decay curve of waste type 2 of the Danish inventory is displayed.

Figure 4-3: Decay of waste type 2 (contaminated aluminium) of the Danish inventory



Source: Own calculation based on data in /DD 2011/ and own estimates for Cl-36, I-129 and Cs-135 (Cs-135 below scale)

As can be seen five nuclides decay in between 100 and 1,000 years to below clearance levels, while two others with long half-life times, Cl-36 and I-129 decay only later on. A third longer-lived nuclide, Cs-135, is below scale from the beginning and therefore does not show up. Those three nuclides were added by estimation, because those were not specified in the Danish inventory data. But in this case they do not add relevant to the sum as they are below clearance levels already today.

The sum curve of the waste crosses this line after 500 years. That means: after this time period the waste has decayed enough and after that period could be cleared and released[9]. Or: before 500 years safety has to be guaranteed by a safety function or measure, beyond that this is not necessary anymore because the residual risk for an individual coming into direct contact with this material or any other use of the material is less than one in a million.

If we see that waste in relation to the characteristics in Table 1-1, we cannot sort it into the category for above-ground-disposal. After 300 years, the administrational control period for surface-near disposal, the waste still exceeds the clearance criterion by about 100-fold, but after 500 years this criterion is satisfied (steep slope of the curve). So for this waste type an above-ground-disposal is a possible management solution, but it is slightly above the edge of being

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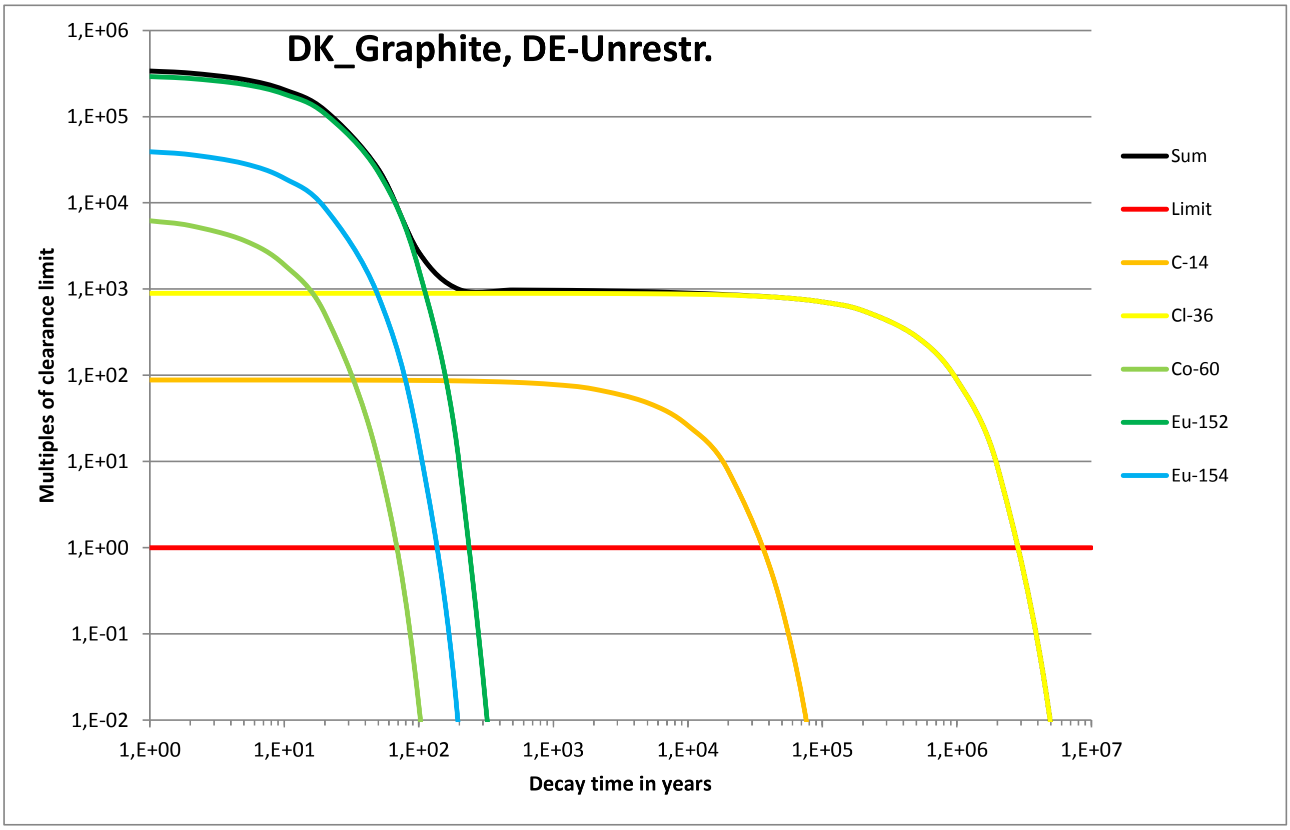
[9] In this case the waste mass for this waste was listed in /DD 2011/ and so was taken for the calculation. In other cases where the mass was not listed in /DD 2011/ an estimate, mostly including the package material, was used. That yields numbers that are by a factor of 2 to 10 smaller than without the package material. The differences are insignificant in most cases.

suitable for this option. In fact, 500 years of necessary institutional control measures for a near-surface disposal facility is unrealistic and unreliable.

4.4. Example 2: type 1 waste

Waste type 1 of the Danish inventory consists of graphite that had been used in one of the research reactors as moderator. Neutron capture in carbon has led to a considerable concentration of C-14 in that waste, combined with contamination of the material with Co-60 and the europium nuclides.

Figure 4-4: Decay of waste type 1 (graphite) of the Danish inventory



Source: Own calculation based on data in /DD 2011/ and own estimates for Cl-36

This curve is different in that it intersects the clearance criterion only after approx. 30,000 years caused by its C-14 content. As the Cl-36 content for this waste type was either not determined or not published, the estimate for this nuclide was based on the C-14 content and on nuclide relations for operational wastes from NPPs. This might be a questionable approach. If this Cl-36 estimate is confirmed by analysis, the intersection takes place after 3 million years instead of 30,000 years. This difference demonstrates how relevant undetermined radionuclides can be. But factually not in this case: the waste is in any case unsuitable for surface-near disposal due to its high C-14 content.

If the EU clearance levels would have been used in this calculation, the result would have been significantly higher. The EU clearance level for C-14 is by a factor of 80 more restrictive than the German regulation. The exceedance of clearance levels by this waste is so by a factor of 80 larger, but due to the steep decay curves the decay times to the intersection of the 1.0 limit are nearly the same.

Isolation or confinement as a safety function is required over times that definitely cannot be achieved by surface-near facilities. This waste can clearly not be called short-lived and is not suitable for near-surface disposal. As C-14 is highly mobile, because, in the long-term, it is either oxidized to carbon dioxide (in a near-surface facility by the oxygen provided) or reduced to a mixture of carbon dioxide and methane (in a geologic disposal facility with small amounts of residual or diffusive water). Both gases, but especially methane, are travelling fast if not enclosed in a gas-tight environment. Long travel times beyond 30,000 years are necessary to resolve the problem.

If this protection is not provided by the designed disposal facility, would it be possible to just delay the release of C-14 from the repository to the environment enough if only the radiation protection limits are respected in the future? To answer this question one could also ask if this carbon-14 inventory could be emitted today, e.g. if the graphite would be burned under carefully controlled conditions in an incineration plant, so emitting the whole inventory immediately to the air. And carefully respecting dose limits by mixing the emissions with added carbon dioxide from co-burning non-radioactive wastes. If the answer to this question is “no”, e.g. because C-14 emissions can cause small but very long-lasting dose contributions after its distribution to the atmosphere, then why should the answer be different in 300 years when the near-surface facility loses its administratively guaranteed safety function and most of the disposed carbon-14 has not been decayed by then?

This demonstrates that putting the sole focus not on proper isolation but on the radiation protection limits is inappropriate. If a facility releases its inventory nearly completely to the environment, but only after a relatively short period of delay, the question whether this exceeds individual protection limits is completely misleading: the facility then is not designed to enclose the wastes but to dilute the wastes and to discharge them. Of course any repository has to respect discharge limits, but its prime task is not to discharge but to enclose.

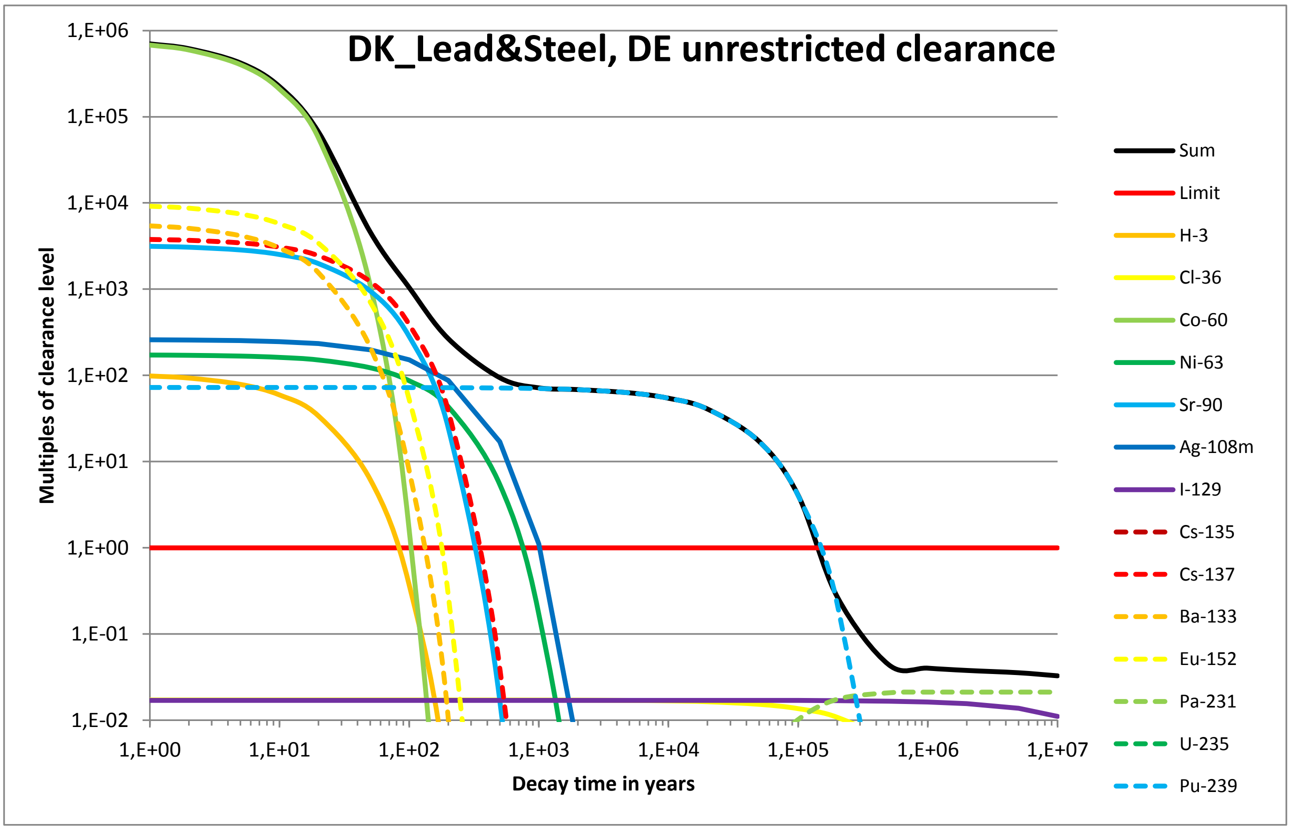
So whatever radioactive doses will result from a certain setting, it cannot be a rational approach to only respect the given limits. And it cannot be rational to only design such a facility for not more than respecting those limits. If proper isolation layers are available in the Danish geology those should be selected, no matter if those are at 100 or at 300 m depth, because the cost difference between those alternatives is too small to be decisive. If the C-14 is to be disposed at 20 m depth and 80% of the disposed inventory will be discharged during the next ice age, the concept can only be evaluated as inappropriate, even though the individual dose limits are respected.

The overall goal of a repository has to be “zero discharge” and “decay within the isolation layer”, any escaping portion can only be accepted if it is only a very small portion of the total that is to be enclosed and if the isolation quality is as good as it can be.

4.5. Example 3: waste type 3 Lead and steel

This waste results, like a few other types, from decommissioning of the research reactors.

Figure 4-5: Decay of waste type 3 (Lead and steel) of the Danish inventory



Source: Own calculation based on data in /DD 2011/ and own estimates for Cl-36 and I-129

As can be seen, exceedance of the clearance criterion for 1,000 years is caused by a number of short-lived nuclides and, beyond 1,000 years, by its Pu-239 content (for 200,000 years). It is clear from that this waste cannot seriously be termed short-lived and is unsuitable for disposal in a near-surface facility.

4.6. All waste types of the Danish inventory

After having provided a few examples to illustrate the method used here and the discussion of a few selected nuclides Figure 4-6 provides an overview of the clearance curves of all waste items of the Danish inventory. In this figure no individual radionuclides are displayed but only the sum curve of each individual waste item.

Note that for a few wastes the curves do not generally have a downward trend but some are increasing with time after a few thousand years. That is caused by radionuclides in the wastes that build up daughter nuclides. In this case the sums are rising, as some of the nuclides in the decay chain have a lower clearance limit than the mother nuclide[10].

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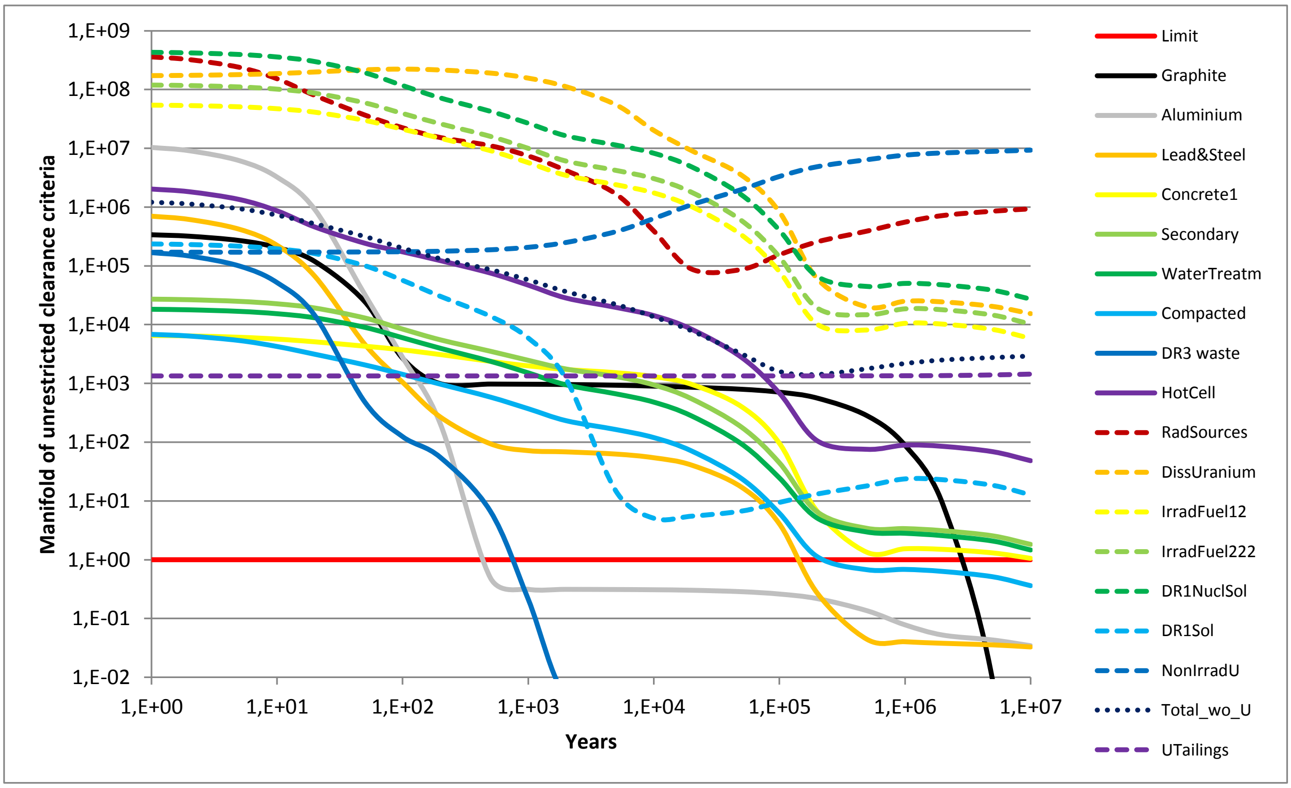
[10] E.g. daughter nuclide radium-226 has a lower clearance limit than the uranium-238 it originates from in the decay chain. The built-up of chain nuclides is a long process for nuclides with long half-life times such as uranium-238, thorium-232 and neptunium-237.

Figure 4-6: Decay characteristics of all wastes of the Danish inventory



Source: Own calculation based on waste descriptions in /DD 2011/

It is clear from that that

- none of the Danish waste types decay within the administratively controllable period of less than 300 years to below clearance levels, so none of it is suitable for near-surface disposal,
- only two of the Danish waste types (Aluminium, DR3 waste) decay enough within the time period to the next predicted ice-age (≈10,000 years) to below clearance levels,
- one waste type (DR1 solution) is only slightly above that criterion (within one order of magnitude) in 10,000 years, and
- all other 18 waste types of the Danish inventory require isolation/confinement times of 100,000 years or beyond. Within 100,000 years these waste types are between two and seven orders of magnitude above the release limit. That is far above the activity levels that can be disposed in a near-surface repository. And this type of longevity is not limited to spent fuel from the research reactors but also concerns such waste types as wastes from the Hot Cell or compacted waste.

The large exceedances of the clearance criterion makes also clear that uncertainties or inaccuracies within only one order of magnitude (factor 10), as the decision to use German regulatory clearance values instead of those from the EU, are largely insignificant: no other conclusions would result if the exceedance of the clearance criterion would be by a factor of 10 smaller or higher.

It can be concluded from that, that it is not rational to sort out certain waste types of the Danish inventory and to dispose those in a differently designed isolation system and to choose a less isolating system to dispose the remaining rest of the wastes.

4.7. Waste radiotoxicity

The method used in /DD 2011/ to compare those wastes is misleading. To only register the activity of the different waste types over time is inappropriate because the basic properties of the different nuclides are not considered. It so assumes that each Becquerel is equal to any other Becquerel of radioactivity. As these are different by five orders of magnitude in respect to doses and risks posed, this is a misleading approach.

And, what is furthermore relevant, the method of displaying activity curves does not provide a rational measure below which a waste's activity can be considered safe. So anything can be left in the dark if the term "short-lived" remains undefined. As this term is often used, especially in countries with extensive near-surface disposal facilities, but never really defined using applicable radiation protection standards, it is easy to use, but overall unreliable.

The Danish wastes are not at all short-lived in the sense that they can be disposed in a near-surface facility with a low quality of barriers made of concrete or HDPE, and subject to ageing/leaching, physical impacts and prone to future ice ages.

If the only criterion that is relied upon is the dose limit: this can (and is) simply misleading, as has been shown in the graphite and C-14 example. If it is today unacceptable to spread radionuclides in large amounts to the atmosphere it would not be acceptable either if this release from a leaking disposal facility is simply delayed by a few hundred years only. If it is possible to concentrate and confine those radionuclides well enough so that more than 99% of the disposed inventory decays far from the biosphere, this path should be taken. No matter if the dose limit is undercut by only one or by seven orders of magnitude, safe confinement is the leading term and not "Disperse and Dilute" to below dose limits under majorly uncontrollable future conditions.

The conclusions drawn in /DD 2011/ from those decay curves are further misleading. It is stated that disposing the uranium in a different manner (in deep boreholes) reduces the radiotoxicity. This is, from the curves shown in Figure 4-6, simply applying for all waste types except for the two shorter lived ones.

And, uranium is one of the least mobile species, if it is enclosed in an isolating geologic layer and not placed in an oxygen-rich environment like in a near-surface disposal. So uranium does not require a different safety function than most of the other wastes, too, and needs no extra requirements. This misinterpretation is a function of the selected analysis and has nothing to do with a rational waste evaluation.

5. A single high-performance or several different repositories with tailored isolation properties?

5.1. Waste-centered tailored isolation requirements

It is clear from the above that the concept of site selection and design goals for individual waste types would not contribute to a solution because the differences in the waste type's properties are not that big: nearly all wastes have to be isolated to a timeframe well beyond 1,000 years, their proper isolation in geologic formations therefore has in any case to withstand ice-ages, and the spreading of mobile radionuclides has to be diminished to a high extent by a strongly reduced hydraulic flow and by strong sorption to prevent from re-entering the biosphere.

As the requirement of constant stability and integrity of the geologic isolation formation majorly defines its suitability, its size is rather an issue of secondary relevance. As diffusion through a compact natural clay layer is not a linear process, small inventories do not require linearly smaller sizes. A clay layer with less than 100 m in vertical size has a stability problem, even though the waste volumes to be emplaced and the diffusion of radionuclides might require less vertical size. A clay layer that extends from the surface to a depth of 100 m has a different stability problem: after the next ice-age about 50 m of its vertical size would not be in that place any more but eroded away, probably but not necessarily exchanged with fresh, but unconsolidated sediments. The consequence of this is that GEUS has worked under inappropriate framework definitions, that their main focus was not on finding a stable host formation (because the stability criteria were not even established and formulated) and their evaluation results are unsuited to identify suitable host formations.

5.2. Waste isolation principles for the Danish inventory

Due to the very different waste types a large variety of wastes occurs. The spent fuel from the research reactors is by three to four orders of magnitude more radioactive than other wastes in the Danish inventory (see Figure 4-6). As travel distances in dry clay layers of e.g. 40 to 50 m (in total 80 to 100 m thick) are sufficient to guarantee that all immobile radionuclides decay completely and that more than 99% of the few mobile species decay within the clay layer it is sufficient to identify such layers, to characterize those and to build a repository in that layer.

The fact that these high-performance isolation requirements would not be necessary for the less radioactive waste types is correct, but does it really make sense then to build two or more repositories to prevent from a “too sophisticated” isolation performance of a single repository that fits all? The technical steps for the disposal of 300 containers of uranium mill tailings waste in an operating repository means, at maximum, prolonged operation of the facility for a year, while the construction and operation of an extra repository for those wastes, e.g. extra boreholes, requires by far more effort.

All other technical requirements are non-linear, too. Construction, operation and closure of a shaft or transport opening is not very different if the repository is at 100 m depth or on the 300 m level. As 10 m depth is definitely unsufficient for isolation and to maintain the long-term integrity of the disposal layer, this “cheaper” solution therefore does not come into question either. So the difference in depth does not really matter if the minimum performance is to be guaranteed.

The borehole technology might be attractive for small inventories in terms of masses. But the authors of /DD 2011/ did not consider the large uncertainties of this technology any further. Those boreholes require a proper identification of suitable and large enough underground layers on an acceptable depth below surface with high-quality isolation properties. Current seismic methods provide data with a resolution of +/- 10 m, so geologic fractures, unsuitable sand lenses, etc. can only be detected if they are larger than that minimum resolution. So each borehole is connected to a large uncertainty that the layer on this specific location is unsuitable. Later corrections cannot be made, but the borehole has to be backfilled to the same quality as those with waste emplaced to protect the neighboring potential emplacement space.

Typical is that this high-quality isolation layer does not appear in the description of the borehole disposal method. It seems that this was neither realized nor taken into account. The evaluation is therefore useless as the task was not understood at all. With that missing of the central element and misunderstanding of the central task, Denmark risks that the whole task will fail.

In a geologic repository the site suitability evaluation can be performed during the site suitability testing, from much smaller distance and hence to a much higher resolution. The repository's underground layout can be adapted to those underground exploration results, unsuitable areas can be avoided, and the risk that the available formation is unsuitable to host all the wastes so can be minimized. But the analysis in /DD 2011/ is too simple to discuss these risks and to compare those with other options. /DD 2011/ only drops this term but is unable to understand the concept fully. After that, the simple borehole approach can turn out as expensive as more than only one underground repository. "It looked so simple in the beginning and from the distance, but as we came nearer and nearer it turned out to be rather complicated."

So it is only wise to tailor the isolation requirements to the highest necessary waste category in order to cover all types of Danish wastes. The idea of tailoring three or four different repositories to the specific requirements of different waste types, and that all on the basis of expected simplicity, is neither technically nor economically nor ecologically sound.

5.3. Inappropriate separation of waste types from the repository inventory

How inappropriate the idea of tailored requirements for any waste type is can best be seen on the example of uranium mill tailings. This waste is by three orders of magnitude above clearance criteria, and it remains there over virtually unlimited times.

This waste requires proper isolation from the environment over virtually unlimited times. Specific requirements for these wastes are:

- to keep uranium isotopes from moving requires oxygen-free conditions (to keep uranium in its four-valent cationic form and prevent from oxidation to the more mobile hexavalent form),
- to prevent already existing or later in-growing radium-226 from moving fast, which can either be performed in a water-free environment, or in an environment rich in sulfate or by sorption on active surfaces, e.g. in clay,
- to prevent radon movement by a gas-tight environment (delay over one month is sufficient).

All these conditions can be provided above ground, they need not necessarily be performed underground. But above ground these conditions cannot be guaranteed over the virtually unlimited times, due to erosion, degrading of cover systems, or in harsh environments (e.g. during an ice-age).

The fact that these wastes are worldwide disposed in above-ground facilities is caused by the fact that it is near to impossible to dispose million tons of waste in deep geologic formations. So the disposal above ground is a compromise between isolation/safety requirements and technical opportunities, given the large volumes involved. In the Danish case this mass is rather limited, so the argument that it is impossible to geologically dispose the material is invalid (150 ISO containers or 1,000 m³ cannot be called a large volume). So why exclude this waste type from the inventory to be safely disposed? Neither the exceedance of the release criterion nor the limited disposal volume is reason enough for a separate solution for this waste, besides the practical question on what else to do with these wastes.

6. Conclusions

1. The method and criteria chosen to evaluate the feasibility of disposal are unsound from a safety standpoint because they simply ignore the basic principles of safe geological disposal.
2. The basic principle of isolating the wastes and to guarantee that the radioactive content decays in safe distance to any future people has not governed the site selection and site evaluation process.
3. Based on this fundamental error, the resulting site-selection process to search for suitable near-surface geologic situations is inappropriate. The criteria should have been instead to identify geologic layers of low or no hydraulic conductivity with a sufficient vertical extension of more than 80 m thickness, in a suitable depth (e.g. 300 to 800 m) and with a geologically predictable long-term integrity in Denmark.
4. The results of the performed site-selection process are useless as they are based on inappropriate criteria.

List of References

DD 2011	Danish Decommissioning: Pre-feasibility study for final disposal of radioactive waste. Disposal concepts. - Main Report, May 2011, [Link]
EU 2013	Council Directive 2013/59/EURATOM of 5 December 2013 laying down basic safety standards for protection against the dangers arising from exposure to ionising radiation, and repealing Directives 89/618/Euratom, 90/641/Euratom, 96/29/Euratom, 97/43/Euratom and 2003/122/Euratom. - Official Journal of the European Union, L 13/1 of 17.1.2014, [Link]
StrlSchV 2012	Verordnung über den Schutz vor Schäden durch ionisierende Strahlen (Strahlenschutzverordnung - StrlSchV) vom 20. Juli 2001 (BGBl. I 2001, Nr. 38, S. 1714, BGBl. I 2002, Nr. 27, S. 1459), zuletzt geändert durch Artikel 5 Absatz 7 des Gesetzes vom 24. Februar 2012 (BGBl.I 2012, Nr. 10, S. 212), [Link]