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Mo-93 from the grave to the cradle

Report from a workshop on molybdenum in radioactive waste and in the environment

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Abstract

Molybdenum-93 (Mo-93) has emerged as an important radionuclide in the safety assessments for the expansion of SFR (final repository for short-lived radioactive waste) and the construction of SFL (final repository for long-lived radioactive waste). In order to get a better picture of the current understanding of the biogeochemistry of Mo and in particular the uncertainties associated with Mo-93 in the current safety analysis a one-day seminar on Mo-93 was organised on 14th June 2016. The present report summarises the presentations given at the workshop and the associated discussions.

Mo-93 ($T_{½} = 4\,000$ a) is generated by neutron activation of stable Mo-92 so it will occur anywhere where Mo-containing material has been exposed to neutron radiation. Reactor parts that are made of Mo alloys are therefore especially important, but Mo exists as impurities in many reactor materials. Since Mo-93 emits only low-energetic gamma rays it cannot be easily detected, and the estimates of Mo-93 therefore largely depend on modelling and measurements of associated more easily measured radionuclides, e.g. Mo-99, Tc-99m, I-131 and Cs-137.The activity of Mo-93 in SFR is currently estimated to ca 5.4×10^9 Bq, but the number is expected to increase to 2.9×10^{10} Bq in 2075. On-going method developments will hopefully improve these estimates.

In the repository Mo-93 in ion exchange resins is assumed to be instantly released, whereas the release of Mo-93 from other reactor parts is controlled by corrosion rates. Once mobilised, Mo-93 is assumed to be present as molybdate, which can be transported by advection and diffusion through the engineered barriers. The thickness of the barriers and the sorption of Mo-93 onto bentonite and concrete will therefore be decisive for how large activities of Mo-93 that may escape to the bedrock. In the bedrock Mo-93 is also assumed to be present as molybdate. In SKB's current radionuclide models it is assumed that there is no retention of Mo-93 in the bedrock (i.e. the K_d value is set to zero), whereas Posiva has set a non-zero K_d for Mo in the bedrock.

In the biosphere Mo is an important micronutrient, for example because Mo-containing enzymes play a central role for the fixation of nitrogen and the reduction of nitrate. The sorption of Mo is generally strong in soils at low pH. Recent experiments have demonstrated that it can bind to both Fe/Al hydroxides and organic matter in the hexavalent oxidation state. In more reducing (sulphidic) environments Mo(VI) will react with sulphide and form Mo(IV), which substantially decreases its mobility. This is an important removal mechanism in sulphate-reducing marine and freshwater environments, which can lead to high concentrations of Mo in sediments. This process also seems to be important in lake and sea sediments in the Forsmark area, which in some cases show markedly higher Mo concentrations than local till. Natural Mo concentrations in the near-surface groundwater range from ca 0.1 to 10 $\mu g\ L^{-1}$ with a trend of higher levels at higher pH. The major source is presumably weathering of albite in local soils, but there is also an atmospheric input of Mo, mainly related to the steel industry. Currently the atmospheric deposition of Mo in Sweden is only monitored at Visingsö.

In the ecosystem models Mo-93 is characterised by relatively low K_d values. Compared to other radionuclides a relatively high fraction of Mo-93 is predicted to accumulate in peat and postglacial sediments. In addition to high mobility, high dose-rate conversion factors and high bioavailability also contribute to the relatively high doses related to Mo-93 in the current models. The dominating exposure pathway is expected to be ingestion of contaminated food. The most critical scenario is when a mire is drained and converted to agricultural land.

Sammanfattning

Molybden93 (Mo-93) har visat sig vara en av de viktigaste radionukliderna i säkerhetsanalyserna inför utbyggnaden av SFR (slutförvar för kortlivat radioaktivt avfall) och konstruktionen av SFL (slutförvar för långlivat radioaktivt avfall). För att få en uppfattning om hur väl molybdens biogeokemi är känd och i synnerhet vilka osäkerheter som hänger samman med Mo-93 i de nuvarande säkerhetsanalyserna ordades ett endagsseminarium om Mo-93 den 14 juni 2016. Föreliggande rapport utgör en sammanfattning av de presentationer som gjordes under mötet och påföljande diskussioner.

Mo-93 ($T_{½}$ = 4 000 a) härrör från neutronaktivering av stabilt Mo-92. Det kommer därför att dyka upp överallt där Mo-haltigt material har exponerats för neutronstrålning. Reaktordelar som är tillverkade av Mo-legeringar är således speciellt viktiga, men Mo förekommer som orenheter i många reaktormaterial. Eftersom Mo-93 vid sönderfall bara avger lågenergetiska gammastrålar, kan den inte mätas på ett enkelt sätt, och skattningarna av Mo-93 bygger därför till stora delar på modellering och detektion av mer lättmätta radionuklider, t.ex. Mo-99, Tc-99m, I-131 och Cs-137. Aktiviteten av Mo-93 in svenskt radioaktivt avfall uppgår för närvarande till ca 5.4×10^9 Bq, men denna siffra förväntas öka till 2.9×10^{10} Bq år 2075. Pågående metodutvecklingar kommer förhoppningsvis att kunna minska osäkerheten i dessa siffror.

I slutförvaret antas Mo-93 i jonbytesmassor frigöras omedelbart, medan mobiliseringen av Mo-93 från andra reaktordelar antas kontrolleras av korrosionshastigheten. Efter mobilisering antas Mo-93 uppträda som molybdat som sedan kan transportas genom advektion eller diffusion genom de tekniska barriärerna. Tjockleken av dessa barriärer och sorptionen av Mo-93 till bentonit och betong kommer därför att vara avgörande för hur höga aktiviteter av Mo-93 som kan nå berggrunden. I berget antas Mo-93 också uppträda som molybdat. I SKB:s nuvarande radionuklidmodell antas det inte ske någon retention av Mo-93 i berggrunden (det vill säga K_d-värdet är satt till noll), medan Posiva har satt ett K_d-värde > 0 för Mo i berget.

I biosfären är Mo ett viktigt mikronäringsämne, bland annat eftersom Mo ingår i enzymer som spelar en central roll för kvävefixering och nitratreduktion. Sorptionen av Mo är överlag stark i jordar vid lågt pH. Nya experiment har visat att Mo kan binda både till Fe/Al-hydroxider och organiskt material i det sexvärda oxidationstillståndet. I mer reducerande (sulfidiska) miljöer kommer Mo(VI) att regera med sulfid och bilda Mo(IV), vilket avsevärt minskar dess mobilitet. Detta är en viktig uppfångningsmekanism i sulfatreducerande färsk- och saltvattensmiljöer som kan leda till höga Mo-koncentrationer i sediment. Denna process tycks också vara viktig i Forsmarks sjö- och havssediment, som i vissa fall uppvisar markant högre Mo-koncentrationer är den lokala moränen. De naturliga Mo-koncentrationerna i ytnära grundvatten ligger normalt i spannet 0.1–10 μg L^{-1} med högre nivåer vid högre pH. Den huvudsakliga källan är förmodligen vittring av albit i de lokala jordarna, med det finns också en deposition från atmosfären, huvudsakligen kopplad till stålindustrin. För närvarande övervakas bara depositionen av Mo i Sverige på Visingsö.

I ekosystemmodellerna karaktäriseras Mo-93 av relativt låga K_d-värden. Jämfört med andra radionuklider förväntas en relativt hög andel av Mo-93 ansamlas in torv och postglaciala sediment. Utöver den höga mobiliteten bidrar även höga doskonverteringsfaktorer och en hög biotillgänglighet till de relativt höga doserna för Mo-93 i de nuvarande modellerna. Den dominerande exponeringsvägen förväntas vara intag av förorenad föda, där det mest kritiska scenariot är när en myr dräneras och omvandlas till jordbruksmark.

Contents

1 Introduction

The management of radioactive waste in Sweden involves three final repositories, one for short-lived waste (SFR), one for long-lived low- and intermediate level waste (SFL) and one for spent nuclear fuel (Figure 1-1). So far only SFR is in operation. As preparations now are being made to expand SFR in order to make room for decommissioning waste from Swedish nuclear power plants, Mo-93 has emerged as an important radionuclide in the safety assessments. Preliminary calculations indicate that Mo-93 could be one of the dominating contributors to the total radiation dose some thousands of years after closure (Figure 1-2). This result was not unexpected, but because Mo-93 has not been important in previous safety assessments it has received relatively little attention so far. However, with the closing down of nuclear reactors and the planned expansion of SFR the matter has now been brought to the fore.

In order to get an overview of how well the behaviour of Mo, and specifically Mo-93, in the repository and in the environment is understood and handled in the safety assessment a workshop on Mo from the perspective of a geological repository was arranged 14th June 2016. The aim was to cover all relevant aspects of Mo-93 and Mo biogeochemistry in the environment all the way from the sources in the nuclear power plants to the planned repository and further on to the surface ecosystems and potential doses to exposed individuals. The workshop took place at Såstaholm Castle outside Stockholm. The list of participants and the final programme for the workshop are found in appendices 1 and 2, respectively. This report is meant to provide a summary of the presentations given at the workshop and the following discussions.

Different presenters used their own estimations of the inventories of Mo-93, which also is reflected in the rendition of the different presentations in this report. It is not the intention of this report to explicitly report the inventory of Mo-93 in the different parts of the Swedish system for radioactive waste.

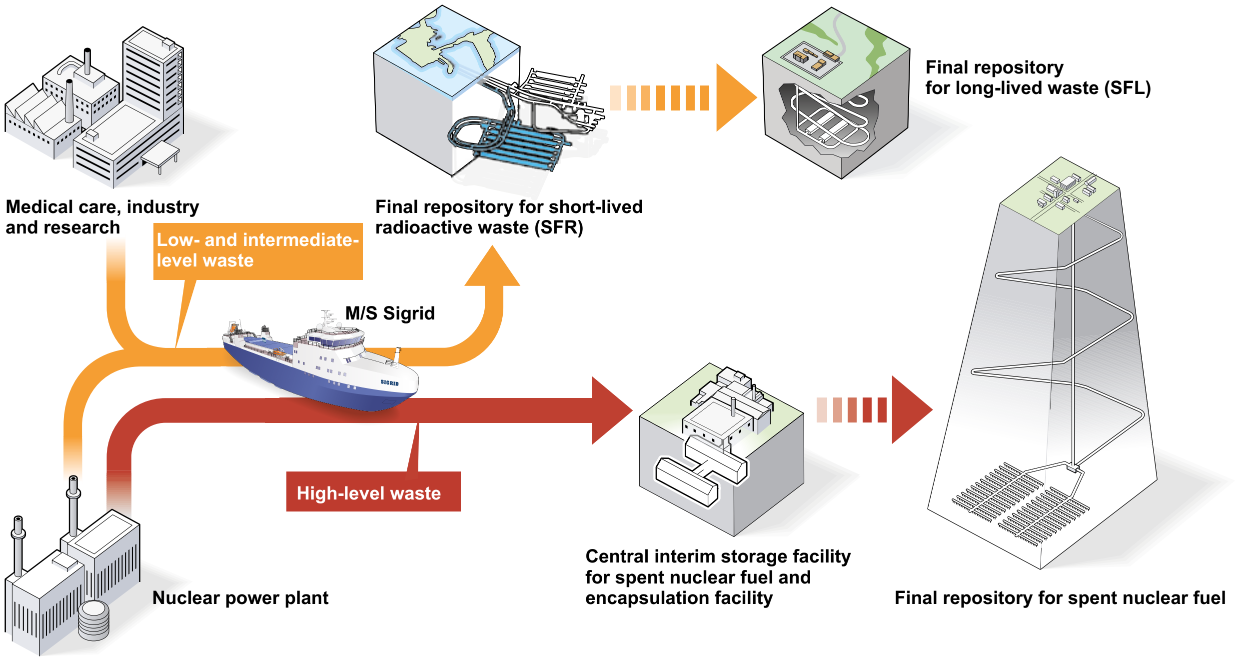

Figure 1-1. *SKB's system for managing Sweden's radioactive waste.*

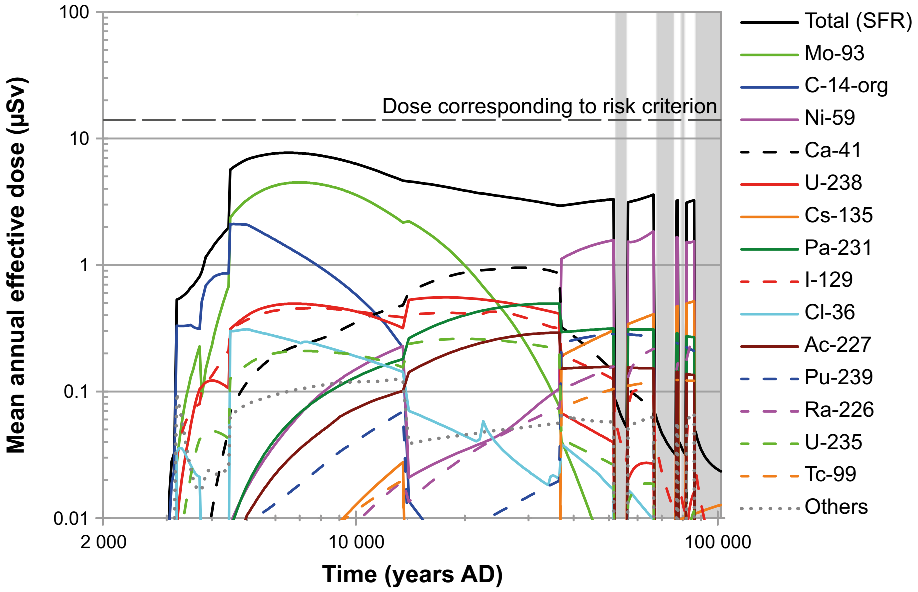

Figure 1-2. *Modelled dose from the final repository for low level waste (SFR). Mo-93 is predicted to dominate the dose during a period some thousands of years after closure. Shaded areas indicate periglacial conditions. The figure is taken from SKB (2015b).*

2 Summary of the presentations and discussions

2.1 Sources of Mo-93 in radioactive waste

The following section is based on the presentations by Merja Pukari, Klas Lundgren, Björn Herschend and Katrin Ahlford.

Mo-93 has a half-life of 4 000 years and is produced in nuclear power plants by neutron activation of Mo-92, a stable naturally occurring Mo isotope. Mo-93 will therefore occur whenever Mo-containing materials have been exposed to neutron radiation. Especially sensitive are reactors parts, which are made of Mo alloys and therefore contain high concentrations of Mo. Examples of this is Inconel 718, which is used in some fuel spacers in light-water reactors (LWRs), and stainless steel type 316, which is used in control rods and some reactor internals in LWRs. Mo-93 is, however, also produced elsewhere, since Mo exists as an impurity in many reactor materials. As a result Mo-93 will also be found in waste from pressurised water reactors (PWRs). The generated activity of Mo-93 is therefore dependent on factors such as the choice of fuel type and reactor material, especially the use of Mo alloys, but also in general the Mo concentration of neutron-exposed surfaces, operation time etc. The introduction of new Mo alloys in reactors has consequently caused an increased uncertainty in the amount of Mo-93 generated.

Mo-93 decays by electron capture to niobium-93 (Nb-93), which is observationally stable. The decay results in a few low-energetic gamma lines, the most intense ones being 16.6 keV (40 %) and 16.5 keV (21 %). No major gamma emissions are, however, found above 20 keV, which means that Mo-93 is not easily detected by radiometric methods. Any direct determination of Mo-93 by gamma spectrometry as well as mass spectrometry requires chemical separation of Mo. A fundamental difficulty is therefore that there is no straightforward method to directly quantify the inventory of Mo-93 in nuclear waste. Instead the inventory of Mo-93 must be estimated from more easily detected radionuclides such as technetium-99m (Tc-99m) and Mo-99. However, while Mo-93 is produced exclusively by neutron activation of Mo-92, Mo-99 derives both from neutron activation of Mo-98 and fission of U and Pu. It is therefore necessary to separate neutron activation from fission in order to estimate Mo-93.

At present the activity of Mo-93 in SFR amounts to ca 5.4×10^9 Bq, but the amount is expected to increase (Figure 2-1). The major source will, however, be the decommissioning of the nuclear power plants. This is predicted to result in 8.1×10^{12} Bq Mo-93 for BWRs, of which 56 % is expected to be found in the control rods. For PWRs the activity will be somewhat lower, 5.7×10^{12} Bq Mo-93.

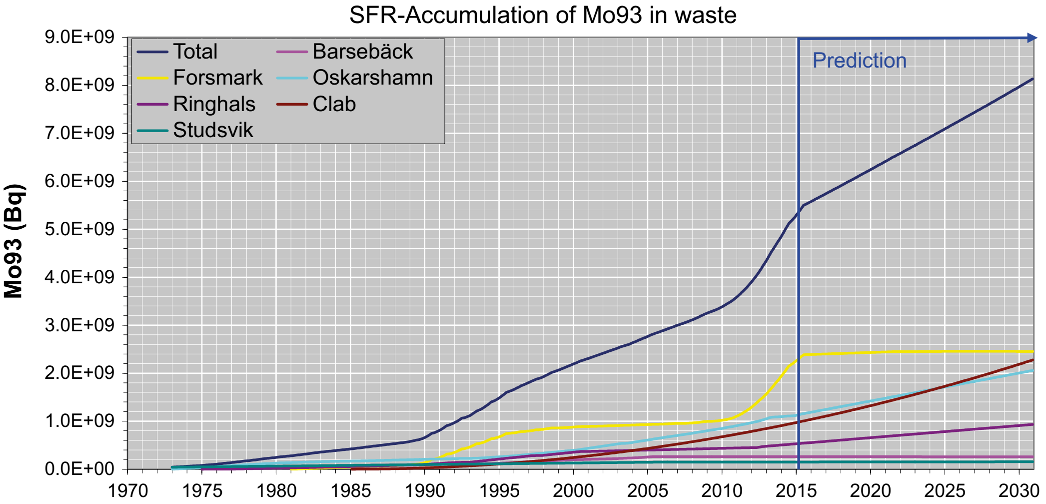

Figure 2-1. Current assessment of Mo-93 in reactor waste for SFR from the nuclear power plants Forsmark, Ringhals, Barsebäck and Oskarshamn plus the Studsvik facility and the interim storage for nuclear fuel (Clab). Early decommissioning of reactors from Oskarshamn and Ringhals is not considered.

In order to improve the assessment of Mo-93 stable Mo is now being measured in reactor water and feedwater at the nuclear power plants in Ringhals and Forsmark, but not in Oskarshamn. Such measurements will improve the assessment the amount of stable Mo exposed to neutron radiation in the reactors, leading to better estimations of the production of Mo-93. There are currently no plans to measure Mo-93 on a regular basis, but it has been proposed that Mo-93 should be measured in reactor water from selected BWRs, pool water from Clab, system surfaces and fuel crud in special campaigns in order to increase the credibility of the employed models for calculating Mo-93.

Because of the difficulties to measure Mo-93 the estimations of Mo-93 in nuclear waste are based on regular measurements of easily detected radionuclides, i.e. Tc-99m, Mo-99, I-131 and Cs-137, and concentrations of the relevant isotopes in the reactor. Many model parameters needed to calculate Mo-93 are fitted to Mo-99, but since Mo-99 derives both from neutron activation and fission, these two Mo isotopes cannot be directly compared. A flow chart for the calculations is shown in Figure 2-2.

The model is currently being improved by adding more datasets of measured radionuclides. For instance, the initial corrosion of fuel spacers is not modelled satisfactory with the present models, but a better model is under development, which will determine Mo-93 in fuel crud based on Co-60 and Mo-93 from the corrosion of fuel spacers based on the Mo-99 that cannot be explained by other sources. The calculated process water concentrations that are used as input to the model will also be reactor-specific.

It is not enough, however, to determine the activity of produced Mo-93. It must also be distributed between specific waste packages. This is done in proportion to the measured Co-60 in each waste package. Although the correlation between Co-60 and Mo-93 is not ideal, this is still considered a reasonable approach.

Depending on the total activity the waste packages will then be deposited in different parts of SFR and SFL. According to SKB's current estimations SFR is expected to store 1.6×10^{10} Bq of Mo-93 from operational waste (e.g. ion exchange resins) and another 1.3×10^{10} Bq of Mo-93 from decommissioning waste (reactor pressure vessels from BWRs, intermediate level scrap metal and ion exchangers from decontamination of reactor pressure vessels). Activated core components and BWR control rods (2.4×10^{13} Bq of Mo-93) will be stored in SFL. Then there will also be ca 6.5×10^{12} Bq of Mo-93 in legacy waste (intermediate level trash and scrap), which will also will be disposed of in SFL. The activity of Mo-93 in this type of waste is highly uncertain, and the estimates are based on Mo-93/Co-60 ratios in activated steel from the Studsvik R2 research reactor. A detailed distribution of Mo-93 and other radionuclides between different compartments of SFR is presented in Table 2-1. The silo is the most sophisticated part of SFR, which will store the most radioactive waste. BMA and BLA are rock vaults for intermediate level and low level waste, respectively.

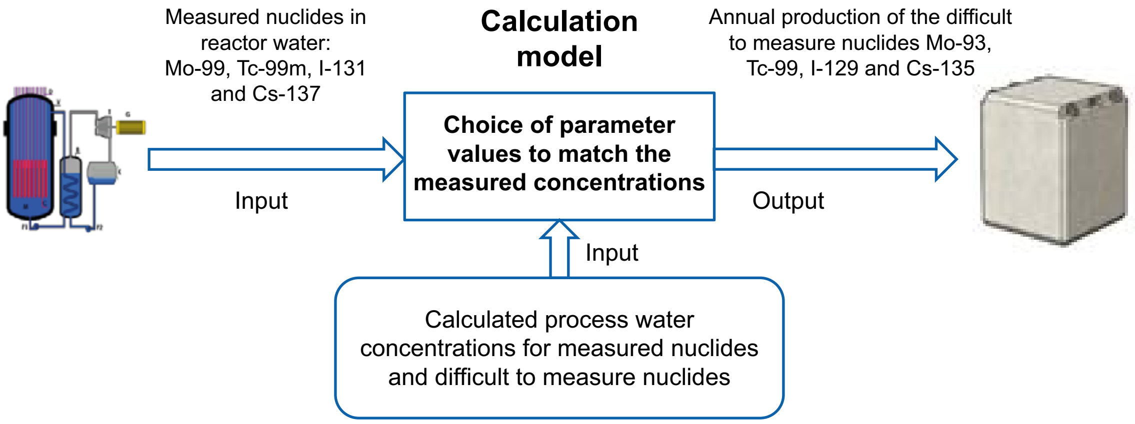

Figure 2-2. *Flow chart for the calculation of the annual production of difficult to measure radionuclides.*

Table 2-1. Best estimate of the radionuclide inventory (Bq) in SFR in year 2075 based on past and future operation of the Swedish nuclear power plants. C-14 is reported as organic, inorganic and induced activity. Data taken from SKB (2015a).

Nuclide	1BMA	2BMA	1BTF	2BTF	Silo	1BLA	2-5BLA	BRT	Total
H-3	8.09E+08	3.31E+12	6.82E+07	1.07E+08	8.97E+09	2.00E+08	1.94E+11		3.52E+12
Be-10	2.21E+05	2.19E+04	1.37E+04	2.48E+04	9.89E+05	6.53E+02	1.26E+03		1.27E+06
C-14 org*	1.47E+11	3.96E+09	9.84E+09	6.07E+09	7.56E+11	7.91E+07	2.25E+08		9.23E+11
C-14 inorg*	1.90E+12	1.44E+10	1.89E+11	2.69E+11	2.72E+12	4.03E+09	9.27E+08		5.10E+12
C-14 ind*		5.09E+09					1.19E+09	1.02E+10	1.65E+10
Cl-36	3.34E+08	2.02E+08	1.44E+07	1.66E+07	8.94E+08	2.17E+07	4.60E+07	7.21E+06	1.54E+09
Ca-41		1.56E+10					3.91E+09		1.95E+10
Fe-55	5.35E+10	1.05E+11	8.33E+07	1.14E+08	2.73E+12	8.78E+06	4.45E+08	1.49E+10	2.91E+12
Co-60	4.08E+11	1.99E+12	1.67E+10	2.36E+10	1.29E+13	1.03E+09	2.59E+10	1.93E+11	1.55E+13
Ni-59	2.10E+12	9.50E+11	3.31E+10	3.83E+10	6.85E+12	3.99E+09	1.15E+10	1.60E+11	1.01E+13
Ni-63	1.47E+14	9.23E+13	2.04E+12	2.27E+12	5.48E+14	3.04E+11	1.12E+12	1.44E+13	8.07E+14
Se-79	2.10E+08	7.29E+06	1.57E+07	1.54E+07	1.05E+09	4.00E+05	5.94E+06		1.31E+09
Sr-90	5.49E+11	3.60E+11	3.48E+10	5.76E+10	3.61E+12	7.42E+08	2.40E+10	2.32E+10	4.66E+12
Zr-93	3.68E+08	1.06E+09	2.29E+07	4.14E+07	4.48E+09	1.09E+06	2.95E+07	1.84E+08	6.19E+09
Nb-93m	1.73E+10	1.31E+13	1.44E+09	2.35E+09	9.33E+12	7.68E+07	1.34E+11	1.06E+12	2.36E+13
Nb-94	3.67E+09	9.12E+10	2.53E+08	4.13E+08	8.67E+10	3.14E+07	9.81E+08	7.94E+09	1.91E+11
Mo-93	1.46E+09	4.52E+09	2.56E+08	2.36E+08	1.96E+10	1.01E+08	9.01E+07	3.00E+09	2.93E+10
Tc-99	6.22E+09	1.42E+09	2.30E+09	5.45E+08	5.00E+10	1.85E+09	4.98E+08	4.49E+08	6.32E+10
Pd-107	5.25E+07	2.55E+09	3.92E+06	3.86E+06	2.75E+08	1.00E+05	1.72E+06		2.89E+09
Ag-108m	1.95E+10	4.06E+10	1.51E+09	2.21E+09	2.30E+11	1.94E+08	1.53E+09	1.62E+09	2.97E+11
Cd-113m	7.98E+08	9.32E+07	7.67E+07	6.34E+07	9.58E+09	1.96E+06	6.13E+06		1.06E+10
In-115		3.13E+05							3.13E+05
Sn-126	2.62E+07	1.75E+07	1.96E+06	1.93E+06	2.05E+08	5.00E+04	7.93E+06	7.53E+05	2.62E+08
Sb-125	4.37E+07	2.62E+08	7.47E+06	1.04E+07	1.32E+11	4.74E+05	4.46E+06	1.34E+07	1.32E+11
I-129	1.46E+08	7.67E+06	2.27E+07	1.02E+07	9.84E+08	4.35E+05	1.94E+06		1.17E+09
Cs-134	1.45E+08	2.26E+08	7.10E+04	8.86E+04	2.20E+11	1.58E+04	1.39E+06		2.20E+11
Cs-135	8.41E+08	5.33E+07	1.03E+08	1.85E+07	4.47E+09	3.07E+06	1.75E+08		5.67E+09
Cs-137	8.15E+12	8.95E+11	7.12E+11	6.22E+11	5.97E+13	1.84E+10	4.95E+11		7.05E+13
Ba-133	4.89E+07	1.43E+08	4.03E+06	6.19E+06	6.16E+08	2.20E+05	1.26E+07		8.31E+08
Pm-147	3.71E+08	4.06E+08	3.84E+06	4.57E+06	3.59E+11	3.02E+05	1.19E+06	1.37E+06	3.60E+11
Sm-151	8.26E+10	3.55E+10	6.51E+09	6.13E+09	4.63E+11	1.68E+08	5.88E+09	3.42E+08	6.00E+11
Eu-152	9.47E+07	1.33E+11	6.19E+07	6.54E+06	8.64E+08	1.02E+08	1.73E+10	5.41E+05	1.52E+11
Eu-154	2.33E+10	6.83E+09	1.98E+09	1.80E+09	5.24E+11	4.01E+07	2.67E+08	9.27E+07	5.59E+11
Eu-155	1.02E+09	3.74E+08	4.96E+07	5.83E+07	9.96E+10	1.54E+06	1.16E+07	2.40E+06	1.01E+11
Ho-166m	1.41E+09	5.22E+08	8.79E+07	1.59E+08	6.83E+09	4.18E+06	9.03E+07	7.99E+03	9.10E+09
U-232	8.85E+04	1.46E+05	1.62E+04	6.73E+03	6.20E+05	2.34E+03	9.35E+03	6.86E+03	8.96E+05
U-234	6.66E+06	3.04E+06	9.86E+05	4.55E+05	3.58E+07	1.33E+05	4.38E+05		4.75E+07
U-235	3.00E+06	7.82E+04	1.84E+07	1.12E+05	1.42E+07	2.98E+08	3.23E+08	1.49E+01	6.57E+08
U-236	2.64E+06	6.00E+06	4.02E+05	3.55E+05	1.58E+07	3.99E+04	2.06E+05	3.92E+05	2.59E+07
U-238	5.95E+06	1.23E+06	8.55E+05	8.75E+05	3.28E+07	7.33E+08	1.77E+08		9.52E+08
Np-237	2.73E+07	7.68E+06	1.07E+06	1.98E+06	5.36E+08	6.75E+04	2.61E+05	4.70E+05	5.75E+08
Pu-238	7.52E+09	4.42E+10	2.09E+09	4.56E+08	7.29E+10	3.47E+08	1.52E+09	2.72E+09	1.32E+11
Pu-239	2.77E+09	6.78E+09	4.68E+08	1.89E+08	1.70E+10	6.60E+07	2.77E+08	4.16E+08	2.80E+10
Pu-240	3.87E+09	9.21E+09	5.20E+08	2.65E+08	2.39E+10	6.74E+07	2.95E+08	5.92E+08	3.87E+10
Pu-241	2.40E+10	1.66E+11	7.30E+09	2.42E+09	3.07E+11	1.29E+09	5.74E+09	9.05E+09	5.23E+11
Pu-242	2.00E+07	5.02E+07	2.96E+06	1.37E+06	1.23E+08	3.99E+05	1.71E+06	3.11E+06	2.03E+08
Am-241	2.91E+10	4.12E+10	6.14E+09	1.83E+09	2.32E+13	5.23E+08	1.94E+09	1.99E+09	2.32E+13
Am-242m	4.46E+07	1.83E+08	7.34E+06	3.21E+06	3.22E+08	1.02E+06	4.84E+06	1.32E+07	5.79E+08
Am-243	2.02E+08	6.62E+08	3.25E+07	1.78E+07	1.60E+09	4.00E+06	1.86E+07	4.14E+07	2.57E+09
Cm-243	1.85E+07	1.03E+08	3.82E+06	4.15E+05	1.89E+08	7.58E+05	3.40E+06	6.38E+06	3.25E+08
Cm-244	6.73E+08	1.07E+10	2.68E+08	2.84E+07	9.26E+09	5.39E+07	2.80E+08	6.76E+08	2.19E+10
Cm-245	1.99E+06	1.01E+07	2.95E+05	1.36E+05	1.49E+07	3.97E+04	2.18E+05	6.83E+05	2.84E+07
Cm-246	5.27E+05	3.34E+06	7.82E+04	3.60E+04	4.29E+06	1.05E+04	6.61E+04	2.24E+05	8.58E+06
Total	1.60E+14	1.14E+14	3.06E+12	3.30E+12	6.72E+14	3.39E+11	2.05E+12	1.59E+13	9.71E+14

2.2 Mo-93 in the repository, the near-field and the geosphere

2.2.1 Mo-93 in SFR

The following section is based on the presentations by Klas Källström, Ola Wessley and Mireia Grivé.

When assessing the safety of SFR there are two principles that need to be considered. First, there must be a substantial retention of radionuclides in the repository. In order to achieve this the design of the repository is a crucial factor. Second, the activity of long-lived radionuclides must be limited. This puts certain requirements on the waste that is brought to SFR. Figure 2-3 illustrates the evolution of the repository and relates the half-lives of key radionuclides to major processes affecting the repository during 100 000 years.

One important limit is when institutional control of the repository no longer can be considered as feasible. This could occur as soon as a few hundred years after closure. Relatively short-lived radionuclides such as Co-60 and Cs-137 can therefore be expected to largely disappear while the repository still is under institutional control, while more long-lived radionuclides such as Ni-59, Am-241, Mo-93 and C-14 must be considered to remain in significant activities when institutional control over the repository no longer is feasible.

The predicted release of Mo-93 from different parts of SFR is shown in Figure 2-4. It is assumed that Mo-93 in spent ion exchange resins is released instantly, whereas release of Mo-93 from reactor pressure vessels and other reactor parts is determined by corrosion rates.

After mobilisation from the waste Mo-93 is assumed to be present as molybdate, which can be transported by advection and diffusion through the engineered barriers to fractures in the bedrock. An increased retention by sorption in the barriers would results in less activity reaching the surface due to radioactive decay in the barriers. An important factor here is the sorption of Mo-93 onto the concrete and bentonite, which constitute the engineered barrier. If the retention is strong enough, the barriers will prevent Mo-93 from leaving the repository in large amounts. Figure 2-5 illustrates that the barrier thickness has a direct effect on the ability of the repository to retain Mo-93 and that concrete is much more efficient than bentonite in retaining Mo-93. For instance, 2.5 m of concrete buffer is estimated to delay the release of Mo-93 by approximately 10 000 years. Given that the half-life of Mo-93 is 4 000 years and that there is no further ingrowth of Mo-93 this would have a significant effect on the release of Mo-93. These numbers can also be compared to the corrosion of 1 cm steel, which is estimated to take up to 100 000 years.

When comparing the total activity of Mo-93 in different parts of SFR to the corresponding void, it is estimated that the highest concentrations potentially would occur in the silo. An upper limit for the Mo-93 concentration in the silo is 1.34 nM. Compared to the concentration of the naturally occurring stable Mo isotopes in the deep groundwater the addition of Mo-93 will be negligible for total Mo concentration, and it is therefore not expected to cause precipitation of any Mo-bearing minerals.

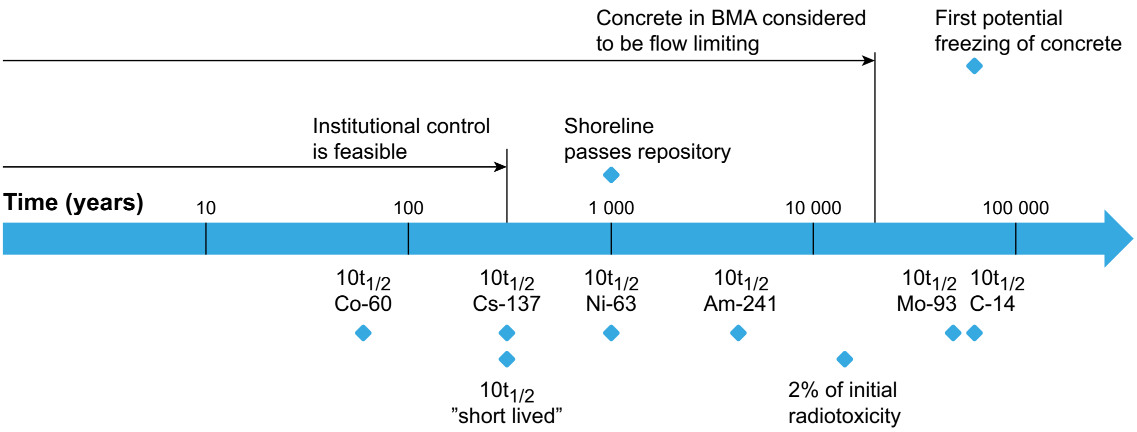

Figure 2-3. *Time line comparing the decay of key radionuclides to important processes during the first 100 000 years after closure. Figure taken from SKB (2015a).*

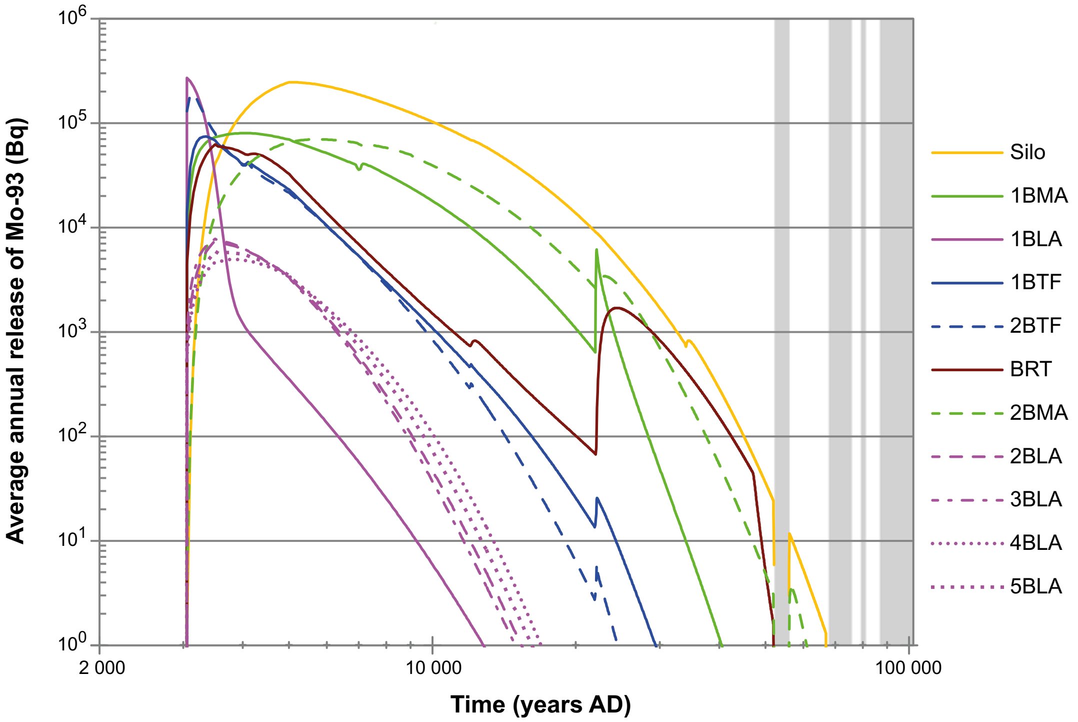

Figure 2-4. *Predicted release of Mo-93 from different parts of the repository for low level radioactive waste. The silo is the most sophisticated repository for the most radioactive waste, mainly from the primary circuit. BMA, BTF and BLA are rock vaults for intermediate level and low level waste. The figure is taken from SKB (2015a).*

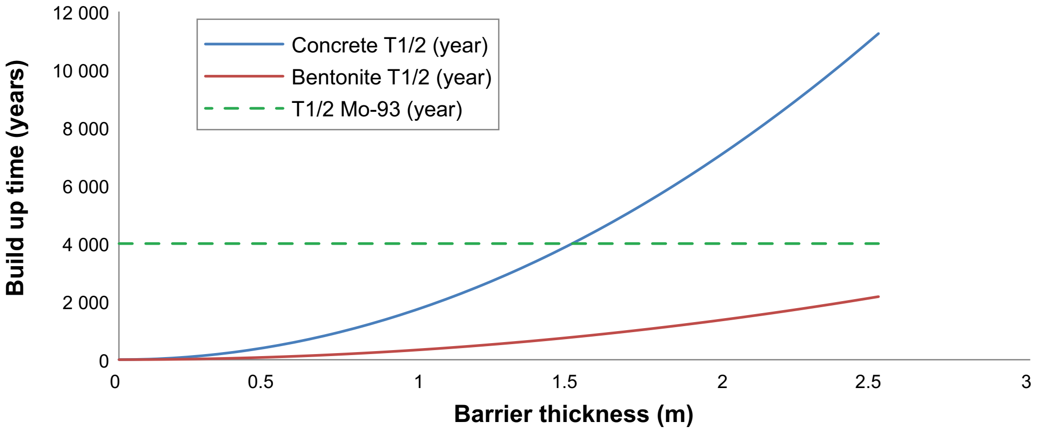

Figure 2-5. *Predicted delay in the engineered barriers in comparison to the half-life of Mo-93.*

The sorption properties of molybdate onto different minerals are in general not very well investigated. Mo sorption onto magnetite, which is one expected corrosion product, has been found to be very low (Rovira et al. 2006). Further research would for example be needed to elucidate the potential reducing effect of structural Fe in magnetite. In cementitious matrices the sorption of molybdate is assumed to be similar to selenate, which in turn is related to the presence of ettringite and AFm phases (alumina, ferric oxide, monosulphate) (Ochs et al. 2016, Chapter 8). Generally, the sorption of oxyanions is believed to be either adsorption onto the surfaces or direct partial or complete substitution of sulphate or hydroxide ions in the interlayer of these phases. There are, however, several open questions related to the sorption of Mo in concrete. One question concerns the role of C-S-H (calcium silicate hydrate) vs. AFm and Aft (alumina, ferric oxide, trisulphate). It is also largely unknown how the sorption properties change as the concrete ages. Furthermore, it is likely that sorption of molybdate is dependent on the occurrence of competing anions. These questions are currently being investigated in the EU-funded Cebama project, where Andra, SKB and other stake-holders also participate. The main retention mechanisms of Mo in cement and the pure cement phases are studied both experimentally and by modelling (Grivé and Olmeda 2015). The project is expected to result in an improved understanding of the retention mechanisms of molybdate in concrete and determination of parameters affecting its mobility.

2.3 Mo-93 in the geosphere

The following section is based on parts of the presentation by Mireia Grivé.

Mo-93 is assumed to occur as molybdate also in the deep groundwater. Since minerals at normal pH are mostly negatively charged, molybdate is not expected to sorb strongly in the bedrock. Possible minerals for molybdate sorption would primarily be those, which have point of zero charge near normal pH, e.g. goethite, hematite, magnetite and certain aluminosilicates. Generally, sorption of Mo is favoured by high Fe and Al oxide content, high clay content and high organic matter content (Bibak et al. 1994, Bibak and Borggaard 1994, Brinton and O'Connor 2003, Goldberg et al. 1996, Lang and Kaupenjohann 2000, Mikkonen and Tummavuori 1993). As in the repository, competition with other anions for sorption sites is also believed to be important for the retention of Mo.

In contrast to the repository and the surface systems the bedrock is currently assumed not to sorb Mo at all in SKB's safety analysis (SKB 2014). This is a conservative assumption, allowing Mo-93 to be transported freely with the groundwater to the surface ecosystems. Because of the relatively short half-life of Mo-93 (4000 years) the activity that would reach the surface would be highly sensitive to retention processes in the bedrock. It would therefore be important to investigate more thoroughly whether a non-zero K_d value for the bedrock could be justified or not. For instance, the role of Mo in microbially mediated redox reactions and the role of sulphide in the retention of Mo would need to be addressed.

2.4 Mo-93 in the surface ecosystems

2.4.1 Mo chemistry in forest ecosystems

The following section is based on the presentation by Jon Petter Gustafsson.

Mo is an important micronutrient, not least for the biogeochemical cycling of nitrogen, where Mo-containing enzymes play a central role for fixation of nitrogen and reduction of nitrate. Mo deficiency has been observed mainly in tropical soil, where weathering has depleted the soils of Mo, but the availability of Mo is also dependent on pH and interactions with other oxyanions such as sulphate and phosphate. Mo is, however, also toxic at high concentrations because it interferes with the uptake of copper. For example, it is well known that Mo may cause copper deficiency in ruminants.

The sorption of Mo in soils is generally strong at low pH, which mainly has been attributed to sorption onto Fe/Al hydroxides. Recent experiments from Swedish soils have, however, demonstrated that the sorption of Mo is also strong in organic soils with little or no Fe/Al hydroxide. In the study of Gustafsson and Tiberg (2015) molybdate was added to two podzolic soil samples, one representing the Oe horizon (mor layer) and one representing the Bs horizon. XANES and EXAFS were then used to determine the oxidation state and coordination of the bound Mo. The results indicated that Mo in all cases was present as Mo(VI), i.e. no reduction had occurred. In the B horizon most of the molybdate was bound to an $Al(OH)_3$-type phase (which was probably allophane, an amorphous aluminosilicate with Al-OH groups known to be important in the B horizons of podzols). Both on ferrihydrite and amorphous $Al(OH)_3$ the EXAFS results showed molybdate to coordinate as an edge-sharing bidentate complex. This was, however, not sufficient to reproduce the measurements in a geochemical model, which required also the presence of an outer-sphere complex at high pH (which would not be observed by EXAFS spectroscopy). On organic matter Mo(VI) is complexed to carboxylate groups at low pH. A limitation with the study was, however, that the use of EXAFS spectroscopy required Mo(VI) to be present in much higher concentrations than those usually found in the natural environment. In reality, it is possible that Mo may also be coordinated to thiol (-SH) groups, which occur in organic matter at low concentration (Gustafsson and Tiberg 2015).

2.4.2 The geochemical behavior of Mo in modern sediments

The following section is based on the presentation by Tais Wittchen Dahl.

Mo is highly soluble in O_2-rich sea water. As a result, Mo is the most abundant transition metal in sea water with a residence time in the oceans of 440 000 years. Mo is slowly scavenged by Mn oxides, but also Fe oxides contribute. Metal oxides are, however, not preserved in sediments so Mo can be released again, causing high dissolved Mo concentrations in the manganeous zone. In sulphidic water molybdate reacts with sulphide to form thiomolybdates. This conversion is quantitative when the H_2S concentration exceeds 11 µM. Subsequent burial and fixation of Mo in the sediments is known to involve a reduction of Mo(VI) to Mo(IV), which can lead to Mo concentrations of more than 100 $\mu g\ g^{-1}$. Both Fe sulphides and organic matter contribute to this Mo fixation. Hence, the availability of sulphide plays a central role for the removal of Mo from the water column and its subsequent immobilisation in the sediments.

Similar enrichment of Mo in sediments has been reported in a freshwater system, Lake Cadagno in Switzerland (Dahl et al. 2010). The lake has a maximum depth of 19 m and is known to be stratified all year round. The bottom water contains 250–300 µM of sulphide, generated from sulphate reduction in the water column and in the sediments. A distinct decrease in the concentration of dissolved Mo and an increase in the concentration of particulate Mo can be observed in the boundary between oxic and sulphidic water.

The exact pathway for Mo fixation in sediments is not fully understood, but it is clear that the reaction of molybdate with sulphide is a key process, which opens up new reaction pathways (Helz et al. 2011, Wang et al. 2011). The final product is not molybdenite (MoS_2), but a configuration with S and Fe that resembles Mo in nitrogenase (Dahl et al. 2013).

2.4.3 Mo in the Forsmark area

The following section is based on the presentation by Fredrik Lidman.

The site investigation in Forsmark has resulted in a large dataset, including more than 500 observations of Mo in sea water, lake water, groundwater, stream water and various types of soils and sediments.

The local till in Forsmark contains Mo concentrations ranging from $< 0.1\ \mu g\ g^{-1}$ to ca 1.4 $\mu g\ g^{-1}$. Globally, most Mo in soil is believed to occur in feldspars and plagioclase, but it can also be present in other minerals such as quartz and biotite (Ahrens et al. 2013). The mineralogy in Forsmark is dominated by quartz, albite, microcline, calcite, magnesiohornblende, illite and clinochlore (Sohlenius and Rudmark 2003). Compared to other analysed elements in the till Mo correlates most strongly to Na (Figure 2-6). Since the major Na-bearing mineral in Forsmark is albite, the Na endmember mineral of the plagioclase solid solution series, this suggests that albite also could a major source for Mo in Forsmark.

In the near-surface groundwater, however, there is no correlation between Mo and Na. Since the biogeochemistry of Mo is so different from that of Na, this lack of correlation is not unexpected even if it is assumed that they both partly derive from weathering of albite. Furthermore, not all groundwater samples represent till. There are also other types of Quaternary deposits in the Forsmark area, and a better classification and description of the different sampling locations are probably necessary in order to allow a more detailed analysis of the spatial variation of Mo and other elements in the groundwater of Forsmark. One overall trend for Mo is, however, that that high Mo concentrations are found particularly in groundwater with high pH, whereas groundwater with low pH tends to have low Mo concentrations (Figure 2-7). It is has not been clarified yet whether this reflects some general gradient in the mineralogy or the type of groundwater that is sampled or whether this is an effect of lower mobility of Mo at low pH.

As previously discussed, one major sink for Mo in the environment is fixation in sediments. The accumulation of Mo in marine sediments is well-documented, but similar processes have been observed also in a freshwater lake in Switzerland (Dahl et al. 2010, 2013). The lakes in Forsmark are arguably very different from Lake Cadagno, for instance in that they are shallow, young and strongly influenced by the presence of calcite in the Forsmark area. A pertinent question for the safety analysis is therefore whether or not Mo-93 can be expected to accumulate in lake sediments in Forsmark.

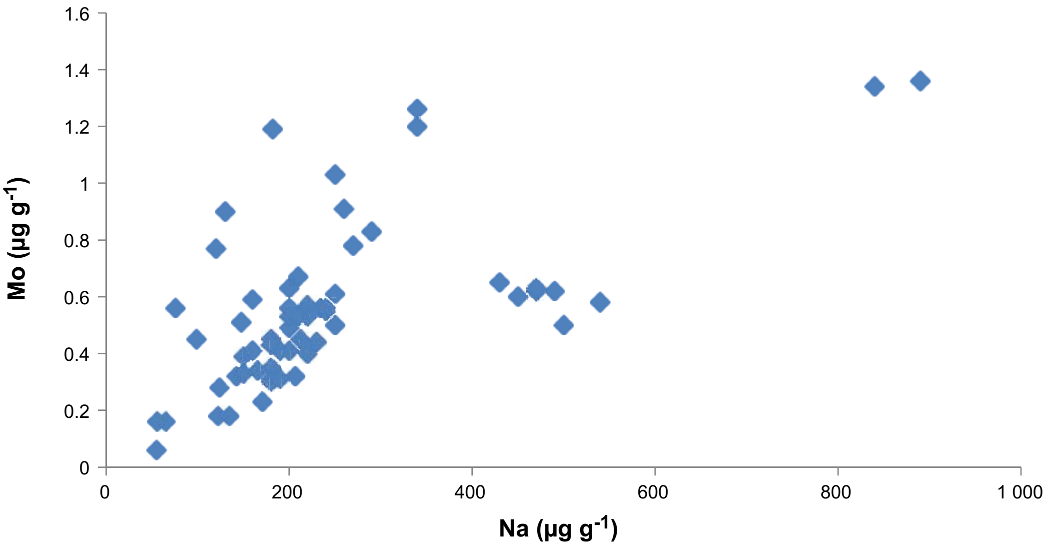

Figure 2-6. *Mo vs. Na in till samples from Forsmark.*

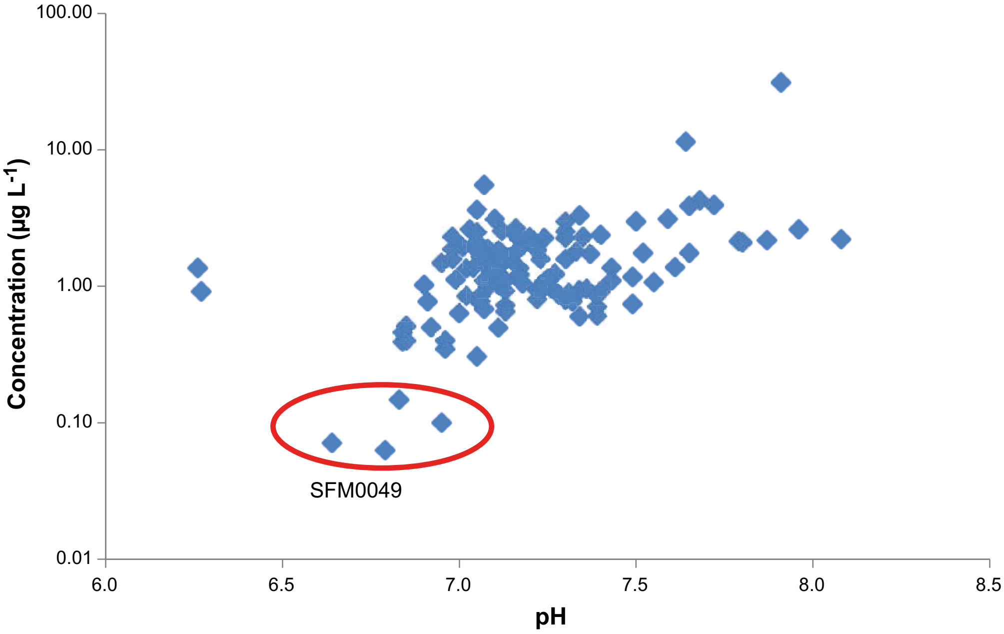

Figure 2-7. *Mo in near-surface groundwater from Forsmark as a function of pH. Particularly low Mo concentrations have been observed in SFM0049, which is characterised by high sulphide concentrations.*

Figure 2-8 illustrates the concentrations of Mo, U and Ni in a sediment core from Stocksjön, a small lake, ca 100 m in diameter, with an average depth of less than 1 m. The Mo concentrations throughout the sediment core are notably higher than in local till, but especially around 0.4 m deep there is a distinct peak with Mo concentrations up to 76 µg g^{-1}. This is notably higher than the Mo concentrations in local till, which has an average of 0.55 µg g^{-1} (n=59) and an observed maximum of 1.36 µg g^{-1}. High Mo/Na and Mo/Ti ratios also suggest that there must have been a preferential enrichment of Mo in these sediments. The Mo peak also coincides with a peak for U, which is another redox-sensitive element. As for Mo, the mobility of U would be expected to decrease substantially if it is reduced, in this case from U(VI) to U(IV). Ni, in contrast, exhibits no peak at this depth, but then Ni is not redox-sensitive. Combined these observations indicate that Mo can be immobilised and accumulate to high concentrations in sediments even in the shallow lakes of the Forsmark area. Elevated Mo concentrations have also been reported from Eckarfjärden, up to 8 µg g^{-1} in gyttja sediments, and 17.9 µg^{-1} in a so-called microbial mat. Futhermore, high Mo concentrations have been observed in marine sediments from Tixelfjärden (17.7 µg^{-1}) and in clayey gyttja from a wetland by Puttan (14.2 µg^{-1}). More information on Mo in the Forsmark area will be presented in an upcoming SKB report.

2.4.4 Atmospheric deposition of Mo

The following section is based on the presentation by Per-Erik Karlsson.

Wet deposition is monitored every month at five sites across Sweden and one in Finland. Mo, however, is currently only monitored at the site on Visingö (Figure 2-9). There the wet deposition ranges from 20–60 µg m^{-2} a^{-1}. A weak correlation has been observed between Mo and Cr, which could indicate that the deposition is dominated by long-range transport. It is likely that the steel industry is the major source for both Cr and Mo.

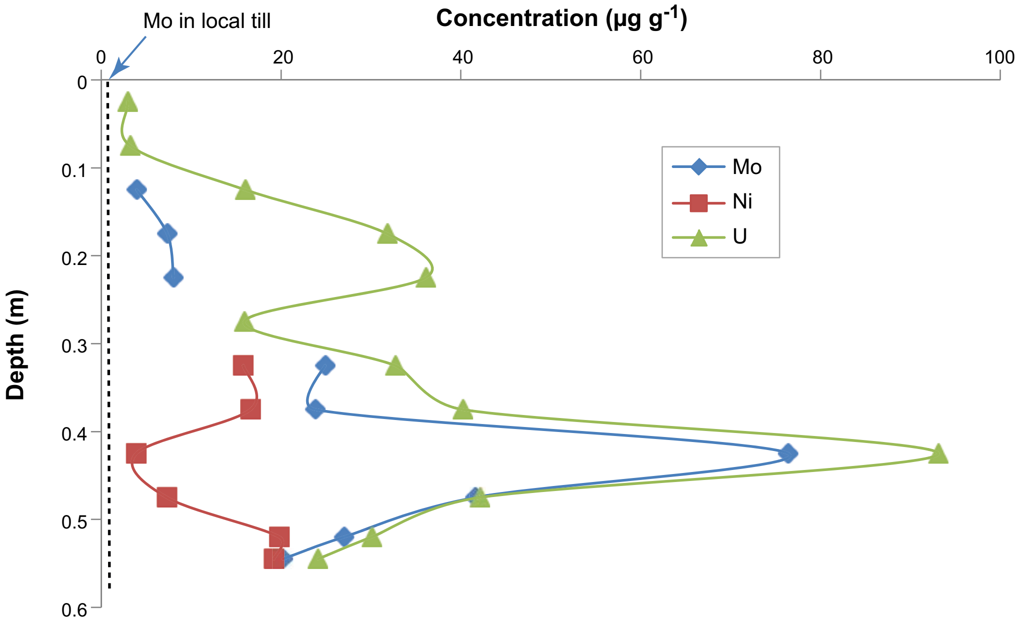

Figure 2-8. *Mo, U and Ni concentrations in sediments from Stocksjön.*

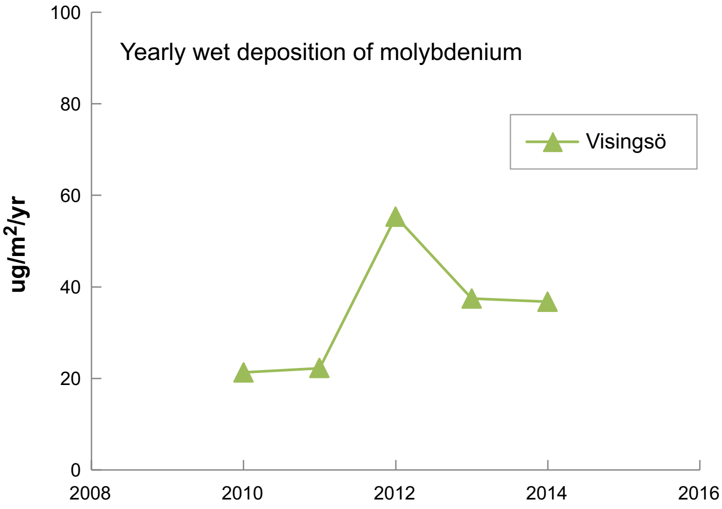

Figure 2-9. *Annual wet deposition of Mo at Visingsö.*

Mosses have also been used to assess the deposition of heavy metals because they have no roots and therefore take up all nutrients (and other substances) from wet and dry deposition. 2–3 year old mosses have been sampled across Sweden every five years since 1975. So far the concentration in mosses has not been quantitatively linked to the deposition, but on a qualitative level the concentrations reflect the reported emissions fairly well. High Mo concentrations have for instance been reported near Sandviken and in Bergslagen, where several iron works are located (Figure 2-10).

Deposition is also monitored in the Swedish throughfall monitoring network (Krondroppsnätet), which has been running since 1985 and has ca 70 active sites today. In this programme air concentrations, deposition (bulk, dry deposition and throughfall) and soil water are measured on a monthly basis. At present, however, Mo is not included in the analyses.

2.4.5 Mo-93 in the biosphere models

The following section is based on the presentation by Peter Saetre.

The transport of Mo-93 and the associated dose is in the surface ecosystem is modelled from a landscape perspective, where different ecosystems such as lakes, streams, mires, forests, agricultural land and sea are represented (Figure 2-11). Site-specific K_d values are used for different materials in the Quaternary deposits, e.g. peat, postglacial sediments, glacial sediments etc. Mo generally has fairly low K_d values, but they tend to be one to two orders of magnitude higher in younger, more organic soils, e.g. peat and postglacial sediments, than in deeper, less organic soils. Compared to many other radionuclides a noticeably high fraction of the accumulated Mo-93 will be found in peat and postglacial sediments (Figure 2-12). The model does not explicitly account for redox processes, but relies on empirical K_d values. It is possible, however, that the higher K_d values that were measured in peat and postglacial sediments to some extent reflects a sulphide-mediated immobilisation of Mo in these environments.

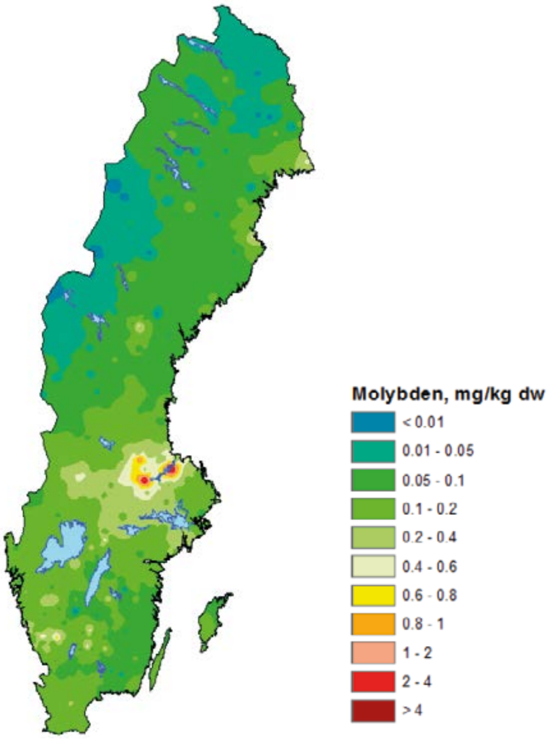

Figure 2-10. *Mo in moss 2015 (from Danielsson and Pihl Karlsson 2016).*

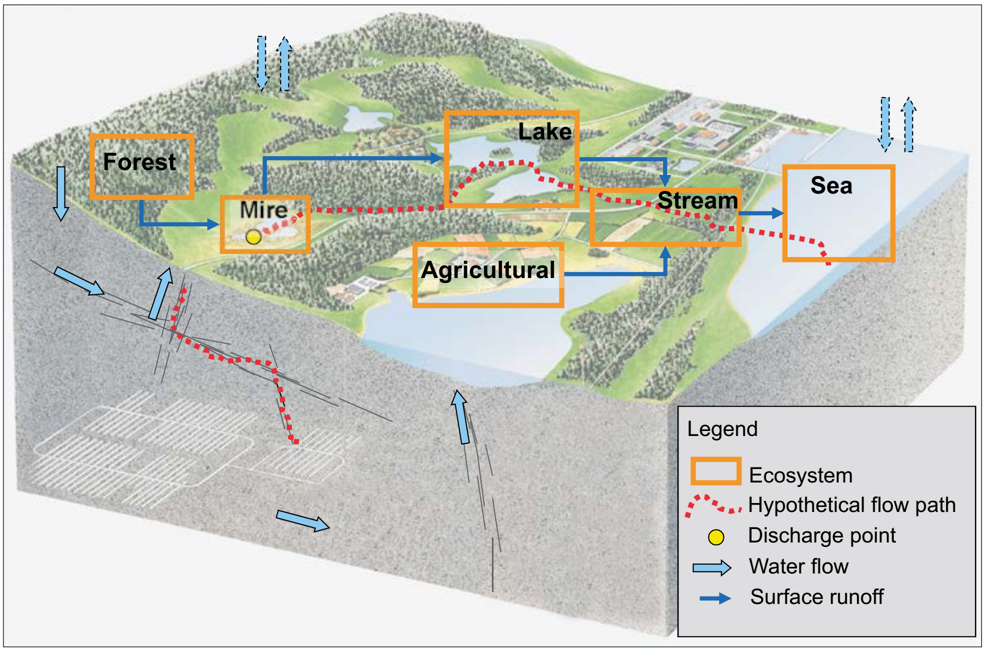

Figure 2-11. *Conceptual model for the transport of Mo-93 and other radionuclides in the surface ecosystems.*

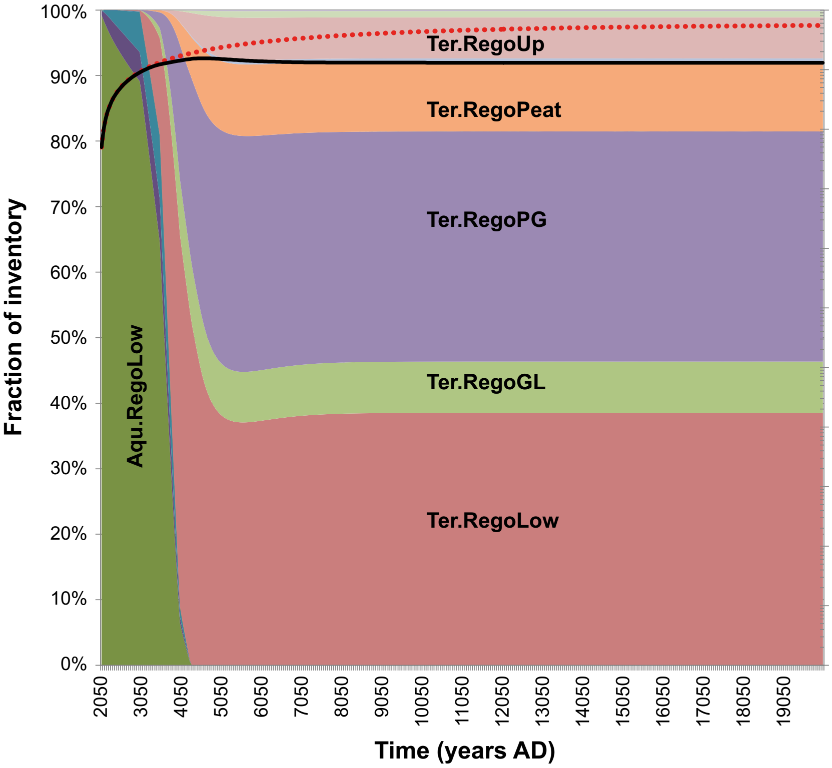

Figure 2-12. *Modelled distribution of Mo-93between different soil layers. Compared to other radionuclides Mo-93 is characterised by high accumulation in organic soil layers, e.g. peat (Ter.RegoPeat) and postglacial sediments (Ter.RegoPG).*

Mo-93 stands for only 0.03 % of the total expected activity in the waste sent to SFR, but there are several factors that combined indicate that Mo-93 still could contribute substantially to the total dose. One reason is the relatively high mobility of Mo in the engineered barriers, the bedrock and the deeper soils in combination with a lower mobility in the upper more organic soils. Furthermore, the dose coefficients for Mo-93 are fairly high, 3.1×10^{-9} Sv Bq^{-1} for ingestion and 2.3×10^{-9} Sv Bq^{-1} for inhalation (Grolander 2013). In addition, the bioavailability of Mo is also fairly high (as reflected in the CR values) so Mo-93 can be expected to be taken up by organisms to a fairly high extent. This combination of radiotoxicity and bioavailability is summarised by the landscape dose conversion factor (LDF) (SKB 2014). The LDF is the annual lifetime-averaged effective dose to a representative individual from the most exposed group resulting from a constant unit release rate of a certain radionuclide to the biosphere, including both external and internal exposure. In the case of Mo-93 the LDF has been estimated to 5.5×10^{-12} Sv Bq^{-1}. This means that if 1 Bq of Mo-93 is released to the surface ecosystems annually over an extended period of time, this will cause a radiation dose of 5.5×10^{-12} Sv every year. Compared to other key radionuclides this is a high number – Mo-93 falls in the upper quartile of the LDF distribution of all modelled radionuclides. For instance, Mo-93 is nearly ten times worse than Cl-36, 200 times worse than Ni-59 and 700 times worse than C-14 when assuming an equal release to the surface systems.

The by far dominating exposure pathway for Mo-93 is expected to be ingestion of contaminated food (99.5 % of the dose generated from Mo-93). The most critical scenario is when mire is drained and converted into agricultural land. In a sensitivity analysis (not considering the landscape evolution) both CR and K_d values for Mo were identified as important parameters. Further improvement of the model could include smart K_d values, which for instance could be dependent on DOC concentrations and pH.

2.4.6 Mo-93 in the Finnish nuclear waste programme

The following section is based on the presentation by Thomas Hjerpe.

The Finnish counterpart to SKB, Posiva, has a radioactive waste programme that in many respects resembles the Swedish approach. The requirements from the regulators are, however, different, which makes it difficult to make direct comparisons. For instance, Posiva considers a time frame of 1 million years, but dose calculations are only made for the first 10 000 years after closure. SKB, however, makes dose calculations up to 1 000 000 years.

So far Mo-93 has been identified as an important radionuclide, but it has not been considered a major problem in Posiva's safety assessment. It should be noted, however, that Posiva only has conducted safety assessments for a spent nuclear fuel repository. The safety assessment of low and intermediate level waste repositories, where Mo-93 is expected to be more important, is the responsibility of the NPP operators. Ca. 3×10^{8} Bq Mo-93 is expected in operational waste and 1×10^{10} Bq Mo-93 in decommissioning waste.

Posiva has identified similar retention processes for Mo as SKB. One notable difference is, however, that Posiva have assigned a non-zero K_d value for Mo in the bedrock. As a consequence, ca 50–80 % of the activity of Mo-93 is retained in the geosphere according to the modelling. Another difference is that the site-specific K_d measurements of Mo in Olkiluoto surface environment are roughly an order of magnitude higher than the corresponding measurements in Forsmark. Similar to Forsmark, the K_d values are higher in topsoils. High Mo concentrations have been observed in macrophytes and roots, supporting that the biological uptake of Mo is fairly high. The dominant exposure pathway for Mo-93 in Posiva's calculations was ingestion of cow milk, but Mo-93 contributed with < 0.02 % of the annual dose to a representative individual in the most exposed group.

3 General discussion

The issue of Mo-93 in nuclear waste is associated with a number of uncertainties. One fundamental uncertainty is the activity of Mo-93 in the nuclear waste, since it is so difficult to measure directly. Measures are now being taken to improve the methods for determining Mo-93. The uncertainties in the current estimations are, however, not expected to be the main problem concerning Mo-93 in the safety assessment.

Further uncertainties concern the speciation of Mo, particularly in the presence of sulphide, and its interaction with different types of materials, not least in the presence of competing anions. On-going research is trying to achieve a better understanding and parameterisation of the interactions between Mo and concrete. It is clear that thicker concrete walls would have a direct effect on the release of Mo-93. However, it must also be considered whether it would be reasonable to assign a non-zero K_d value to the bedrock, as Posiva have, or whether the assumption that Mo-93 will be transported freely with the groundwater in the model is more correct. It is important that this is dealt with in a consistent way for SFR and SFL.

So far Mo-93 has been assumed to be present as molybdate in the near-field and in the bedrock. Given the impact of sulphide on the mobility of Mo it might be worth to consider whether the sulphide concentrations anywhere in the near-field or in the bedrock could be high enough to react with and reduce molybdate. That would have far-reaching implications for the mobility of Mo-93 and its possibilities to reach the surface ecosystems.

In the surface ecosystems immobilisation of Mo in sediments appears to be the key process that can cause high concentrations of Mo. There is strong evidence that such accumulation is happening in Stocksjön, but it needs to be assessed how wide-spread this immobilisation of Mo is in Forsmark. Primarily one could expect this preferential accumulation of Mo to occur in lake sediments, sea sediments and wetlands.

The K_d values currently used in for Mo-93 the biosphere model are relatively low. It is uncertain to what extent they are able to capture the immobilisation of Mo in sulphide-rich environments, although there is a clear trend of higher K_d values in more organic soils. It is also known that the sorption of Mo onto various surfaces is strongly dependent on pH. If pH in the future will be lower, this could be expected to decrease the mobility of Mo. One way to deal with these and similar problems would be to develop smart K_d values, which could account for pH, organic matter, sulphide etc. At the same time there is a need to keep the models simple but yet not stupid.

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Appendix 1

Final programme for the workshop

The seminar took place on 14 June 2016 at Såstaholm Castle, Stockholm. The final agenda is shown below.

#	Activity Detail	Speaker	Start Time	
1	Introduction	Ulrik Kautsky	09:00:00	09:10:00
3	Mo-93 inventory model	Merja Pukari and Klas Lundgren, Alara Engineering	09:10:00	09:30:00
4	Mo-93 in low and intermediate level waste	Björn Herschend Katrin Ahlford, SKB	09:30:00	09:50:00
5	Mo aspects in the near-field of low- and intermediate level waste repositories	Klas Källström and Ola Wessely, SKB	09:50:00	10:05:00
6	Coffee break		10:05:00	10:35:00
7	The geochemical journey of Mo from the NF to the rock	Mireia Grive, Amphos	10:35:00	10:55:00
9	Discussions		10:55:00	11:20:00
	Mo deposition	Per-Erik Karlsson , IVL	11:20:00	11:40:00
10	Lunch		11:40:00	12:40:00
11	Molybdenum chemistry in forest soils	Jon Petter Gustafsson, SLU & KTH	12:40:00	13:10:00
12	The geochemical behaviour of Mo in modern sediments	Tais Wittchen Dahl , Copenhagen Univ.	13:10:00	13:40:00
13	Discusssions		13:40:00	14:00:00
14	Mo in the site investigation areas	Fredrik Lidman, SLU	14:00:00	14:20:00
15	Mo-93 in biosphere models	Peter Saetre, SKB	14:20:00	14:30:00
16	Coffee break		14:30:00	15:00:00
17	Mo-93 in Finland	Thomas Hjerpe, Facilia AB	15:00:00	15:20:00
20	Final discussions		15:20:00	16:10:00

Appendix 2

List of participants

Participant	Organisation	E-mail Address (replace ABC with @)
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