

**R-10-27**

**Chemical conditions in present and  
future ecosystems in Forsmark –  
implications for selected radionuclides  
in the safety assessment SR-Site**

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December 2010

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**Update notice**

The original report, dated December 2010, was found to contain editorial errors which have been corrected in this updated version.

# Summary

This report is a background report for the biosphere analysis of the SR-Site Safety Assessment. This work aims to describe the future development of the chemical conditions at Forsmark, based on the present chemical conditions at landscape level taking landscape development and climate cases into consideration. The results presented contribute to the overall understanding of the present and future chemistry in the Forsmark area, and specifically, to the understanding of the behaviour of some selected radionuclides in the surface system. The future development of the chemistry at the site is qualitatively discussed with focus on the interglacial within the next 10,000 years. The effects on the chemical environment of future climate cases as Global Warming and cold permafrost climates are also briefly discussed.

The work is presented in two independent parts describing background radionuclide activities in the Forsmark area and the distribution and behaviour of a large number of stable elements in the landscape. In a concluding section, implications of the future chemical environment of a selection of radionuclides important in the Safety Assessment are discussed based on the knowledge of stable elements.

The broad range of elements studied show that there are general and expected patterns for the distribution and behaviour in the landscape of different groups of elements. Mass balances reveal major sources and sinks, pool estimations show where elements are accumulated in the landscape and estimations of time-scales give indications of the potential future development. This general knowledge is transferred to radionuclides not measured in order to estimate their behaviour and distribution in the landscape. It could be concluded that the future development of the chemical environment in the Forsmark area might affect element specific parameters used in the radionuclide model in different directions depending on element. The alternative climate cases, Global Warming and Permafrost, might have less influence on relative parameters such as  $K_d$  and CR if the underlying processes are unchanged compared to the temperate case.

## Sammanfattning

Den här är en bakgrundsrapport till biosfärsanalysen inom säkerhetsanalysen SR-Site. Arbetet syftar till att beskriva den framtida utvecklingen av de kemiska förhållandena i Forsmark baserat på nuvarande förhållanden, den framtida landskapsutvecklingen och alternativa klimatscenarier. Resultaten som presenteras bidrar till den allmänna förståelsen av de nuvarande och framtida kemiska förhållandena i Forsmark, och specifikt till förståelsen av hur några utvalda radionuklider beter sig i ytsystemet. Den framtida kemiska utvecklingen diskuteras kvalitativt med fokus på interglacialen under de kommande 10,000 åren. Effekten av alternativa klimatscenarier som global uppvärmning och kallt klimat med permafrost diskuteras också kortfattat.

Arbetet redovisas i två oberoende delar som dels beskriver bakgrundsaktiviteter i Forsmarksområdet för några radionuklider, dels fördelning och uppträdande av ett stort antal stabila element i landskapet. I ett sammanfattande avsnitt diskuteras därefter konsekvenserna av den framtida kemiska miljön för ett urval radionuklider som identifierats som viktiga i säkerhetsanalysen.

Den stora spännvidden av element som studerats visar att det finns generella och förväntade mönster i landskapet för olika grupper av element. Med massbalanser identifieras viktiga källor och sänkor, uppskattningar av de totala poolerna visar var element ackumuleras i landskapet och uppskattningar av tidsskalor ger indikationer om den potentiella framtida utvecklingen. De här generella kunskaperna överförs till radionuklider som inte mäts för att ge en bild av deras fördelning och uppträdande i landskapet. Det konstateras att utvecklingen av den framtida kemiska miljön i Forsmark skulle kunna påverka de elementspecifika parametrar som används i radionuklidmodellen i olika riktning beroende på element. De alternativa klimatscenerierna global uppvärmning samt kallt klimat med permafrost kommer möjligen ha mindre betydelse för relativa parametrar som  $K_d$  och CR om de underliggande processerna är relativt oförändrade jämfört med det tempererade klimatfallet.

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# 1 Introduction

Radioactive waste and spent nuclear fuel from Swedish nuclear power plants are managed by the Swedish Nuclear Fuel and Waste Management Co, SKB. Both waste and spent fuel are planned to be placed in a geological repository according to the KBS-3 method /SKB 2010a/. According to KBS-3, copper canisters with a cast iron insert containing spent fuel are to be enclosed by bentonite clay and deposited at approximately 500 m depth in saturated, granitic rock. Approximately 12,000 tonnes of spent nuclear fuel is forecasted to arise from the Swedish nuclear power programme, corresponding to roughly 6,000 canisters in a KBS-3 repository.

Between 2002 and 2008, SKB performed site investigations with the intention on finding a suitable location for a repository. Investigations were focused on two different sites along the eastern coast of southern Sweden; Forsmark in the municipality of Östhammar and Laxemar-Simpevarp in the municipality of Oskarshamn. Data from the site investigations have been used to produce comprehensive, multi-disciplinary site descriptions for each of the sites. The resulting site descriptions were reported in /SKB 2008/ (Forsmark) and /SKB 2009/ (Laxemar-Simpevarp). Based on available knowledge from the site descriptions and from preliminary safety assessments of the planned repository, SKB decided in June 2009 to select Forsmark as the site for the repository. This decision was based on a large number of empirical evidence suggesting Forsmark to be more suitable for a geological repository. The location of Forsmark is shown in Figure 1-1. An application for the construction of a geological repository for spent nuclear fuel at Forsmark is planned to be filed in 2011.

According to the regulations from the Swedish Radiation Safety Authority, SSM, a safety assessment of the planned repository has to be performed before the construction of the repository starts (SSMFS 2008:21). The assessment should focus on potential developments that may lead to the release of radionuclides. SKB launched the project SR-Site to conduct the safety assessment, summarised in the SR-Site main report /SKB 2011a/.



*Figure 1-1. Location of the Forsmark and Laxemar-Simpevarp sites.*

The safety assessment SR-Site focuses on three major fields of investigation: performance of the repository, the geosphere and the biosphere. The biosphere part of SR-Site, SR-Site Biosphere, provides estimates for human exposure after closure given a unit release, expressed as *Landscape Dose Conversion Factors*. Multiplying these factors with modelled release rates from the geosphere results in estimates of the annual doses used to assess compliance with the regulatory risk criterion. The effects on the environment of a potential release from the repository are also assessed in SR-Site Biosphere.

Within the SR-Site Biosphere project a number of subtasks could be distinguished:

- description of the site and the site development under different future conditions,
- description of properties and processes of importance for modelling radionuclide behaviour in present and future ecosystems,
- identification and description of areas in the landscape that potentially can be affected by release of radionuclides from the planned repository (Biosphere Objects),
- calculation of the radiological exposure to humans at constant unit release in future ecosystems in order to estimate Landscape Dose conversion Factors (LDF's), and also to calculation of radiological exposure to the environment,
- description and justification of the simplifications and assumptions made in the biosphere radionuclide model and in the underlying models.

The present report, which is further described Section 2, mainly deals with the first, second and last bullets in the list above.

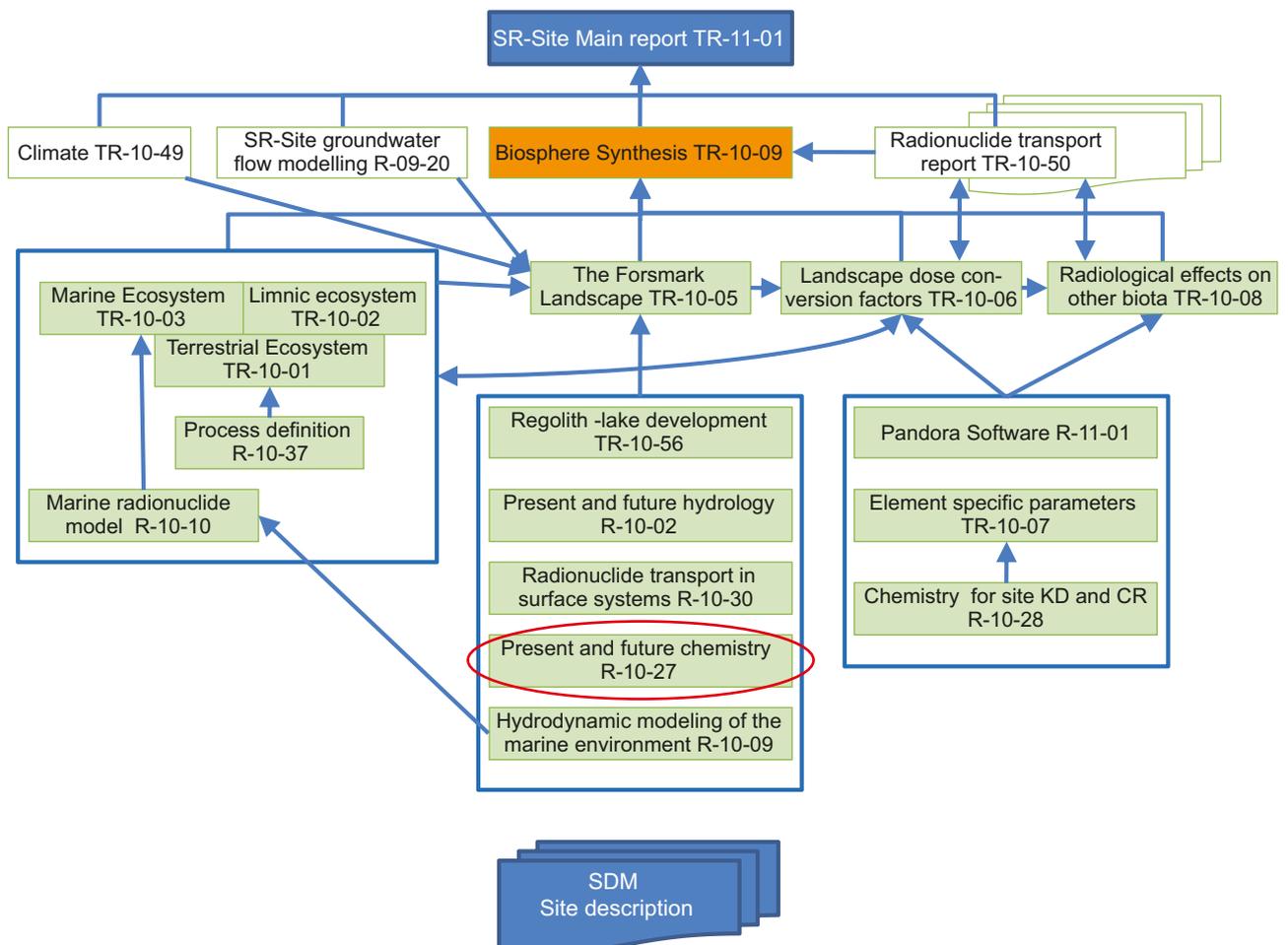
The SR-Site Biosphere project is part of a larger framework constituting the previous Site Descriptive Models (SDM – cf. Table 3-1), the present Safety Assessment /SKB 2011a/, and the Environmental Impact Assessment /SKB 2011b/. The SDM provides a description of the present conditions at the site, which is used as a basis for developing models describing the future conditions in the area within the SR-Site Safety Assessment. The general objectives of the SR-Site modelling and the specific objectives of the SR-Site Biosphere modelling are presented in /SKB 2010a/.

## 2 This report

This report is a background report for the biosphere analysis of the SR-Site Safety Assessment /SKB 2011a/, contributing to the general understanding of the present chemical conditions at Forsmark with references to the past and future development of the site. This report (R-10-27) provides information to the **Forsmark Landscape report** /Lindborg 2010/ and **Biosphere Synthesis report** /SKB 2010a/, according to the report structure in Figure 2-1.

### 2.1 Objective and scope

The current work aims to describe the future development of the chemical conditions at the site, based on the present chemical conditions at landscape level taking landscape development and climate cases into consideration. The results presented in this report contribute to the overall understanding of the chemistry in the Forsmark area, and specifically, to the understanding of the behaviour of important radionuclides in the surface system.



**Figure 2-1.** The hierarchy of reports produced in the SR-Site Biosphere project. Arrows indicates major interactions during project work flow of analysis and results, but interaction has been substantial between most parts of the project throughout the process. This report (R-10-27) is encircled in red mainly provides information to The Forsmark Landscape Report /Lindborg 2010/ and the Biosphere Synthesis Report /SKB 2010a/.

The future development of the chemistry at the site is qualitatively discussed with focus on the interglacial within the next 10,000 years. In a longer time perspective, i.e. 100,000–1,000,000 years, chemical conditions will alter radically depending on the phase of the glacial cycle, e.g. glaciations and land submerged by sea water /Söderbäck 2008/. As these stages are less important regarding dose to humans according to the dose modelling /Avila et al. 2010/, focus is laid on the chemical environment during the terrestrial period. The effects on the chemical environment of future climate cases as Global Warming and cold permafrost climates are also briefly discussed.

A large number of radionuclides are of interest when considering a potential release from a deep repository into the biosphere. In most cases it is not possible to study the actual radionuclides themselves, so the distribution patterns of naturally occurring radionuclides or their stable isotopes have been used to understand the behaviour of the radionuclides that may originate from nuclear waste, i.e. their analogues. By exploring the distribution and fluxes of a large number of stable elements within the landscape, the range in behaviour of different elements, or groups of elements, could be used in predicting the behaviour of specific radionuclides in the ecosystems. The rationale for including all available elements from the SKB sampling programme in the evaluations of this report was to achieve maximum variation, despite the sometimes weak connection to the radionuclides at interest.

Some specific objectives of the work described in this report are:

- to compile chemical data from different ecosystems in order to provide an understanding and quantification of major fluxes and pools of elements in the Forsmark area at landscape level,
- to estimate time spans for current processes in the Forsmark area,
- to qualitatively discuss the future development of the chemistry in the Forsmark area based on the present conditions and future cases,
- to discuss future implications for relevant radionuclides based on the knowledge of the future development of the chemistry of stable analogues.

## 2.2 Report structure

This report contains two introductory chapters describing the general objectives of the SR-Site Safety Assessment, the SR-Site biosphere project (Section 1) and the more specific objectives of this report in Section 2. The Forsmark site is introduced in Section 3 together with a short summary of previous work with focus on chemical conditions in the surface system.

The major work is presented in two independent parts describing background radionuclide activities in the Forsmark area in Section 4 and the distribution and behaviour of stable elements in the landscape in Section 5. In Section 6, which is based on previous sections, implications of the future chemical environment of a selection of radionuclides important in the Safety Assessment are discussed.

## 3 The Forsmark area

This section gives a short summary of previous work and serves as a background to this report with focus on chemical conditions in the surface system based on the site description (SDM), and SR-Site (cf. Section 1). The Forsmark area is briefly presented, followed by a short description of the present chemical conditions at the site. The past landscape development leading to the present chemical conditions is outlined together with a compilation of factors that probably will be important in the development of the future chemical environment.

### 3.1 Setting and general characteristics

The Forsmark area is located approximately 120 km north of Stockholm, in northern Uppland within the municipality of Östhammar (cf. Figure 1-1). Figure 3-1 shows the regional model area focused by the site investigation and within the site descriptive modelling.

/Lindborg 2008/ gives a thorough description of the surface system. Possible future cases and the succession of the landscape are described in /Lindborg 2010/. Different aspects of the Forsmark area have been described in a large number of reports, e.g. geology, hydrochemistry, hydrogeology, ecology etcetera. A selection of important background reports with focus on the surface system is compiled in Table 3-1, together with references to the comprehensive site descriptions.

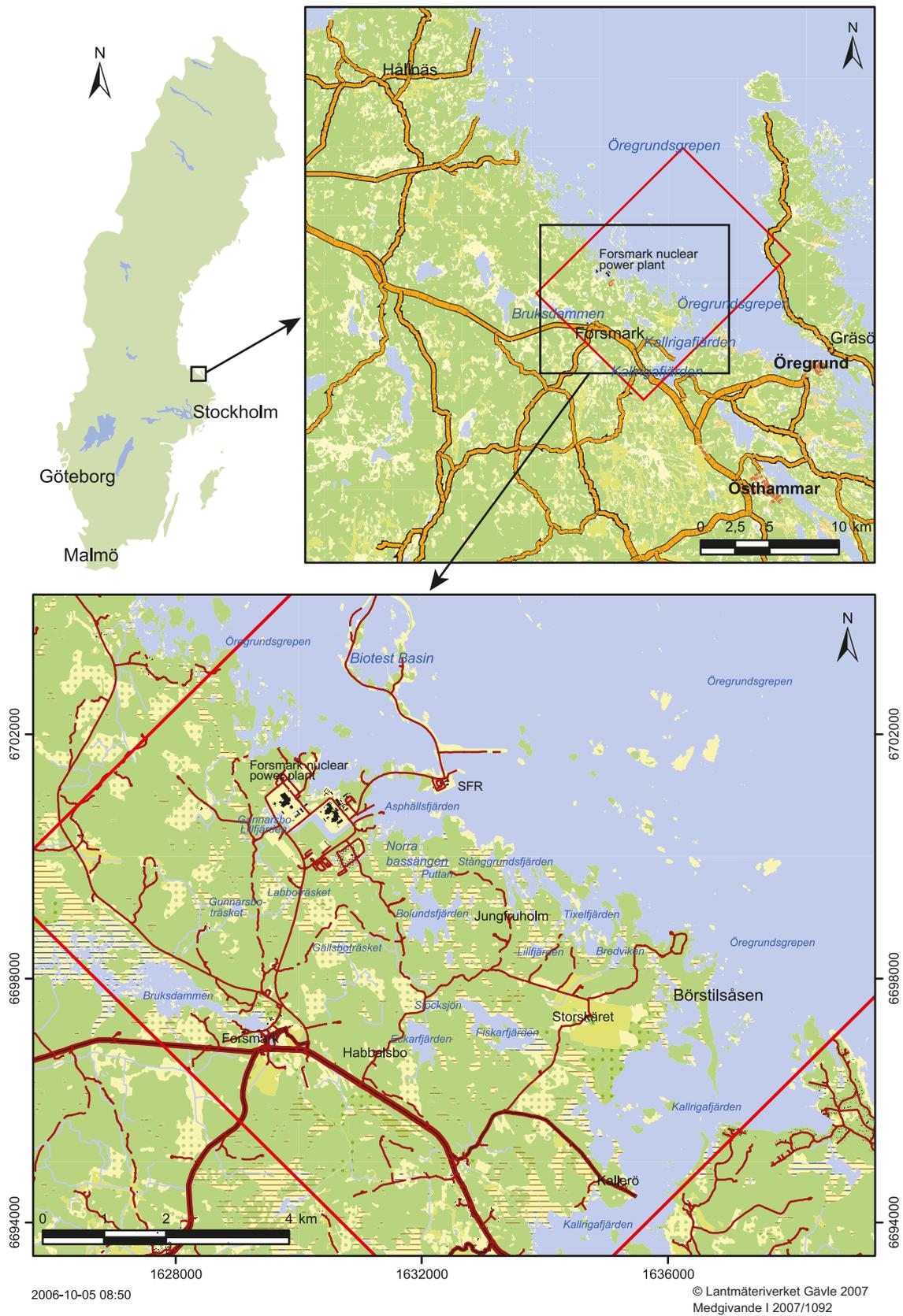
### 3.2 Present chemical conditions

Fresh surface waters and shallow groundwater in the Forsmark area are generally characterised by high contents of marine ions, high pH and high alkalinity, as well as very high concentrations of calcium compared with the general conditions in Sweden. These site-specific characteristics can be explained by marine remnants left by a regressing Baltic Sea, together with glacial remnants in the form of the calcite-rich till layer deposited during the latest glaciation /Sonesten 2005, Tröjbom and Söderbäck 2006a, Tröjbom et al. 2007/.

The strong influence from calcite has had a profound effect on the development of the terrestrial and limnic ecosystems in the Forsmark area. Secondary precipitation of calcium as calcite, including co-precipitation of dissolved phosphorus, mediates the development of the nutrient-poor oligotrophic hardwater lake type, typical of this region. In comparison with other parts of Sweden, lakes in the Forsmark area are characterised by low concentrations of phosphorus and high concentrations of nitrogen and dissolved organic carbon /Sonesten 2005/. The rich supply of calcium also influences soil formation and the development and structure of the terrestrial ecosystems /Löfgren 2010/.

### 3.3 Development since the last deglaciation

The chemical conditions observed in the Forsmark area today is a consequence of past landscape development, present and historic land use, and anthropogenic inputs. The ongoing shoreline regression creates a spatial gradient from the coast in an inland direction, which represents a timeline in landscape maturity. This means that the spatial gradient of today may be extrapolated and translated into a succession of landscape maturation for the future. The present chemical conditions found in the modelled area in Forsmark therefore represent a historic time span of about 10,000 years since the last deglaciation. In Figure 3-2 this is illustrated as a box moving over time from the sea-bottom environment to the terrestrial environment, although the illustration is somewhat limited by the fact that climate, vegetation cover and anthropogenic influence via e.g. atmospheric deposition and land use have varied during the period.

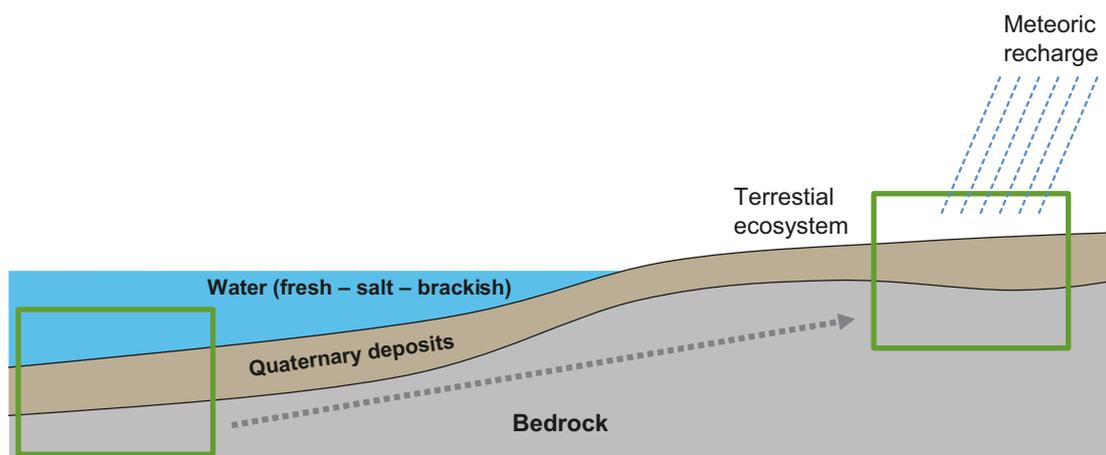


**Figure 3-1.** The location of the Forsmark site, where the detailed map includes the geographical names used throughout this report. The red rectangle shows the Forsmark regional model area focused by the site investigation and the site descriptive modelling.

**Table 3-1. Compilation of reports describing the surface system at Forsmark with focus on surface chemistry and issues discussed within this report (R-10-27).**

Report title	SKB Report	Reference
<b>Site descriptions</b>		
Surface system Forsmark. Site descriptive modelling SDM-Site Forsmark	R-08-11	/Lindborg 2008/
Biosphere analyses for the safety assessment SR-Site – synthesis and summary of results	TR-10-09	/SKB 2010a/
<b>Hydrochemical evaluations</b>		
Hydrochemistry in surface water and shallow groundwater. SDM-Site Forsmark	R-07-55	/Tröjbom et al. 2007/
Chemical characteristics of surface systems in the Forsmark area. SDM-Site Forsmark	R-06-19	/Tröjbom and Söderbäck 2006a/
Chemical characteristics of surface waters in the Forsmark area. SDM-Site Forsmark	R-05-41	/Sonesten 2005/
<b>Hydrology and hydrogeology</b>		
Description of surface hydrology and near-surface hydrogeology at Forsmark. SDM-Site Forsmark	R-08-08	/Johansson 2008/
Numerical modelling of surface hydrology and near-surface hydrogeology at Forsmark. SDM-Site Forsmark	R-08-09	/Bosson et al. 2008/
Modelling of present and future hydrology and solute transport at Forsmark. SR-Site Biosphere	R-10-02	/Bosson et al. 2010/
<b>Ecosystem descriptions</b>		
The terrestrial ecosystems at Forsmark and Laxemar-Simpevarp	R-08-01 TR-10-01*	/Löfgren 2008/ /Löfgren 2010/
The limnic ecosystems at Forsmark and Laxemar-Simpevarp	R-08-02 TR-10-02*	/Nordén et al. 2008/ /Andersson 2010/
The marine ecosystems at Forsmark and Laxemar-Simpevarp	R-08-03 TR-10-03*	/Wijnbladh et al. 2008/ /Aquilonius 2008/
<b>Geology</b>		
Description of the regolith at Forsmark. SDM-Site Forsmark	R-08-04	/Hedenström and Sohlenius 2008/
<b>Landscape development</b>		
Geological evolution, palaeoclimate and historical development of the Forsmark and Laxemar-Simpevarp areas. SDM-Site	R-08-19	/Söderbäck 2008/
Landscape Forsmark, Data, methodology and results for SR-Site	TR-10-05	/Lindborg 2010/

\* SDM reports updated within the SR-Site Biosphere project.



**Figure 3-2.** A hypothetical cross-section of the Forsmark area today, illustrating the spatial gradient that can be extrapolated and translated into a succession of landscape maturation for the future. The left box may represent the conditions soon after the deglaciation (8800 BC), whereas the right box represents conditions in the most elevated parts of the area which have been exposed to precipitation and chemical weathering for several thousands of years.

The historical development of the chemistry in the surface system at Forsmark can be divided into four stages based on differing hydrological conditions /Tröjbom et al. 2007/:

(1) Deglaciation of exposed glacial sediments, submerged by the freshwater Baltic Ice Lake. These deposits contained large amounts of limestone originating from the sea floor of Gävlebukten, 100 km north of the Forsmark area /Ingmar and Moreborg 1976, Hedenström and Sohlenius 2008/.

(2) Freshwater conditions were replaced by the brackish Littorina Sea, with increasing salinity, where density turnover infiltrated sea water through the underlying sediments /Laaksoharju et al. 2008/.

(3) Close to the emerging coast when a shallow sea still covered the area, the topographical discharge gradient may have transported traces of deep groundwater into glacial and post-glacial deposits. This flow pattern is still present close to the coast line /Bosson et al. 2010/.

(4) As soon as land emerged, precipitation and recharge of meteoric water created new hydrological flow patterns recharging through the regolith and discharging in streams and lakes. At this stage, aeration by meteoric recharge, in combination with a supply of organic carbon, altered redox conditions in the shallow groundwater. Increased supply of H<sup>+</sup> ions, mainly supplied through decomposition of organic matter, is the ultimate driving force for weathering reactions that take place in the regolith and bedrock /Tröjbom et al. 2007/.

### **3.4 Factors forming the future chemical environment**

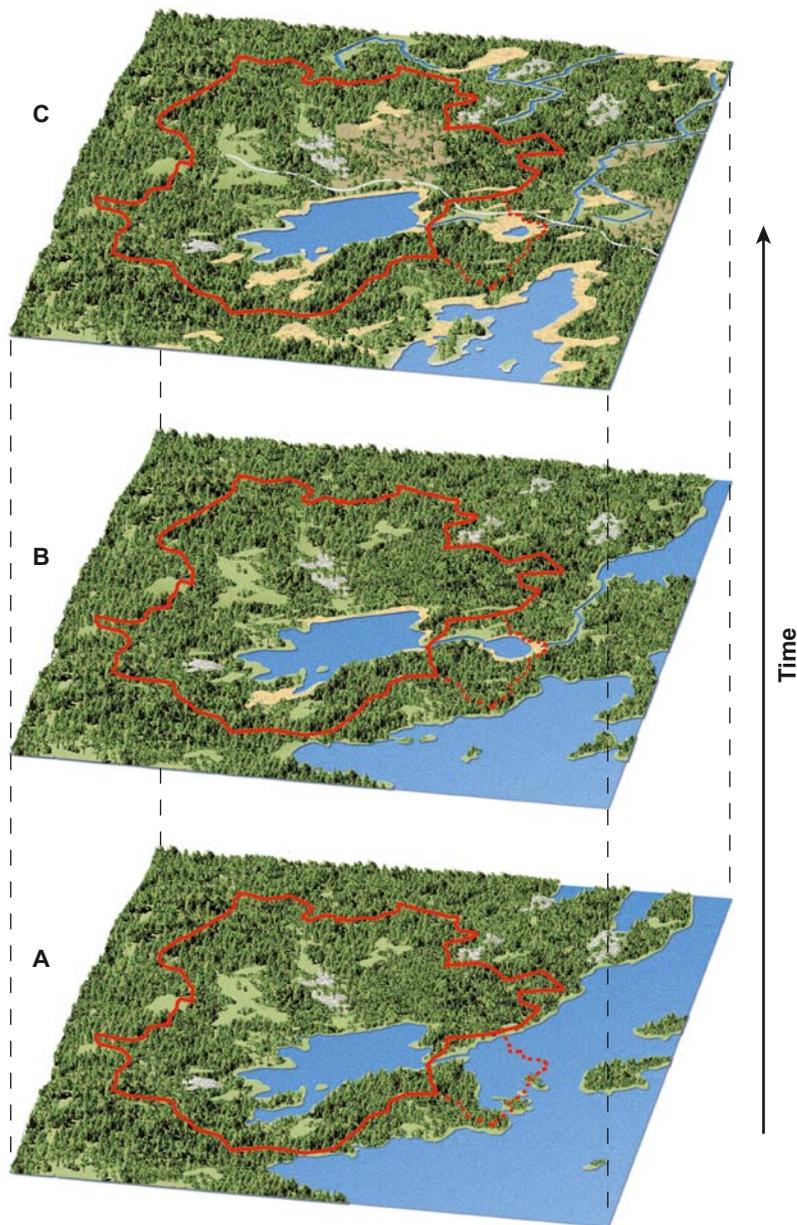
Several factors will influence the development of the chemical environment of the Forsmark landscape in the future. External abiotic factors as land uplift, climate and atmospheric deposition set the limits, whereas internal factors as primary production and land use may have profound influence via feedback mechanisms. In this section a number of factors potentially important for the forming of the future chemistry of the Forsmark area are identified.

#### **3.4.1 Land rise**

The ongoing land rise and the withdrawal of the Baltic Sea will continue and new land and new lakes will emerge. As this process proceeds, land use and vegetation cover will change, new lakes are transformed into mires and further into potential arable land as illustrated in Figure 3-3 (cf. /Andersson 2010 and Löfgren 2010/ for descriptions of the limnic and terrestrial ecosystems, respectively). The land rise process creates a gradient of young soils close to the coast to more mature soils in the inland. The emerging Forsmark landscape is not a system in steady-state. Instead, a number of processes at different time-scales interact and form variable conditions over time. Most measurements from the present Forsmark area represent a time span of only a few thousands years since the withdrawal of the sea. Data from the Forsmark region may extend this time span to at most approximately 10,000 years.

#### **3.4.2 Catchment morphology and lake ontogeny**

The chemical conditions measured in lakes and streams reflect the geometry, size and composition of the terrestrial parts of the catchments as well as the morphology and influence from lakes and wetlands within the hydrological network upstream. Most hydrochemical measurements from the Forsmark area represent small catchments with little influence from older, more mature parts of the landscape. Investigations of sediment stratigraphy in the inland catchment of Forsmarksån have shown that the oligotrophic hardwater stage, which characterises the present Forsmark area, does not necessarily have to arise. Instead could brown-water lakes be formed directly after isolation from the Baltic Sea as a result of influence from large catchments upstream /Brunberg and Blomqvist 2003/.



**Figure 3-3.** Potential development of a coastal area with two delineated sub-catchments (red solid and red dotted line, respectively) in east-central Sweden over approximately 800 years (A–C). Following the shoreline transgression, the smaller area (red dotted line) goes from being a shallow marine basin (A), to a lake-centred sub-catchment (B), and the subsequent expansion of reed belts in the littoral zone then transforms the lake into a wetland (C).

The typical ontogeny of the lakes in this area is a short, optional, oligotrophic hardwater stage lasting about 1,000 years followed by the development of brown-water lakes and finally wetland formation. The isolation of deeper depressions of the sea floor may result in the formation of eutrophic lakes, not present in the Forsmark region today. The deeper parts of Öregrundsgrepen will in the future possibly develop into a number of relatively large and deep eutrophic lakes with a hydrochemistry more resembling larger lakes further south along the coast. In the long term all lakes could be regarded as temporary since they will eventually be filled up and transformed to either wetland or drier land area (cf. /Lindborg 2010 and Andersson 2010/).

### 3.4.3 Climate and hydrology

Climate changes, either anthropogenic or natural, could change the prerequisites in either shorter or longer perspective by changing temperatures, precipitation patterns and water balance. Simulations show that there is a large range in possible climates for the Fennoscandian region in a 100,000 year time perspective /Kjellström et al. 2009/.

During a warm and wet climate (i.e. the Global Warming case), there will be only minor differences in the relative water balance compared to the present temperate climate, although the absolute runoff will increase significantly due to higher precipitation. The increased runoff under wet climate conditions affects the turnover and residence times of water in lakes and streams, which influence the transport of matter within and between the different ecosystems in the landscape. The effect of dilution of a hypothetical release from the repository on the surface might be higher under wet climate conditions due to the higher amount of water circulating in the surface system /Bosson et al. 2010/.

During permafrost conditions the water balance will be significantly altered. Since the evapotranspiration is very low the runoff leaving the system via the surface water streams is larger for the cold climate than under temperate climate conditions, even if the applied precipitation is lower. This is also a result of the fact that a large portion of the precipitation is snow, which gives a relatively short and intensive runoff during the snow melt when the evapotranspiration is still relatively small. The infiltration capacity under permafrost conditions is low resulting in a larger amount of direct runoff from the surface to the streams. The total amount of water transported to the streams via the saturated zone is however the same under temperate and cold conditions with permafrost /Bosson et al. 2010/.

When the land rises the groundwater table will be slightly lowered compared to the present conditions. This leads to decreasing amounts of water transported in the upper part of the profile, which has a high transport capacity. Thus, the fast transport of water to the surface stream network after a rain event might be reduced. A larger portion of the water will infiltrate to the deeper part of the regolith and the water will to a larger extent be transported in the saturated zone towards the streams, i.e. the contribution from the subsurface saturated zone to the runoff increases as the land rises, whereas the contribution from the overland part decreases /Bosson et al. 2010/.

Changing temperatures and hydrological patterns according to the cases described above may influence primary production and weathering rates, which in the prolongation could lead to more or less altered chemical conditions in the surface system.

### 3.4.4 Ecosystem development and primary production

The formation of a vegetation cover on the emerging land has had profound effects on the development of the chemical conditions of the surface system. Decomposition of organic matter generates  $H^+$  which is the driving force for weathering reactions in the deposits as well as a source of organic carbon and other nutrients in the surface water /Tröjbom et al. 2007/. In the young Forsmark area these processes are still not in steady state and equilibrium has not been established between accumulation and decomposition of organic matter. This will occur at 2,500 to 6,000 years from land rise according to /Löfgren 2010/.

As primary production in the terrestrial system is the ultimate driving force for weathering reactions in the regolith, a warmer and wetter climate leading to increasing primary production will also lead to increasing weathering rates in the regolith. Conversely, will a cold (and dry) climate with reduced primary production capacity probably lead to decreasing weathering rates in the surface system.

### 3.4.5 Atmospheric deposition

It is also plausible that atmospheric deposition patterns will change due to altered anthropogenic emissions in the (relatively near) future. Deposition of some elements will most probably increase in future whereas anthropogenic emissions and deposition of other elements, e.g. heavy metals as Pb, Cd, Hg, Cu and Zn perhaps will decrease. When the Baltic coast moves further east due to the land rise process, marine influences via atmospheric deposition will be reduced in the present Forsmark area.

### **3.4.6 Land use and human impact**

Over time, land use has had profound effects on the terrestrial and limnic ecosystems and on the chemistry observed in the surface system. Cultivation, breeding livestock, fertilization, forest management, draining of wetlands and lowering of lakes are all factors that potentially affect weathering rates, leakage of nutrients and conditions for retention in the limnic ecosystem. Depending on future land use, hydrochemistry could therefore be considerably influenced.

The part of the Forsmark area currently submerged by the sea contains larger areas of clay than the currently emerged part which is dominated by till /Hedenström and Sohlenius 2008/. This implies that weathering rates and area-specific discharge will increase for some elements when the sea bottom turns into land, especially if these areas are drained and cultivated similarly to comparable arable land in the region today. Area-specific losses of e.g. phosphorus are at least an order of magnitude larger for arable land than for forest land /Tröjbom et al. 2007/, and a greater proportion of arable land could significantly increase the nutrient status and primary production in the fresh surface water of streams and lakes (cf. Section 3.4.2).



According to /Avila et al. 2010/ the most important radionuclides, considering dose to humans, were:  $^{226}\text{Ra}$ ,  $^{129}\text{I}$ ,  $^{135}\text{Cs}$ ,  $^{36}\text{Cl}$ ,  $^{79}\text{Se}$ ,  $^{99}\text{Tc}$ , and  $^{237}\text{Np}$ . In Table 4-1, these nuclides are compiled together with a listing of which elements could be assumed as their stable analogues. The purpose of this listing is to explain the assumptions behind the discussion in Chapter 6. Considerations and argumentation for assigning analogues to individual radionuclides are presented in Appendix A – Description of the selected radionuclides.

**Table 4-1. Compilation of radionuclides included in the safety assessment and the stable elements assumed as analogues in this report.**

Element	Name	Isotopes	Stable analogues
Ag	Silver	$^{108m}\text{Ag}$	Ag
Ac	Actinium	$^{227}\text{Ac}$	(La)
Am	Americium	$^{241}\text{Am}$ , $^{243}\text{Am}$	(Eu)
C	Carbon	$^{14}\text{C}$	C
Ca	Calcium	$^{41}\text{Ca}$	Ca, (Sr)
Cd	Cadmium	Cd	Cd
Cl	Chlorine	$^{36}\text{Cl}$	Cl, (Br)
Cm	Curium	$^{244}\text{Cm}$ , $^{245}\text{Cm}$ , $^{246}\text{Cm}$	(Gd)
Cs	Cesium	$^{135}\text{Cs}$ , $^{137}\text{Cs}$	Rb, (K)
Eu	Europium	Eu	Eu
Ho	Holmium	$^{166}\text{Ho}$	Ho
I	Iodine	$^{129}\text{I}$	I
Mo	Molybdenum	$^{93}\text{Mo}$	Mo
Nb	Niobium	$^{94}\text{Nb}$	Nb, (Zr), (Ti)
Ni	Nickel	$^{59}\text{Ni}$ , $^{63}\text{Ni}$	Ni, (Co)
Np	Neptunium	$^{237}\text{Np}$	Pm
Pa	Protactinium	$^{231}\text{Pa}$	Pr
Pb	Lead	$^{210}\text{Pb}$	Pb
Pd	Palladium	$^{107}\text{Pd}$	(Ni)
Po	Polonium	$^{210}\text{Po}$	(Se), (Sb), (Pb)
Pu	Plutonium	$^{239}\text{Pu}$ , $^{240}\text{Pu}$ , $^{242}\text{Pu}$	(Sm)
Ra	Radium	$^{226}\text{Ra}$	Ba, (Sr), (Ca)
Se	Selenium	$^{79}\text{Se}$	Se, (S)
Sm	Samarium	$^{151}\text{Sm}$	Sm
Sn	Tin	$^{126}\text{Sn}$	Sn, (Pb)
Sr	Strontium	$^{90}\text{Sr}$	Sr, (Ca)
Tc	Tecnetium	$^{99}\text{Tc}$	(Cl)
Th	Thorium	$^{229}\text{Th}$ , $^{230}\text{Th}$ , $^{232}\text{Th}$	Th, (Pb)
U	Uranium	$^{233}\text{U}$ , $^{234}\text{U}$ , $^{235}\text{U}$ , $^{236}\text{U}$ , $^{238}\text{U}$	U
Zr	Zirconium	$^{93}\text{Zr}$	Zr, (Ti)

## 4.2 Site specific radionuclide activity data

In this section site specific radionuclide activity measurements from the Forsmark area are compiled<sup>1</sup>. These data were measured within the site investigation programme in water, biota, sediment and soil to serve as future references. Nine isotopes are naturally occurring isotopes (<sup>129</sup>I, <sup>226</sup>Ra, <sup>222</sup>Rn, <sup>228</sup>Th, <sup>230</sup>Th, <sup>232</sup>Th, <sup>234</sup>U, <sup>235</sup>U, <sup>238</sup>U), and eight are artificial isotopes emitted from nuclear power plants or nuclear bomb tests (<sup>238</sup>Pu, <sup>239</sup>Pu, <sup>240</sup>Pu, <sup>242</sup>Pu, <sup>99</sup>Tc, <sup>229</sup>Th, <sup>233</sup>U and <sup>236</sup>U). These in total 7 elements are marked in blue in the periodic system in Figure 4-1.

The number of samples analysed is presented in Table 4-2. Since the activities of some of the analysed isotopes were low, several samples showed results below the reported detection limits (shown within brackets in Table 4-2).

Isotopes of <sup>234</sup>U, <sup>235</sup>U, <sup>238</sup>U, <sup>230</sup>Th, <sup>232</sup>Th, <sup>226</sup>Ra and <sup>222</sup>Rn were analysed in deep groundwater, near surface water and surface water. As shown in Table 4-2 there are up to 108 samples in deep groundwater and 35 samples in near surface groundwater. In surface water there are up to 48 samples taken but many of these samples are below the reported detection limits.

The isotopes <sup>129</sup>I, <sup>238</sup>Pu, <sup>239</sup>Pu, <sup>240</sup>Pu, <sup>242</sup>Pu, <sup>226</sup>Ra, <sup>99</sup>Tc, <sup>228</sup>Th, <sup>229</sup>Th, <sup>230</sup>Th, <sup>232</sup>Th, <sup>233</sup>U, <sup>234</sup>U, <sup>235</sup>U, <sup>236</sup>U, <sup>238</sup>U were also analysed in biota, sediment and humus layers in two campaigns during the site investigation /Roos et al. 2007, Grolander and Roos 2009/. Soil samples were collected from different terrestrial habitats and sediment samples were taken from both limnic and marine environments. Biota samples represent terrestrial vegetation, terrestrial herbivores, aquatic vegetation and fish. There are between 1 and 7 samples available as shown in Table 4-2.

**Table 4-2. Total number of isotope analyses compiled in this report. Numbers within brackets represent the number of samples (of total) reported below detection limits.**

Isotope	Source of isotope	Number of water samples			Number of biota, soil and sediment samples					
		Deep ground-water	Near surface ground-water	Surface water	Terrestrial biota		Aquatic biota		Soil/humus layer	Sediment
					Vegetation	Herbivores	Vegetation	Fish		
<sup>129</sup> I	N, A				7	2	7	1	1	2
<sup>238</sup> Pu	A				2(2)	2(2)	5(5)	5(5)	1	2(1)
<sup>239</sup> Pu	A				7	2(2)	7	5	1	2
<sup>240</sup> Pu	A				2	2(2)	5	5	1	2
<sup>242</sup> Pu	A				7	2(2)	7	5	1(1)	2(2)
<sup>238</sup> Pu/ ( <sup>238</sup> Pu+ <sup>240</sup> Pu)					2		3		2	1
<sup>226</sup> Ra	N, A	91 (2)	10	29 (22)	2(2)	2	5(5)	5(5)	4	2
<sup>222</sup> Rn	N, A	87	10	13 (3)						
<sup>99</sup> Tc	A						1			1(1)
<sup>228</sup> Th	N, A	3		5	7		7	5	3	
<sup>229</sup> Th	A				5	2(2)	2		1(1)	2(2)
<sup>230</sup> Th	N, A	104 (11)	35 (16)	48 (24)		2	1		4	2
<sup>232</sup> Th	N, A	84 (13)	21 (11)	36 (20)	7	2	7	5	4	2
<sup>233</sup> U	A				2(1)	2(2)	5(3)	5(4)	1(1)	2(2)
<sup>234</sup> U	N, A	108 (3)	35 (2)	48 (13)	2(1)	2	5(3)	5(4)	4	2
<sup>235</sup> U	N, A	85(12)	21(12)	31 (19)	2(1)	2	5(3)	5(5)	1	2
<sup>236</sup> U	A				2(2)	2(2)	5(5)	5(5)	1(1)	2(2)
<sup>238</sup> U	N, A	108 (3)	35 (2)	48 (13)	2	2	3	1	4	2
<sup>234</sup> U/ <sup>238</sup> U		22	3	5	5		2		3	

N: natural occurring isotope, A: artificial isotope.

<sup>1</sup> Data extracted from the SICADA database 2009-11-10, access might be given on request.

#### 4.2.1 Background activities in water

Radionuclide activities were analysed within the site investigation, but the extent of these investigations are considerably smaller than the broad monitoring and investigations regarding stable elements in different media. Available measurements of radionuclide activities within the site investigation program in water, biota, sediment and soil are compiled in Table 4-2.

The results from the isotope analyses in deep groundwater, near surface groundwater and surface water are presented in Table 4-3. In this table, minimum, median and maximum values are listed for these naturally occurring isotopes and conclusions from this compilation of data are summarised below:

The activity concentrations of the uranium isotopes  $^{238}\text{U}$ ,  $^{235}\text{U}$  and  $^{234}\text{U}$  vary between the different water types, with the highest concentrations in deep groundwater, lower concentrations in near surface groundwater and the lowest concentration in surface water. The variation is also generally large within a water type. The median  $^{238}\text{U}$  activity in deep groundwater at Forsmark is 110 mBq/L which could be compared to the median activity of 32 mBq/L in measurements from 54 Swedish wells reported by /Östergren et al. 2003/. The  $^{234}\text{U}$  median activity in deep groundwater in Forsmark is 270 mBq/L which is significantly higher than the 66 mBq/L reported by /Östergren et al. 2003/. The activity is as expected highest for  $^{234}\text{U}$  and lowest for  $^{235}\text{U}$ .  $^{234}\text{U}$  has the highest activity because of its shorter half life even though the most common uranium isotope is  $^{238}\text{U}$  with a natural abundance of 99.284%. The *activity ratio*  $^{234}\text{U}/^{238}\text{U}$  has been calculated for the samples and the median ratio is 2.6 in deep groundwater, 1.2 for near surface groundwater and 1.1 for surface water. In Swedish groundwater the activity ratio  $^{234}\text{U}/^{238}\text{U}$  is in general 2.4 and the ratio is normally higher in groundwater than in surface water according to /Östergren et al. 2003/. This means that the activity ratio of the uranium isotopes in the Forsmark area is similar to the normal Swedish conditions. The *mass ratio* of  $^{234}\text{U}/^{238}\text{U}$  has also been measured in several samples and was found to be approximately 0.0072 which indicates a natural composition of uranium isotopes. It can therefore be concluded that even though the activities of uranium isotopes are higher in the Forsmark area than the average for Swedish groundwater, the uranium isotope composition appears to be of natural origin.

The activity is low for the thorium isotopes in all water types.  $^{232}\text{Th}$  show similar activity in the different water types, whereas  $^{230}\text{Th}$  show large variations in activity within the same water type but no clear differences between the different water types could be seen.  $^{228}\text{Th}$  was not measured in near surface water but the activity was significantly higher in deep groundwater than in surface water.

The  $^{226}\text{Ra}$  isotope shows high activities in all water types at Forsmark compared to normal conditions in Sweden. The activity is highest for deep groundwater (median 1,300 mBq/L), lower for shallow groundwater (median 60 mBq/L) and lowest for surface waters (median 6 mBq/L). According to /Aastrup 1981/ the Swedish median  $^{226}\text{Ra}$  activity in the groundwater of the regolith is 4.2 mBq/L, which is one order of magnitude lower than the median activity for shallow groundwater in the Forsmark area. The Swedish median  $^{226}\text{Ra}$  activity for deep groundwater in the bedrock is 3.7 mBq/L according to /Aastrup 1981/, which is as much as three orders of magnitude lower than the activity measured in the deep groundwater at Forsmark.

The  $^{222}\text{Rn}$  concentration follows the same pattern as the  $^{226}\text{Ra}$  concentration with high concentration in deep groundwater (270 Bq/L), lower in near surface groundwater (32 Bq/L) and lowest concentration in surface waters (0.034 Bq/L). The normal radon concentration in Swedish groundwater in soil is 10–300 Bq/L and in deep groundwater the normal concentration is 70–500 Bq/L, cf. /Grolander 2009, Skeppström 2005/. This means that the radon concentration in Forsmark is within the normal range for Swedish conditions.

It can be concluded that the activities of U, Ra and Rn isotopes are highest in deeper groundwater and decreasing towards the surface waters.

It was expected to find similar patterns for the Th isotopes as for U, Ra and Rn isotopes. Instead the thorium measurements show no significant difference between the different water types but a large variation within the water types. The reason for this is not clear.

For Rn there is a variation within the water types. This could be explained by local groundwater flow patterns. Discharging deep groundwater can be assumed to contain high levels of Rn and recharging groundwater can be assumed to contain no or very low levels of Rn. This is especially true for Rn that is an inert noble gas that does not react with the chemical environments along its flow path and therefore can be assumed to follow the groundwater flow path. The large variation in Rn activities between different sampling sites can perhaps be used to assess the origin of the groundwater, which is discussed further in /Grolander 2009/.

#### 4.2.2 Background activities in soil, sediments and biota

The results from the analyses in biota, soil and sediments are compiled in Table 4-4 where the minimum, median and maximum concentrations are presented. Conclusions from these results are summarised below:

There are few measurements of  $^{129}\text{I}$  and it is hard to draw any conclusions. The activities measured in the different biota types however seems to be within the same range. The highest activities were detected in humus layers.

$^{242}\text{Pu}$  were not found in any of the samples, while small amounts of the other Pu isotopes were found in most of the samples. Herbivores and fish constitute exceptions with no Pu isotopes detected, which indicates that Pu is accumulated in primary producers but not in consumers. The major part of the samples (where it could be detected) had a  $^{238}\text{Pu}/^{239}+^{240}\text{Pu}$  ratio corresponding to the 3–5% observed in nuclear bomb test fallout in the fifties and sixties in the northern hemisphere /Roos et al. 2007/.

$^{226}\text{Ra}$  activities were measured in terrestrial biota, aquatic biota, humus layers and sediments. In general it can be concluded that the activities are higher in the primary producers than in the consumers, which is valid both for terrestrial and aquatic biota. Median  $^{226}\text{Ra}$  activities are 580 mBq/kg for fish and 23,000 mBq/kg for aquatic primary producers. This means that the  $^{226}\text{Ra}$  activity is more than 50 times higher in aquatic primary producers than in fish. For terrestrial biota  $^{226}\text{Ra}$  activities are generally lower compared to aquatic biota with median activities in herbivores of 270 mBq/kg and 1,300 mBq/kg in primary producers. This means that there is a 5 times higher concentration of  $^{226}\text{Ra}$  in primary producers than in herbivores. This pattern suggests that primary producers accumulate more  $^{226}\text{Ra}$  than consumers and that the accumulation is higher in aquatic biota than in terrestrial. This pattern can to some extent be explained by the fact that only edible parts of the herbivores and fishes were analysed and since  $^{226}\text{Ra}$  is likely to accumulate in skeleton it is possible that the analysis of soft tissue will give a biased representation of the total amount of  $^{226}\text{Ra}$  in herbivores and fishes.

The  $^{226}\text{Ra}$  activities in the humus layer are very similar to the activities in the sediment. This means that  $^{226}\text{Ra}$  probably behave in a similar way in terrestrial and aquatic deposits.  $^{226}\text{Ra}$  concentrations in sediment from lakes and sea basins in the Forsmark area range from 8,500 to 30,000 mBq/kg with a median of 19,000 mBq/kg while the  $^{226}\text{Ra}$  concentration of sediment in the Baltic Sea ranges from 10,000 to 100,000 mBq/kg according to /HELCOM 2009/. This means that the  $^{226}\text{Ra}$  concentrations in the sediments in the Forsmark area are in the lower range of the activities found in the sediments of the Baltic Sea.

The naturally occurring Th isotopes were detected in varying activities in the analysed samples while the artificial isotope  $^{229}\text{Th}$  was not found in any of the samples of biota, humus or sediments. This indicates that no  $^{229}\text{Th}$  has been added to the natural system at the Forsmark site. The lowest activities were found for  $^{228}\text{Th}$ , with activities in terrestrial vegetation lower than in aquatic vegetation. For  $^{230}\text{Th}$  and  $^{232}\text{Th}$  large variations were found within the same group of samples and the reason for this large variation are unclear. It is hard to see any general patterns for the distribution of Th isotopes because of the large variations in activities. The activities of  $^{232}\text{Th}$  in sediments range from 5,800 mBq/kg to 22,000 mBq/kg with a median activity of 14,000 mBq/kg while the measured  $^{232}\text{Th}$  activities in the Baltic Sea varied between 10,000 mBq/kg to 50,000 mBq/kg according to /HELCOM 2009/.

No activities of the two artificial isotopes  $^{233}\text{U}$  and  $^{236}\text{U}$  were found in the analysed samples. For the natural uranium isotopes  $^{234}\text{U}$ ,  $^{235}\text{U}$  and  $^{238}\text{U}$  the activities varied much within the same sample group which makes it hard to draw any general conclusions on accumulation of these isotopes. It can be concluded that the lowest activities are found in herbivores and that the highest are found in humus layers and sediments.

The artificial isotope  $^{99}\text{Tc}$  was analysed in one sediment sample and one algae sample. The sediment sample fell below the reporting limit while  $^{99}\text{Tc}$  was detected in the algae sample.

### 4.3 Concluding remarks for the radionuclide section

The main purpose with the measurements of radionuclide activities compiled in this section was to describe background activities in the Forsmark area. The representativity of these measurements is usually not sufficient to draw conclusions of the distribution and behaviour of the radionuclides in the environment. Knowledge based on the distribution patterns of stable isotopes described in Chapter 5 are used in Chapter 6 to discuss possible implications of the radionuclides of the future development of the chemical environment in Forsmark.

The compilation of the radionuclides and their stable analogues in Table 4-1 should be regarded as a clarification of the assumptions behind the discussions of Chapter 6, and not as an attempt to review the many aspects of the analogue concept.

**Table 4-3. The results from the isotope analyses done in deep groundwater, near surface groundwater and surface water in the Forsmark investigation area. The results are presented as minimum, median and maximum activities. Cf. Table 4-2 for number of samples.**

Isotope	Groundwater mBq/L			Near Surface Groundwater mBq/L			Surface Water mBq/L		
	Min	Median	Max	Min	Median	Max	Min	Median	Max
$^{226}\text{Ra}$	<15	1,300	23,000	<15	60	300	<10	6	38
$^{222}\text{Rn}$	17,000	270,000	8,300,000	17,000	32,000	400,000	<15	34	250
$^{228}\text{Th}$	52	55	86				0.39	0.62	4.1
$^{230}\text{Th}$	<0.23	0.4	160	<0.3	1.6	25	<0.35	0.42	100
$^{232}\text{Th}$	<0.24	0.2	1.4	<50	0.86	5.6	<0.001	0.2	0.74
$^{234}\text{U}$	<50	270	4,000	<50	90	650	<50	18	100
$^{235}\text{U}$	<50	4.0	68	<0.02	2.0	7.8	<50	0.54	1.3
$^{238}\text{U}$	<50	110	1,900	<0.02	75	430	<50	15	100
$^{234}\text{U}/^{238}\text{U}$ (mass)	0.00710	0.00728	0.00743	0.00720	0.00726	0.00735	0.00720	0.00720	0.00720
$^{234}\text{U}/^{238}\text{U}$ (activity)	0.96	2.6	6.5	0.96	1.16	2.17	1.00	1.12	1.54

**Table 4-4. The results from the isotope analyses in soil, sediment, aquatic and terrestrial biota in the Forsmark investigation area. The results are presented as minimum, median and maximum activities. Cf. Table 4-2 for number of samples.**

Isotope	Terrestrial biota (mBq/Kg)						Aquatic biota (mBq/Kg)						Humus layer (mBq/Kg)			Sediment (mBq/Kg)		
	Vegetation			Herbivores			Vegetation			Fish			Min	Median	Max	Min	Median	Max
	Min	Median	Max	Min	Median	Max	Min	Median	Max	Min	Median	Max						
<sup>129</sup> I	0.16	3.4	6.6		0.24		0.45	1.4	2.1	0.28		0.28		4.4		0.33	0.41	0.49
<sup>238</sup> Pu	<4	20	43		<0.2		<60	4	240		<3		210		<20	26	73	
<sup>239</sup> Pu	<10	100	210		<1		<10	1,300	2,500	<10		23	1,500		100	610	1,100	
<sup>240</sup> Pu	<10	70	150		<1		<10	920	1,800	<10		10	1,300		48	370	680	
<sup>242</sup> Pu		<0.4			<0.1			<0.2			<0.5		<3.5			<1.6		
<sup>239</sup> Pu/ <sup>240</sup> Pu %	<4		360	<20		0.16	3.6	130	4,300	<4		37	2,800		150	20	1,800	
<sup>238</sup> Pu/ ( <sup>238</sup> Pu+ <sup>240</sup> Pu) %	9.8	9.9	10				2.7	5.5	6				7.5			4.1		
<sup>226</sup> Ra	300	1,300	7,600	70	270	470	3,800	23,000	42,000	320	580	20,000	13,000	17,000	29,000	8,500	19,000	30,000
<sup>99</sup> Tc								1,500									<600	
<sup>228</sup> Th	0.01	0.15	1.6				3.5	5.0	6.6				7.0	9.9	14			
<sup>229</sup> Th		<19			<3			<9			<20		<150		<69			
<sup>230</sup> Th	0.0000024	0.0000180	1,200	3.6	16	29	0.001	260	29,000	<38		240	0.01	0.02	8,900	5,600	17,000	29,000
<sup>232</sup> Th	3	16	980	2	6.5	11	17	480	17,000	1	6	120	6,800	9,600	12,000	5,800	14,000	22,000
<sup>233</sup> U		<5.2			<0.9			<2.3			<0.2		<41		<18			
<sup>234</sup> U	17	86	6,900	7	35	63	260	10,000	69,000	34	200	5,000	38,000	51,000	180,000	8,500	98,000	190,000
<sup>235</sup> U	6.2	1,300	2,700	0.3	1.7	3.1	11	430	2,800	1.2	7.3	200	1,800		380	4,000	7,700	
<sup>236</sup> U		<0.2			<1.2			<0.1			<0.2		<1.4		<0.60			
<sup>238</sup> U	16	85	58,000	7	30	54	220	9,000	59,000	23	150	4,300	37,000	46,000	160,000	8,000	86,000	160,000
<sup>234</sup> U/ <sup>238</sup> U %	0.0071	0.0073	0.0073				0.0072	0.0072	0.0072				0.0072					

## 5 Stable elements – transports and pools in the Forsmark landscape

In this chapter estimations of pools and fluxes of stable elements in the terrestrial ecosystem and the limnic ecosystem are coupled and evaluated at landscape level. The distribution of elements between different biotic and abiotic pools, together with estimations of element fluxes in and out of the pools give an overall picture of major sources and sinks in the landscape. General patterns among elements and element groups can reveal common processes in the landscape and can also indicate if elements show analogous behaviour in the ecosystems. The range in behaviour of different elements could also be used in predicting the behaviour of radionuclides in the ecosystems (cf. Chapter 6).

This section is divided into three major parts dealing with i) flux estimations by the use of a mass balance model ii) pool estimations based on detailed ecosystem models and iii) a combined evaluation of pools and fluxes which reveals estimations of time scales. Descriptions of methods, input data and validation data for the three parts are found in Section 5.1. The results regarding mass balances and fluxes are described in Section 5.2. In Section 5.3 pool estimations are described and in Section 5.4 estimations of time-scales.

### 5.1 Descriptions of methods and input data

In Section 5.1.1 the mass balance model used for flux estimations is described together with a compilation of primary input data and reference data used for validation. The basis for the estimations of pools in the landscape is described in Section 5.1.2. Denominations and abbreviations used throughout in this report are compiled in Table 5-1.

#### 5.1.1 Mass balance model for flux estimations

The mass balance approach used in this work is a simple budget calculation linking several sources of information at landscape level in order to quantify major fluxes in the landscape. The strength of this exercise is not a detailed modelling, but the broad range of elements included in the analysis. Besides the quantification of fluxes in the landscape, this modelling approach could also be seen as a step to validate and cross-reference site specific measurements against literature data and data from catchments nearby.

#### *Model structure and implementation*

The empirical mass balance model couples information from several sources and sinks at subcatchment level. The smallest spatial unit in the mass balance model is the subcatchment, which is further divided into the terrestrial and limnic system. Each ecosystem is further divided into three compartments representing the major pools regolith, biota and water according to the schematic description in the upper Figure 5-1. These subcatchments are interconnected via streams in the model according to the actual hydrological network as shown in the lower Figure 5-1.

A simplified mass balance for a subcatchment could be formulated according to the equation below based on the schematic description in Figure 5-1. The following major data sources were included in the mass balance: 1) element transports based on hydrochemical measurements in streams and discharge measurements in the Forsmark area. 2) Site specific deposition data, or if not available, literature data of disposition in the region and 3) accumulation in the limnic sediments based on measurements of element contents and modelled sedimentation rates.

$$\text{Deposition} + \text{Import} + T\_Acc/Wea = \text{Export} + L\_Acc$$

where

$$T\_Acc/Wea [g/yr] = ASD\_const [g/m^2/yr] \cdot \text{land area [m}^2]$$

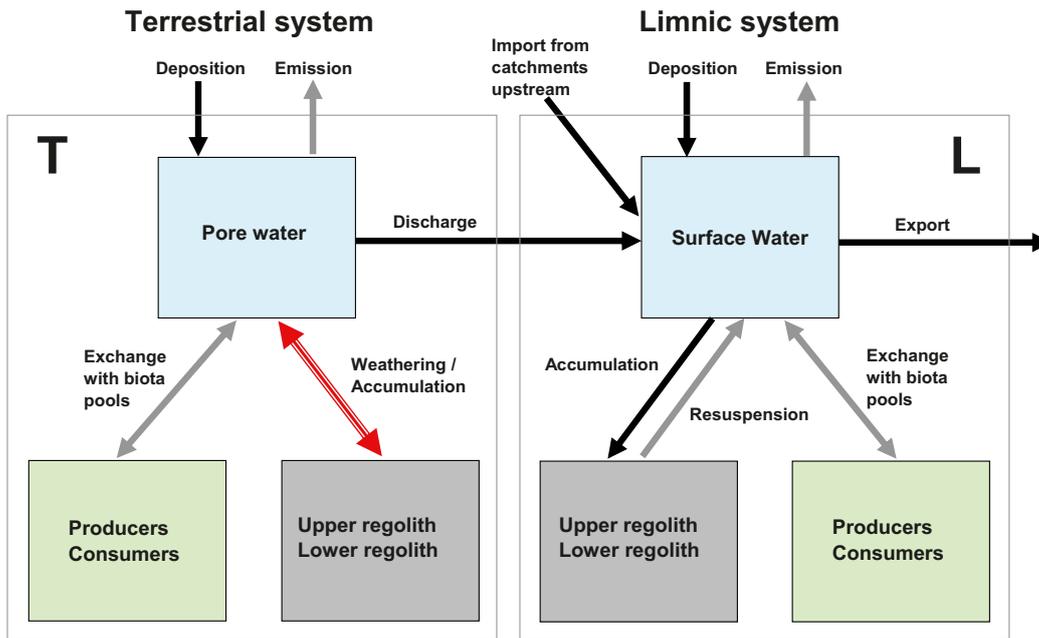
If  $ASD\_const > 0$ , there is net weathering in the terrestrial system

If  $ASD\_const < 0$ , there is net accumulation in the terrestrial system.

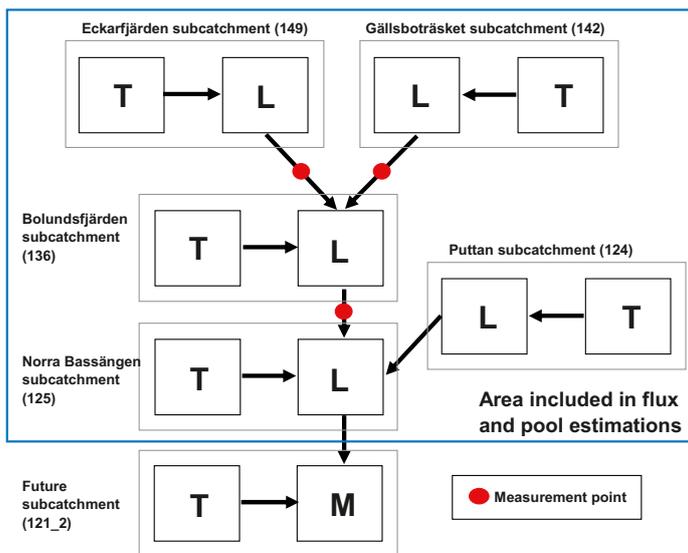
**Table 5-1. Compilation of denominations and abbreviations used in this report.**

<b>Fluxes</b>	<b>Abbreviation</b>	<b>Description</b>
Deposition	InAir	Atmospheric deposition
Deposition	T_InAir	Atmospheric deposition on land
Deposition	L_InAir	Atmospheric deposition on lake
Weathering/accumulation	T_Acc/Wea	Mass balance term representing either net weathering or net accumulation in the terrestrial system
Accumulation	T_Acc	Accumulation in the terrestrial system
Accumulation	L_Acc	Accumulation in lake sediments
Weathering	T_Wea	Net weathering in the terrestrial system
Discharge	T_OutWater	Discharge from terrestrial system to surface water
Discharge	T_OutRegoLow	Transport via groundwater discharge from RegoLow layer to surface water
Import	L_InWater	Sum of discharge and export from upstream catchments
Export	L_OutWater	Export via surface water
<b>Pools</b>	<b>Abbreviation</b>	<b>Description</b>
Biota	T_Bio	Total biota pool in the terrestrial system
Biota	L_Bio	Total biota pool in the limnic system
Producers	T_Prod	Producers in the terrestrial system
Producers	L_Prod	Producers in the limnic system
Consumers	T_Cons	Consumers in the terrestrial system
Consumers	L_Cons	Consumers in the limnic system
Regolith	T_Rego	Total regolith pool in the terrestrial system
Regolith	L_Rego	Total regolith pool in the limnic system
Upper regolith	T_RegoUp	Upper regolith layer in the terrestrial system
Upper regolith	L_RegoUp	Upper regolith layer in the limnic system
Lower regolith	T_RegoLow	Lower regolith layer in the terrestrial system
Lower regolith	L_RegoLow	Lower regolith layer in the limnic system
Water	L_Water	Total contents in water in the limnic system
Particulate fraction	L_Part	Particulate fraction of water in the limnic system
Dissolved fraction	L_Diss	Dissolved fraction of water in the limnic system
<b>Parameters</b>	<b>Abbreviation</b>	<b>Description</b>
Discharge constant	ASD_const	Element specific area specific discharge constant
Retention parameter	L_Ret	Landscape retention parameter
Landscape partitioning coefficient	LPC(L_Ret)	Landscape partitioning coefficient based on the landscape retention parameter
Landscape partitioning coefficient	LPC(pool)	Landscape partitioning coefficient based on pool and flux estimations

## Compartments of a subcatchment



## Connection of subcatchments



**Figure 5-1.** In the upper figure the terrestrial and limnic compartments of a subcatchment are shown. Coloured boxes represent pools in the terrestrial system (T) to the left and in the limnic system (L) to the right. Fluxes are shown as arrows where black denotes estimated fluxes, grey net fluxes assumed negligible in long term and the striped red arrow marks the mass balance term which either represents net weathering or net accumulation in the terrestrial system. The subcatchments (consisting of a terrestrial and limnic part) are interconnected according to the actual hydrological network as shown in the lower picture. The blue box marks the subcatchments included in flux and pool estimations. Red dots marks the calibration points, the basin numbers are shown within brackets and “M” denotes the marine basin downstream.

The area specific discharge constant (ASD\_const), which is common for all catchments for an element, was the only parameter varied during calibration of the model, and thus reflecting a common characteristic for the whole Forsmark area (cf. Table 5-1 for abbreviations). When solving the mass balance, weathering and accumulation in the terrestrial system was lumped together in a mass balance term (T\_Acc/Wea) representing the net effect of chemical and physical weathering processes (cf. /SKB 2010b/) as well as accumulation in soil and wetlands. In case of a positive mass balance term this is an estimation of the overall net weathering in the terrestrial system, with no regard to redistribution within the terrestrial ecosystem (wetlands are included in the terrestrial compartment, i.e. the redistribution from terrestrial areas to wetlands is not distinguished in the model). Conversely, represent a negative mass balance term the net accumulation in the terrestrial system, i.e. if for example a significant fraction of the main input via atmospheric deposition is retained in the terrestrial system. If the assumptions in the list above are less valid for an element and, if a neglected flux in fact is not negligible, it was by definition included in the mass balance term which should be held in mind when the results are evaluated.

The mass balance budgets, which couples total fluxes over a year, were based on several assumptions summarised below:

- Input via deposition was assumed equal on both lake and land surfaces. The bulk precipitation samplers used in the Forsmark area collect both wet and dry deposition.
- Emissions to the atmosphere were for all elements assumed negligible. This assumption is not valid for e.g. N where unknown fractions are brought from the atmospheric pool via nitrogen fixation and emitted to the atmosphere via denitrification. The net effects of these processes on the element were in practice included in the mass balance term described above. Also “emissions” of other elements not attributed to gas exchange could distort the mass balances via emissions of dust, pollen and migration of animals, as discussed in Section 5.5.
- The biota pools (producers and consumers) were assumed to be in steady state in relation to individual elements, i.e. no net accumulation occurs.
- There is no resuspension from the lake sediments, i.e. the accumulation term represents the long term burial in the sediments.
- All export from the landscape occurs via surface water transport of elements. It was consequently assumed that there is no transport via groundwater passing surface water divides (i.e. groundwater divides coincide with surface water divides) /Bosson et al. 2010/.

### ***Description of input data and reference data used for validation***

Estimations of annual export of elements via surface water discharge are based on concomitant measurements of concentrations and discharge in streams during June 2004 to May 2006 at three hydrochemical measurement points in the Forsmark area (cf. lower Figure 5-1). Input data and uncertainties are described in detail in Appendix B.

Data on atmospheric deposition originates from several sources. If available, site specific measurements of precipitation were used to estimate the total yearly deposition in the Forsmark area. Otherwise deposition data from the Laxemar-Simpevarp site were used instead, or if data from both these sites are missing literature data from Sweden were used. These data, which comprise direct input data in the model, are described in Appendix B.

The net accumulation per year in the lake sediments was estimated by multiplying the net accumulation rate with site specific element concentrations of the sediments as described in Appendix B.

In order to validate the results of the mass balances for the Forsmark area, area specific transports from catchments nearby as well as from other parts of Sweden were compiled for comparisons. Data from four sites were selected: Forsmarksån, a medium sized watercourse located close to the Forsmark site, the Laxemar-Simpevarp area which represents a landscape slightly larger than the Forsmark area in southern Sweden, the small catchment Storbergsbäcken located close to the Kalix River in northern Sweden and finally Svartberget/Krycklan/Svartberget, which is a small, thoroughly studied catchment located in northern Sweden (cf. Appendix B for further details).

In order to estimate the potential of discharging groundwater from deeper levels of the regolith to influence concentrations observed in the surface water, a hypothetical mass transport from the T\_RegoLow layer was estimated. By multiplying a typical concentration of the T\_RegoLow layer by the modelled average groundwater flux from this level, a hypothetical mass transport, potentially reaching the surface, was obtained. Considerations and assumptions behind the selection of representative data for this estimation are summarised in Appendix B.

### **Model calibration and validation**

For each element, a separate model was calibrated by varying a general ASD\_constant until maximal agreement was achieved between modelled and observed transports at three points marked as red dots in the lower Figure 5-1 (cf. /Tröjbom et al. 2007/ and /Johansson et al. 2008/ for detailed descriptions of the hydrochemistry and hydrology of the sampling sites PFM000069, PFM000070 and PFM000107).

There were not enough data to achieve an independent validation of the spatial variation modelled among the catchments, i.e. by data points not used in the calibration step. Average levels for the whole Forsmark area could, however, be validated if e.g. modelled area specific discharge is compared to estimations from catchments nearby as well as other parts of Sweden. These data are described under 'reference data' below.

### **Presenting of results**

All model results in Section 5.2 are summarised for the whole catchment of Norra Bassängen, marked by the blue box in the lower Figure 5-1. This means that the detailed spatial resolution used in the calibration procedure described below, mainly was an intermediate step in the modelling contributing to model stability and the overall confidence of the model results. As there is no import from upstream catchments for this area, the mass balance for the whole catchment of Norra Bassängen could be written according to the equation below. These are also the terms shown in e.g. Figure 5-4. Abbreviations are explained in Table 5-1.

$$\Sigma T\_InAir + \Sigma L\_InAir + \Sigma T\_Wea = Export + \Sigma L\_Acc + \Sigma T\_Acc$$

where

T\_Wea    Positive T\_Acc/Wea  
T\_Acc    Absolute value of negative T\_Acc/Wea

The overall retention in the landscape could be described by a parameter that relate the accumulation in the terrestrial and limnic ecosystems to the total input via atmospheric deposition and net weathering. This parameter, L\_Ret, is a relative measure describing the relationship between inputs and outputs of the system:

$$L\_Ret = \frac{Accumulation}{Deposition + Weathering} = \frac{\Sigma T\_Acc + \Sigma L\_Acc}{\Sigma T\_InAir + \Sigma L\_InAir + \Sigma T\_Wea}$$

If the system is at steady state, i.e. the inputs are balanced by the outputs, the L\_Ret parameter moves towards zero. Depending on the system borders and the time span, this parameter may approach a value other than zero which corresponds to the long term burial in relation to the input. L\_Ret values close to one may either indicate that the system is far from steady state, or that the element is very immobile. On the other hand, if the L\_Ret parameter is close to zero the system is probably close to steady state and the mobility in the landscape is probably high for this element.

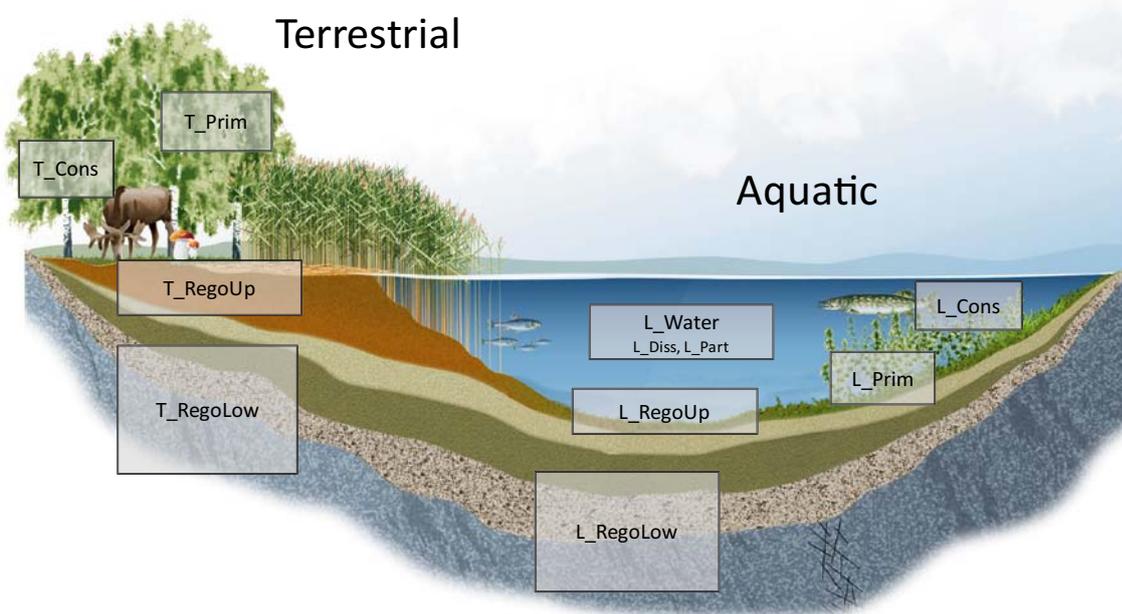
#### **5.1.2 Quantification major pools in the landscape**

The total element contents in the major pools were estimated for the terrestrial, limnic and marine ecosystems. These estimations were previously reported in /Löfgren 2008, 2010, Nordén et al. 2008, Andersson 2010, Wijnbladh et al. 2008, Aquilonius 2010/ and only a short summary of the methodology is given below. Pool estimations of the marine ecosystem is not included in this report and the Baltic coast is regarded the outer boundaries of the mass balance model previously described.

Each ecosystem was divided into a number of biotic and abiotic pools according to Figure 5-2. The biotic pool was divided into consumers and producers and the abiotic pool into the upper organic layers and the mineral layers below. In the limnic system, the amounts of elements in the lake water were divided into dissolved and particulate species. The estimation of the total element content in each pool was based on carbon budgets for each ecosystem and site specific element concentrations relative to carbon in the pools.

The terrestrial pools of consumers, producers and the upper regolith represent the total element content of all plants, animals and the organic soil layer based on information from each specific catchment. The upper regolith pool (T\_RegoUp) was defined as the organic soil layer (usually 0.5 m thickness), whereas the size of the lower regolith pool (T\_RegoLow) was defined as the upper 0.5 meter of the inorganic soil layer, i.e. the 0.5–1.0 m of the soil profile. The rationale for including only the upper 0.5 meters in the estimation of the T\_RegoLow pool in /Löfgren 2010/ was that this layer is in contact with the root zone and thus potentially participating in the biochemical cycles. In order not to introduce alternative estimations in the reporting they were left unchanged in this report, although this means that the T\_RegoLows pool are underestimated approximately 8 times as the average thickness of the regolith layer on land areas was estimated to 4 meters /Hedenström et al. 2008/. This discrepancy was however taken into account when the results were interpreted in Section 5.4.

In the limnic ecosystem, the size of the pools of producers, consumers, particulate matter and dissolved matter represent the total element content of all plants and animals as well as the particulate and dissolved fractions in the water of the lakes. The limnic L\_RegoUp pool represents the upper 1 cm of the sediment, whereas the L\_RegoLow layer represents the whole sediment profile except for this upper centimetre /Nordén et al. 2008/.



**Figure 5-2.** The terrestrial system is divided into four major compartments: the organic soil layer consisting of litter and humus (T\_RegoUp), the mineral soil layer (T\_RegoLow), producers (T\_Prod) and consumers (T\_Cons) /Löfgren 2010/. The limnic system is divided into six major compartments: The upper and lower regolith pools (L\_RegoUp and L\_RegoLow), the biota pool divided into producers and consumers (L\_Prod and L\_Cons) and the lake water pool divided into the dissolved and particulate fractions (L\_Diss and L\_Part) /Andersson 2010/.



Major sinks are accumulation in lakes and export via watercourses. The mass balance term, i.e. the difference between known sources and sinks in the mass balance (cf. Section 5.1.1), may either be positive or negative. A positive mass balance term means that the known sinks exceeds the known sources and in this case this term represents an additional supply from e.g. weathering reactions in the terrestrial system. A negative mass balance term means conversely that the known sources exceed the known sinks and in this case this term represent net accumulation in the terrestrial system (e.g. elements supplied via atmospheric deposition are accumulated in the terrestrial system). From Figure 5-4 it could be concluded that the relative distribution of sources and sinks differs significantly among elements and element groups.

For elements in the upper half of Figure 5-4, input from the atmosphere is equal to or exceeds the known sinks (export via surface water and accumulation in lake sediments). This implies that a significant fraction of these elements is retained in the terrestrial system. In case of e.g. Pb, Zn, Cd, Cu and Hg only c. 10% of the atmospheric deposition input reaches the Baltic Sea via discharge or is accumulated in the lake sediments. This general pattern also applies to metals as Ni, Nb, V, Ga, Cr, Be, W, Co, Th, Rb, Mn as well as metalloids and non metals as Se, Ga, P, Be, I, Sb, N and As. In the Svartberget/Krycklan catchment in northern Sweden as much as 90% of the Pb found in the soil originates from atmospheric deposition /Klaminder et al. 2006/. In the Storbergsbäcken catchment, /Land and Öhlander 2000/ found that the present day atmospheric input of Pb is considerably higher than the outputs via stream water.

For elements in the lower half of Figure 5-4 the sinks exceed the estimated input from the atmosphere which implies that there is a net contribution of these elements from a source within the terrestrial ecosystem. Weathering of minerals and ongoing flushing of marine remnants are possible terrestrial sources for these elements, but there is still a significant portion originating from atmospheric deposition. Metals such as Fe, Y, Sc, Zr, Mo, Sr, Ba belong to this group as well as several elements of marine origin, e.g. Cl, Na, Li, S, Br and K.

For six elements in the lower part of Figure 5-4, atmospheric deposition only balances a minor fraction of the known sinks. This means that weathering reactions and other processes in the terrestrial system are the main sources of these elements. Once released into the surface water, these elements could be accumulated in lake sediments or further exported to the Baltic Sea. Main constituents of minerals in rocks and regolith as Al, Si, Ca and Mg, as well as U belong to this group. In case of C, the mass balance term represents input from the atmosphere and weathering of calcites.

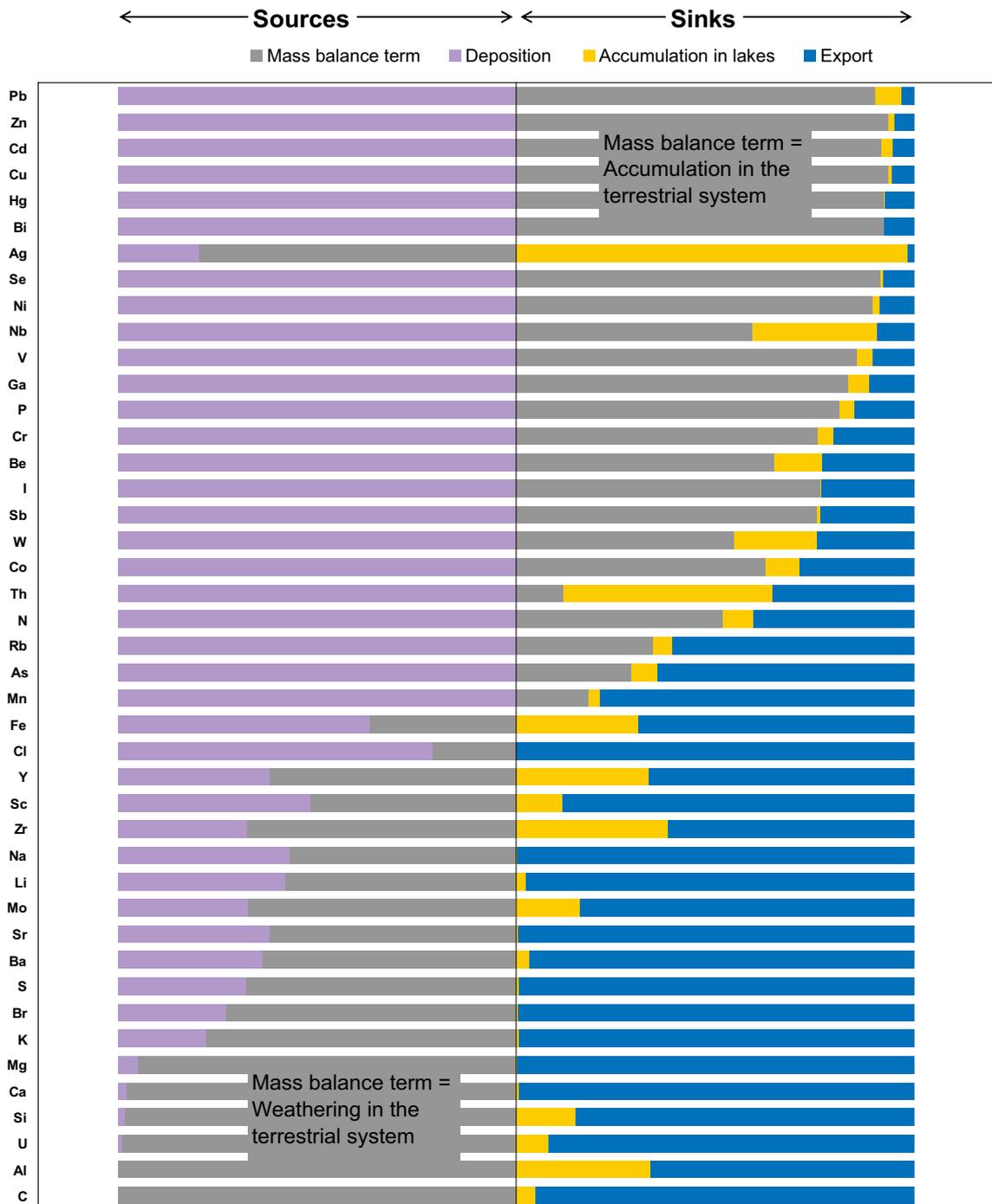
Besides these major groupings in Figure 5-4, several minor conclusions could be drawn from this figure:

Weathering of minerals is the main source for U in the Forsmark area. Concentrations of this element are significantly higher in the Forsmark area compared to other parts of Sweden /Tröjbom and Söderbäck 2006a/. An explanation could be locally elevated contents of U in regolith and rock, in combination with increased mobility of this element due to the exceptionally high alkalinity /Smellie et al. 2008/.

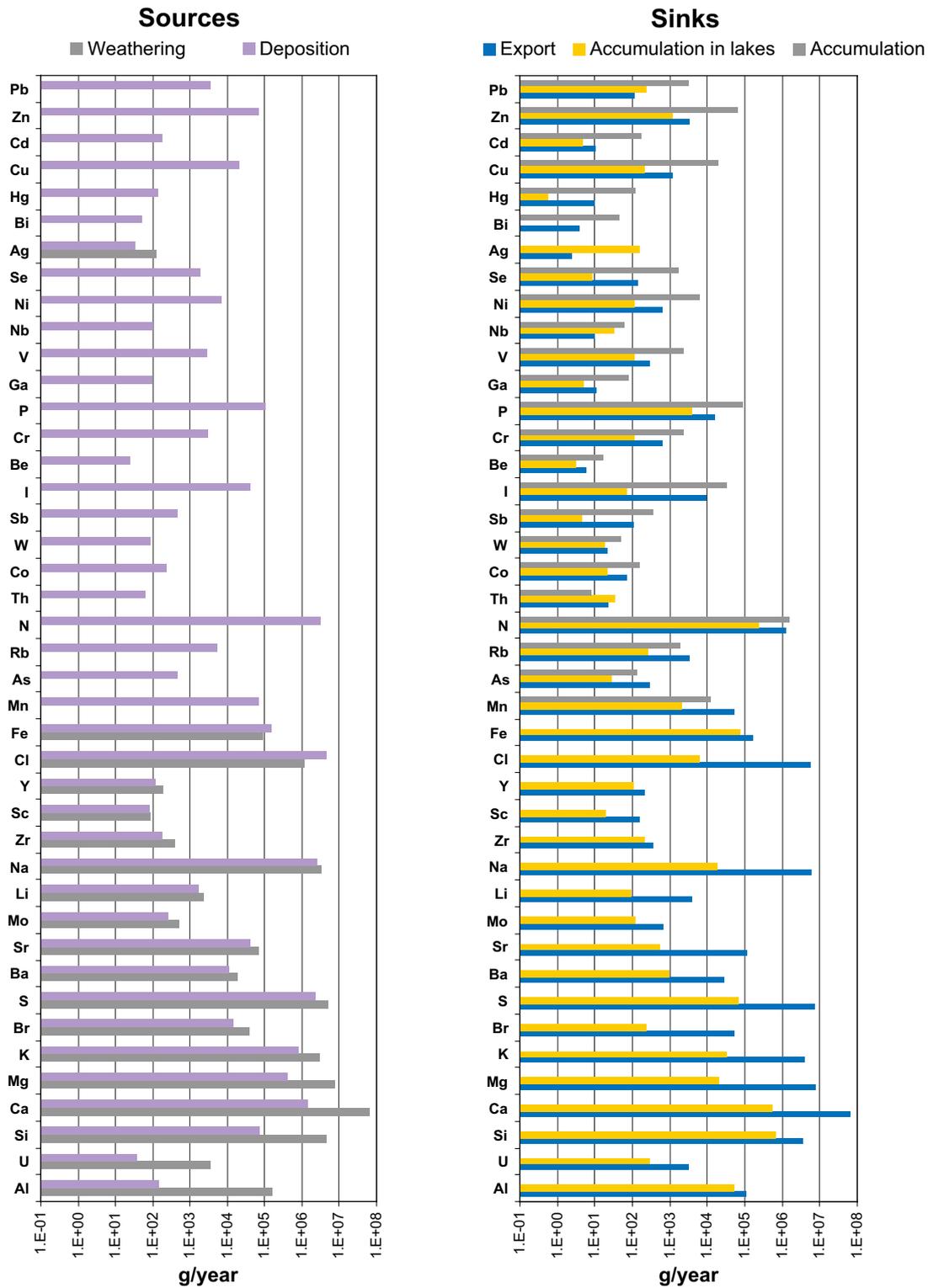
For three elements mainly associated with mineral sources, Zr, Sc and Y, the atmospheric input is in the same order of magnitude as the weathering (positive residual in the mass balance). This could be an artefact due to the lack of site specific deposition data and may be an indication of overestimated deposition for this element group. Zr is usually associated with hard-weathered minerals released through abrasive erosion (cf. rare earth metals and uncertainties discussed in Appendix B, Section B.2.8).

For elements abundant in sea water, e.g. Sr, Li, Na, Cl, atmospheric deposition of sea water aerosols is substantial in the coastal area of Forsmark. Weathering input (which also includes flushing of marine remnants besides weathering of minerals) is of the same magnitude as atmospheric deposition for these elements (cf. /Tröjbom et al. 2007/).

Three non-metals, N, P and I show a high degree of retention in the terrestrial system. In the case of N, a significant portion of the atmospheric input is likely emitted back to the atmosphere via denitrification processes, not quantified here. There is probably an even higher reflux to the atmosphere due to the input through nitrogen fixation, which is not included in the atmospheric input term. Phosphorus enters the system as atmospheric deposition in the form of particles or phosphate, or is released by weathering of phosphorus bearing minerals (cf. uncertainty discussion in Section 5.5).



**Figure 5-4.** Relative comparisons of element fluxes at landscape level in the Forsmark area. Exchange with the atmosphere is always treated as a source, e.g. atmospheric deposition (pink colour). Major sinks are accumulation in lakes (yellow) and export via watercourses (blue). The mass balance term (grey), corresponding to the difference between the above mentioned fluxes, either represents a source on the left side (weathering in the terrestrial system) or a sink on the right side (accumulation in the terrestrial system).



**Figure 5-5.** Absolute comparisons of element fluxes at landscape level in the Forsmark area expressed in g/year. Note the log-scales. In the left panel, the mass balance term (grey colour) represents weathering in the terrestrial system, whereas it represents accumulation in the terrestrial system in the right panel. The total catchment area is 8.5 km<sup>2</sup>, of which the lakes comprise 0.66 km<sup>2</sup>.

Phosphorus is effectively retained in the ecosystems by biotic uptake. The co precipitation of phosphorus in the calcium rich Forsmark area is also a possible factor that reduces the export via surface water. A large portion of the iodine input, mainly originating from marine sources, is also accumulated in the terrestrial system according to the mass balance.

For many metals, the sink representing accumulation in lake sediments is usually small compared to the accumulation in the terrestrial system. In such case, accumulation in lake sediments is of minor importance compared to the accumulation in the terrestrial system at landscape level. The absolute relationships between the terrestrial and limnic sinks are mainly determined by the relative size of the systems where the terrestrial system is significantly larger. It is, however, relevant to make relative comparisons among elements to reveal if some elements are to a greater extent retained in any of the systems. Nb and Th are for example two exceptions from this pattern where a greater relative fraction is retained in the limnic system than in the terrestrial system in comparison to most other metals.

Also a great portion of Fe, Y, Zr and Al accumulates in the limnic system compared to the export via discharge. Some of these elements are associated with minerals with low solubility (e.g. Zr bearing minerals), which means that they probably are to a higher degree transported as particles. Also Ti (not available for the mass balance) and lanthanides should belong to this group of elements.

In the bullet list below, some uncertainties and anomalies in Figure 5-4 are discussed for selected elements. General uncertainties are discussed in Section 5.5.

- Br – There is no reliable deposition data available for Br. The airborne deposition estimation is based on the assumption that Br mainly originates from marine sources with a constant Br/Cl ratio and therefore is proportionalised to the site specific Cl deposition (cf. Appendix B, Section B.1.2). The discrepancy between Br and Cl in the mass balances, where a greater portion of Br seem to be released from the terrestrial system, could perhaps reflect the lower mobility of Br and a delayed flushing of relict marine remnants compared to Cl.
- Ag – The distribution pattern of this element differs from other heavy metals (e.g. Cu, Zn, Pb and Cd) and it could not be excluded that this pattern is an artefact. There is no site specific deposition data for this element and a literature value from southern Sweden is used instead. The accumulation in the lake sediments is large compared to the export via discharge and a possible explanation to this pattern could be precipitation of e.g. AgCl in the limnic system. If the accumulation in the sediments is overestimated the positive residual is also overestimated which in turn implicates that the weathering in the terrestrial system is exaggerated. Other investigations show Ag concentrations in lake sediments which are c. 20 times lower than the data used in the mass balance, whereas other heavy metals do not show this discrepancy between the investigations /Engdahl et al. 2008/. The interpretation is not straightforward, but if these latter data are more representative for the Ag contents of limnic sediments, the overall pattern becomes more similar to other heavy metals, although with a greater fraction retained in the limnic system.
- According to the mass balances of Nb and Th, a relatively large portion of these elements are retained in the limnic sediments compared to other metals. There is also room for retention in the terrestrial system if the input via atmospheric deposition is apportioned among the known sinks. Nb is usually associated to Fe, which is retained in the limnic system as hydroxy-oxides /Åström et al. 2008/.
- Fe and Mn are redox sensitive elements with usually similar behaviour in the environment. According to the mass balances, the overall patterns for these elements differ in that respect that the atmospheric input is larger for Mn, whereas the long term accumulation in lakes is more accentuated for Fe. The relatively higher atmospheric input for Mn could perhaps either be explained by the relatively greater importance of the sea water input (cf. limnic pools in Figure 5-12) or the composition of regolith and bedrock. The discrepancy between Mn and Fe in the limnic system is most likely due to the occasionally reducing environment in bottom water of the lakes leading to dissolution of precipitates of Mn prior to Fe. The incorporation of Mn in biota in the terrestrial system is also relatively more important for Mn compared to Fe according to Figure 5-11.

It should be emphasized that the relative distribution patterns in Figure 5-4 are strongly dependent on the uncertainties associated with the flux estimations. The major conclusion from this mass balance exercise is that the overall patterns expected and reflect the range in behaviour in the landscape for different elements. The overall trend lining up the elements from Pb at the top to Al at the bottom reflects the contrasting properties of these elements; Pb is an air borne pollutant bound to particles and organic matter in the terrestrial and limnic ecosystems /Steinnes and Friedland 2006/, whereas Al has local mineral origin through weathering where a significant portion accumulates in lakes in the form of mineral particles. The exact location of a single element on this scale is however more uncertain due to uncertainties in background data (cf. Section 5.5). This is clearly illustrated by the REE group and related elements which are handled in Appendix B, Section B.2.8.

## 5.2.2 Validation by area specific transports in other catchments

To give an indication if the fluxes used in the mass balances are reasonable, area specific mass transports from the Forsmark area were compared with transports from four other sites in Sweden: 1) The catchment of Forsmarksån located close to the Forsmark site, 2) Storbergsbäcken and 3) Svartberget/Svartberget/Krycklan in northern Sweden, and 4) the Laxemar-Simpevarp area in southern Sweden. These in all five sites all represent relatively small catchments close to the Baltic coast, but with differing conditions regarding geological properties, atmospheric deposition and climate (cf. Section 5.1.1).

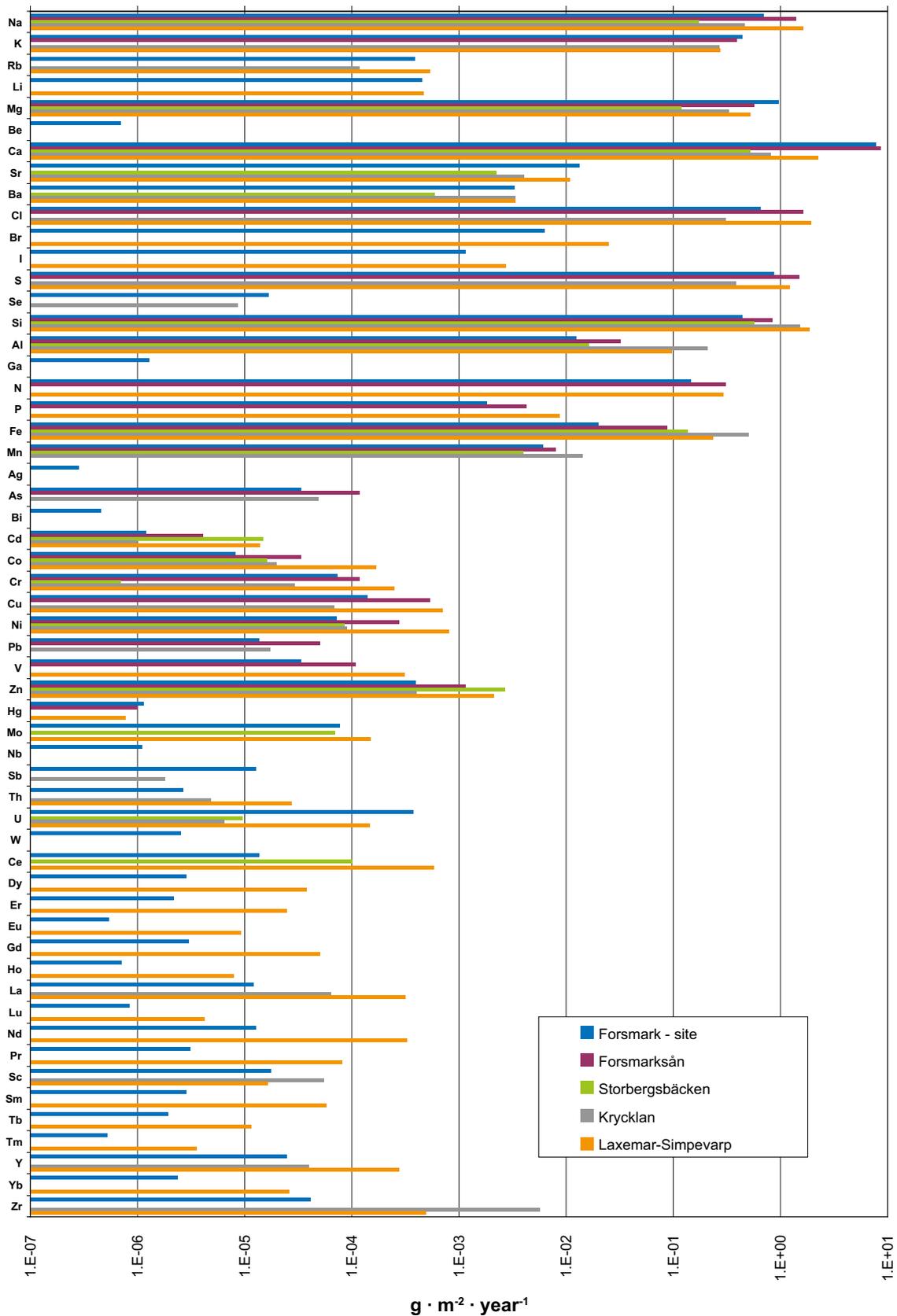
When area specific transports from the Forsmark site are compared with transports observed in the Forsmarksån catchment nearby, the general picture is that they agree well for most elements. The detailed comparison in Table 5-2 between the Forsmark site and the catchment of Forsmarksån shows that the area specific transports is comparable for most major elements during 2004–2006. In Figure 5-6 and Figure 5-7, area specific transports from the Forsmark area, Forsmarksån, Storbergsbäcken, Svartberget/Krycklan and Laxemar-Simpevarp area could be compared for several elements (all data are tabulated in Appendix B, Table B-2). Conclusions are summarised below:

Ca is the main constituent of the dissolved ions with an area specific transport almost 10 times higher than the second most abundant element. The area specific discharge of Ca is approximately 10 g/m<sup>2</sup>/year in both the Norra Bassängen catchment in the Forsmark area (7.8) and the catchments of Forsmarksån nearby (7.2) during 2004–2006. It should be noted that the bars in Figure 5-7 represent different time periods. From the statistics in Table 5-2 it could be concluded that there is probably no clear difference between the larger catchment of Forsmarksån and the Forsmark site regarding the area specific transport of Ca. This also implicates that the Ca weathering gradient from coast to inland is not particularly strong. The weathering of the calcite rich till starts when meteoric water percolates the regolith and is thought to decrease over time when the calcite is consumed (cf. Section 5.4). Compared to the other reference sites, the Forsmark region (the Forsmark area and the catchment of Forsmarksån) marks out by almost an order of magnitude higher area specific transport of Ca.

**Table 5-2. Area specific transports in Forsmarksån and the Norra Bassängen catchment (g/m<sup>2</sup>/year). Statistics of area specific transports per year are based on monthly measurements. The figures marked in bold represent the same period.**

		Ca	Mg	Na	K	S	Cl	Tot-N	Tot-P	Si	TOC
<b>Forsmarksån catchment (large inland catchment)</b>											
1978–2008	25-percentile	6.7	0.4	1.1	0.28	0.8	1.3	0.20	0.003	0.41	3.5
	Median	8.5	0.5	1.2	0.35	1.0	1.4	0.24	0.005	0.60	4.3
	75-percentile	9.9	0.6	1.5	0.41	1.3	1.8	0.30	0.006	0.72	4.8
2004–2006	Mean	<b>7.2</b>	<b>0.5</b>	<b>1.2</b>	<b>0.32</b>	<b>0.5</b>	<b>1.4</b>	<b>0.23</b>	<b>0.004</b>	<b>0.40</b>	<b>4.2</b>
2007–2008	Mean	8.8	0.6	1.4	0.39	0.5	1.6	0.31	0.004	0.85	5.8
<b>Norra Bassängen catchment (Forsmark area)</b>											
2004–2006		<b>7.8</b>	<b>1.0</b>	<b>0.7</b>	<b>0.44</b>	<b>0.87</b>	<b>0.65</b>	<b>0.15</b>	<b>0.002</b>	<b>0.44</b>	<b>2.8</b>





**Figure 5-7.** Comparison of area specific transports between the Forsmark site and four reference sites ( $g/m^2/yr$ ). The inland catchment of Forsmarksån close to the Forsmark site, Storbergsbäcken in Northern Sweden close to Kalix River, Svartberget/Krycklan in northern Sweden close to Umeå and the Laxemar-Simpevarp area in southern Sweden.

The area specific transport of U is considerable higher in the Forsmark area compared to the catchments of Storbergsbäcken and Svartberget/Krycklan (U is not measured in Forsmarksån). Discharge of U is almost two orders of magnitude higher in the catchment of Norra Bassängen, which may be an indication of elevated concentrations in the regolith or bedrock and/or that U is highly mobile at the hydrochemical conditions prevailing in the Forsmark area. Compared to the Laxemar-Simpevarp area the discrepancy is less pronounced, cf. /Smellie et al. 2008/.

The area specific transports of REE are at least one order of magnitude higher in the Laxemar-Simpevarp area compared to the Forsmark area. There are no data on REE from the catchment of Forsmarksån. A single observation from Storbergsbäcken point in the same direction as Laxemar-Simpevarp data, i.e. higher area specific transports than Forsmark, whereas data from Svartberget/Krycklan are intermediate. According to Appendix B, Section B.2.8, flux estimations are probably uncertain for these elements and the observed discrepancy could be analysis artefacts. This discrepancy is probably attributed to local contents of these elements in the bedrock and the regolith, cf. /Tröjbom and Nordén 2010/.

Area specific transports of the nutrients N and P are slightly higher in the catchment of Forsmarksån compared to the Norra Bassängen catchment in the Forsmark area. A larger proportion of arable land in the former catchment is a possible explanation to the slightly elevated area specific transports of nutrients. Area specific transports of N and P in the Laxemar-Simpevarp area are also of comparable size.

The overall conclusion from this comparison is that the area specific transports estimated for the Forsmark area are in level with the transports observed in the catchment of Forsmarksån nearby. Compared to other sites in Sweden, some of the discrepancies could be explained by the specific hydrochemical conditions prevailing in the Forsmark area, e.g. the calcite rich till and the resulting high alkalinity. The location of the sites close to the Baltic Sea explains the generally elevated transports of marine ions at these sites compared to typical conditions in Sweden /Sonesten 2005/. These ions either originate from flushing of relict remnants in the deposits or ongoing atmospheric deposition (sea spray).

### 5.2.3 Potential contributions from groundwater discharge

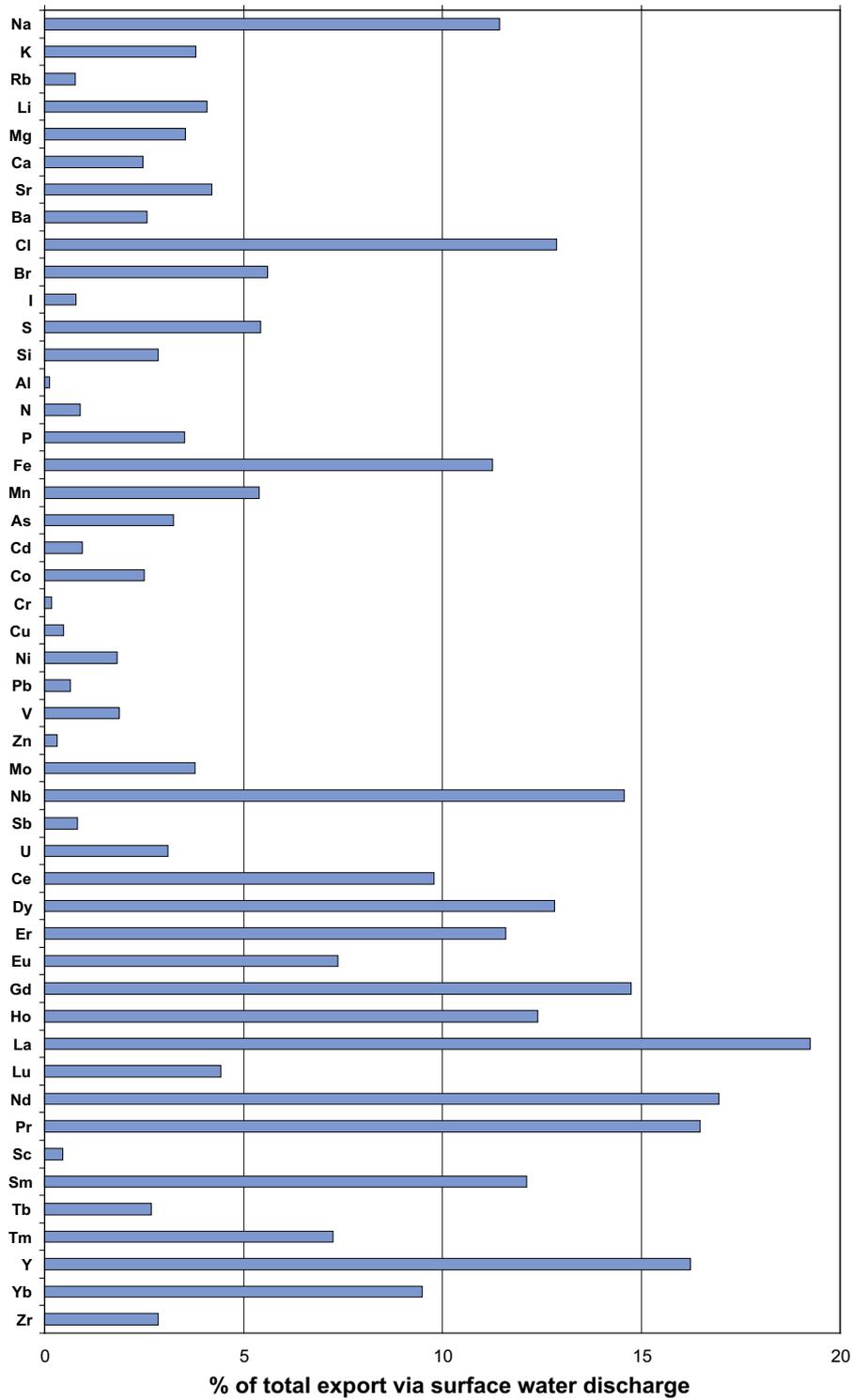
The maximum *potential* contribution of elements via groundwater discharge from deeper levels of the regolith to surface water discharge was estimated by multiplying typical element concentrations in the T\_RegoLow layer with the yearly groundwater flux from the T\_RegoLow layer to the surface (cf. Appendix B, Section B.1.5). This estimation is independent from the mass balance described in Section 5.2.1 and may support interpretations about the origin of elements in the surface system. It should be noted that this exercise is rough and the results could only give an indication if deep groundwater discharge is a possible source of significance.

According to this rough estimation only a fraction (maximum 20%) of the element transports at ground level can be explained by groundwater discharge from the lower regolith layer. The major patterns and conclusions are summarised below:

Flushing of marine remnants from the regolith layer is a potential source of marine ions in the surface system as shown in Figure 5-8 (e.g. Na and Cl). According to this estimation approximately 10–15% of the Cl in the surface water discharge could on average in the Forsmark area originate from the regolith layer. Locally this fraction could be significantly higher if the groundwater is brackish (the typical groundwater selected in this exercise represents a non-brackish, altered groundwater type influenced by cation exchange processes and meteoric recharge (cf. Appendix B, Section B.1.5).

Due to the low redox potential in the saturated zone, dissolved Fe and Mn occur at elevated concentrations in groundwater at depth. Discharge of groundwater of low redox potential may therefore add these elements to the surface system. When this water is oxygenated at the surface these metals precipitate in mires and lakes or are transported as colloids or larger particles to the Baltic Sea.

For many of the transition metals (e.g. Cu, Zn, Ni, Co) discharge of groundwater could theoretically contribute only to a minor fraction of the amounts observed in the surface system. As shown by the mass balances, the main source of these metals is atmospheric deposition (cf. Section 5.2.1).



*Figure 5-8. Hypotetical maximal contribution from the RegoLow layer to the surface water discharge in streams in the Forsmark area.*

Si, Al and Zr are elements associated to weathering of minerals (cf. Section 5.2.1). The release of these elements into the water phase depends on weathering reactions which in turn are driven by H<sup>+</sup> that mainly originates from decomposition of organic matter at the surface. Abrasive erosion may also put these elements into the water phase as particles or colloids. According to this estimation, the deeper levels of the regolith are probably not an important source for the release of these elements at the surface.

The hypothetical contribution of rare earth elements, REE, to the surface system from the T\_RegoLow layer is not exceeding 20%. As stated before, uncertainties associated to this element group are large and this pattern may be an artefact. This rough estimate however indicates that discharge from deep levels of the regolith is not a main source for these elements in the surface system, but compared to most other elements potentially more important.

Nutrients as N and P are usually related to decomposition of organic matter and deeper parts of the regolith are therefore not a probable source for these elements, as indicated by this estimation (especially not for N).

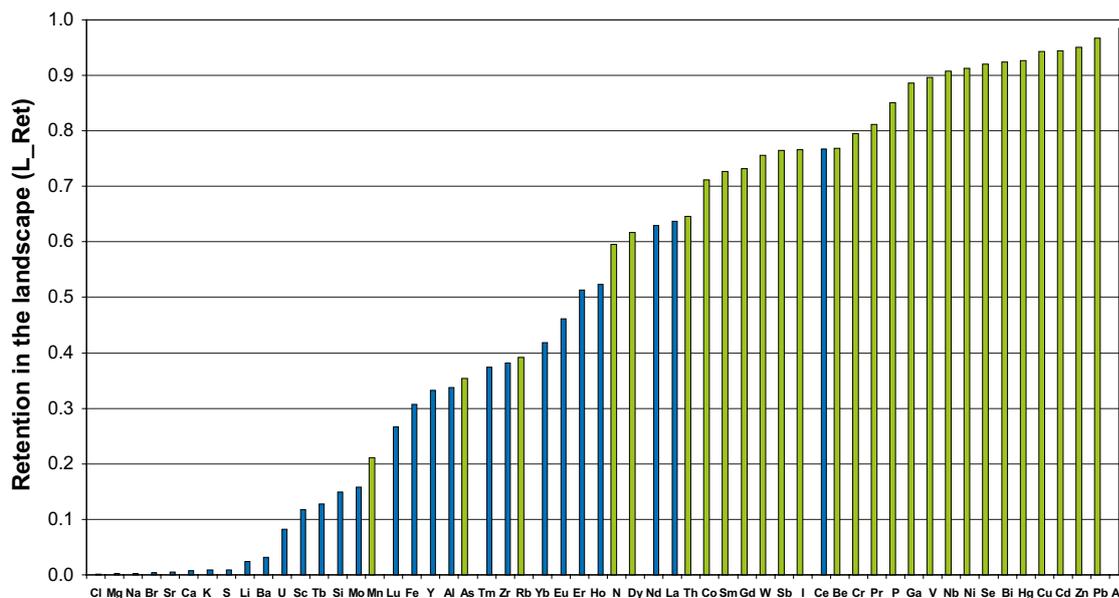
Nb, which is one of the selected nuclides that may be released from the deep repository (cf. Chapter 4), marks out in Figure 5-8 compared to many other metals by showing a slightly higher potential for groundwater influence at the surface. According to the mass balance in Section 5.2.1 the main source for Nb in the surface system seems to be atmospheric deposition in combination with high retention in the limnic system.

#### 5.2.4 Overall retention in the landscape

The overall retention in the landscape could be described by a parameter that relates the accumulation in the terrestrial and limnic ecosystems to the total input via atmospheric deposition and net weathering. This parameter, L\_Ret, is a relative measure describing the relationship between inputs and outputs of the system (cf. Section 5.1.1). If the system approaches steady state the L\_Ret parameter moves towards zero. Depending on the system definitions and the time span considered, this parameter may approach a value other than zero which corresponds to the long term burial in relation to the input. L\_Ret values close to one might either indicate that the system is far from steady state, or that the element is very immobile. On the other hand, if the L\_Ret parameter is close to zero the system is probably close to steady state and the mobility in the landscape is probably high for this element.

$$L\_Ret = \frac{Accumulation}{Deposition + Weathering}$$

The L\_Ret parameter has been calculated from the mass balances in Section 5.2.1 for a large range of elements based on average conditions in the Forsmark area reflecting the present situation. From Figure 5-9 could be concluded that several elements in the left part show negligible retention (e.g. Cl, Mg, Na, Br, Sr and Ca), which indicates that these elements are highly mobile and approximately at steady state. Several heavy metals, e.g. Pb, Zn, Cd and Cu show on the contrary very high retention in the landscape which either reflects a condition very far from steady state, or low mobility in the landscape as a whole. In case of e.g. Pb both explanations are valid: the low mobility of this metal in combination with increased anthropogenic emissions during the last centuries probably forms an advancing Pb front in the soil profile that will reach the discharging surface water in the future. At that time, the L\_Ret parameter will show a lower value corresponding to the long term burial in the landscape, cf. /Klaminder et al. 2006/ and /Steinnes and Friedland 2006/.



**Figure 5-9.** Overall retention in the landscape. The colour of the bar indicates if the element is released (blue) or accumulated (green) in the terrestrial system.

### 5.3 Evaluation of pools of elements in the landscape

Element pools have previously been estimated for the terrestrial and the limnic ecosystems in the Norra Bassängen catchment based on carbon budgets and element concentrations in a large number of organisms, soil, sediment and water. In this section the results from these studies are reviewed in order to facilitate comparisons between ecosystems at landscape level (cf. Section 5.1.2). For detailed descriptions of these estimations the reader is referred to /Löfgren 2010, Andersson 2010/. Uncertainties regarding the pool estimates are discussed in Sections B.2.5 and B.2.6. These data are further evaluated in Section 5.4 where they are coupled to fluxes at landscape level to give a conception of time-scales and residence times of the pools.

It should be emphasized that these pool estimations are dependent on the underlying assumptions. This is especially valid for the terrestrial T\_RegoLow pool, for which the vertical extent was set to 0.5 m in the estimation in /Löfgren 2010/, although the average thickness of the regolith layer on land areas is estimated to 4 meters /Hedenström et al. 2008/. The rationale for including only the upper 0.5 meters in the estimation of the T\_RegoLow pool in /Löfgren 2010/ was that this layer is in contact with the root zone and thus potentially participating in the biochemical cycles. In order not to introduce alternative estimations in the reporting these figures were left unchanged in this report, although this means that the T\_RegoLow pool is underestimated approximately 8 times (cf. Section 5.1.2). This implicates that the dominance of the T\_RegoLow pool is even larger which is accounted for in the estimations in Section 5.4.

The pool estimations are summarised over the same area as the transport estimations in Section 5.2 and the general patterns are discussed in brief below. Relative comparisons among the pools are visualised in Figure 5-11 to Figure 5-13 and absolute numbers are found in Figure B-4 to Figure B-6 in Appendix B, Section B.4. In this section, the presentation of the pool data is divided into four figures, each combining the pools in different ways to facilitate comparisons:

- In Figure 5-10 the pools of the terrestrial and limnic ecosystems are compared. The producer and consumer pools are combined in a general biota pool per ecosystem. In the left panel, the RegoUp and RegoLow pools are summed per ecosystem (cf. Section 5.1.2 for definitions). From this figure, it is clear that the total terrestrial and limnic regolith pools are dominating and of comparable size within the catchment of Norra Bassängen (note that this relation is dependent on the assumption of the vertical extent of the T\_RegoLow layer cf. Section 5.1.2). In the right panel, the RegoLow layers have been excluded and the pools represent the amounts available in the short term.

- In Figure 5-11, the relative sizes of the pools are shown for the terrestrial ecosystem. In the left panel all pools are included, whereas the T\_RegoLow pool has been excluded in the right panel to make details visible. The selected pools in the right panel thus represents the amounts potentially available in short term.
- In Figure 5-12, the relative size of the pools is shown for the limnic ecosystem. In the left panel all pools are included, whereas the L\_RegoLow pool has been excluded in the right panel to make details visible. The selected pools in the right panel thus represents the amounts potentially available in short term.
- In Figure 5-13, limnic producers and consumers are compared. In the terrestrial ecosystem the consumer pool is negligible for all elements compared to the producer pool and therefore not shown in a separate figure.

For most nutrients in the terrestrial system the producer, consumer and T\_RegoUp pools are not negligible compared to the inorganic T\_RegoLow pools (e.g. N, P, K, S, Ca, Zn, Cu and Cl). This is especially evident for Cl where a large fraction of the total amount of this element is found within the producer pool (mainly as tree biomass). Cl, which is an essential nutrient, actively taken up by plants, is also enriched in the producer pool compared to the related, but not essential, element Br. The amounts of Cl and Br in the T\_RegoLow pools are both low due to the high mobility and leaching of these elements. According to the right panel of Figure 5-11, where the T\_RegoLow pools have been excluded, all essential nutrients mark out by a higher relative fraction in the producer pool compared to geogenic elements, i.e. the patterns reflect the active uptake/accumulation of these elements in biota.

Some elements, not essential for biota, are also enriched in the terrestrial producer and RegoUp pools compared to most other elements, e.g. Hg, and Cd. These elements show distribution patterns similar to essential trace elements such as Cu and Zn.

Some elements seem to accumulate in the terrestrial T\_RegoUp layer but not to any greater extent in the biota pools, e.g. Pb, Sb, As and Ag. These elements, which are not actively taken up by the plants, are probably associated to particulate and/or organic matter in the T\_RegoUp layer. This is well known for Pb and results in limited mobility and accumulation for this metal in the terrestrial ecosystem (cf. mass balance in Section 5.2.1). The metalloid As on the other hand, is according to the mass balance more mobile compared to Pb and a greater fraction of the atmospheric input seems to reach the surface water system.

U is also enriched in the terrestrial T\_RegoUp layer compared to geogenic elements as Si and Al, but is not to any greater extent taken up by the biota. The mobility of U seems to be high in the Forsmark area, probably due to the unique hydrochemical environment, and much of the U released by weathering reactions in the terrestrial system reaches the Baltic Sea according to the mass balance in Section 5.2.1.

When the relative distributions of the pools of the limnic system are compared in Figure 5-12, the L\_RegoLow layer totally dominates the picture except for Cl, Br and to some extent Na. Dissolved in the lake water, these mobile marine ions comprise a significant fraction of the total amounts in the limnic system, particularly Cl and to some lesser extent Br. Na additionally originates from non-marine sources, e.g. weathering and erosion of regolith and bedrock, further retained in the limnic sediments as mineral particles. If the dominating L\_RegoLow pools are excluded from Figure 5-12, the relative distribution of the pools in the right panel gives a more detailed picture.

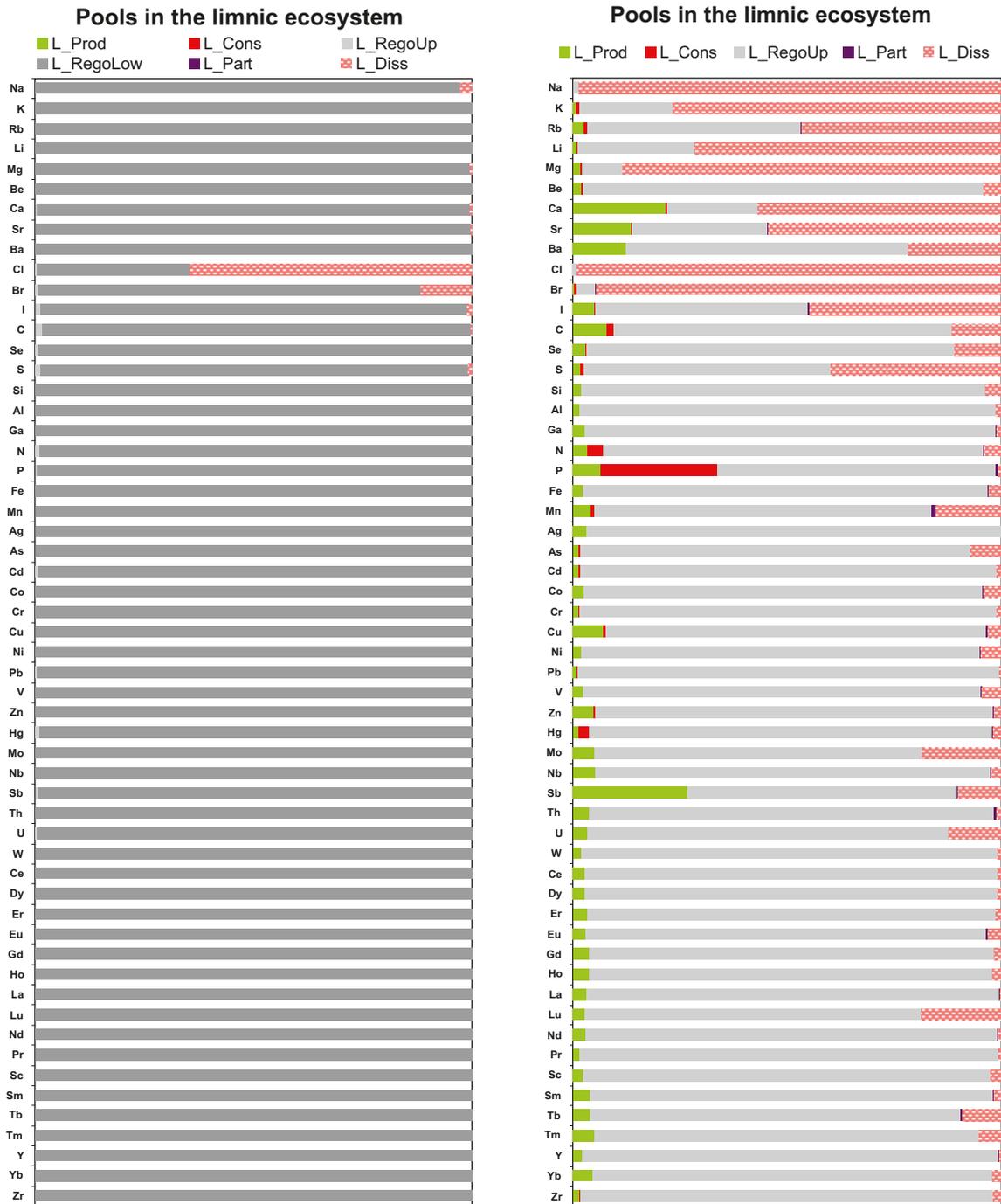
In the limnic system the dissolved pools comprise a significant fraction of the total amounts of marine ions (e.g. Cl, Br, Na, Mg) according to Figure 5-11. When the L\_RegoLow pools are excluded, this pattern is further strengthened among the remaining pools which represent the total amounts available in short term. Approximately half of the total amounts of marine elements such as K, Li, Rb, and I, S are found in the dissolved pools if the L\_RegoLow pools are excluded. In the Forsmark area marine influences either originate from recurring intrusions of Baltic sea water into the lower located lakes, or by flushing of relict marine remnants /Tröjbom et al. 2007/.



**Figure 5-10.** Comparison between terrestrial and limnic element pools in the Forsmark area. In the left panel, the terrestrial and limnic consumer and producer pools are combined in the 'T\_Bio' and 'L\_Bio' pools, respectively. The upper and lower regolith pools are combined in the 'T\_Rego' and 'L\_Rego' pools. Particulate and dissolved fractions of the lake water are combined in the pool denoted 'L\_water'. In the right panel the RegoLow pools are excluded and the regolith is represented by 'T\_RegoUp' and 'L\_RegoUp', respectively. This latter selection represents the pools available for element cycling in a short perspective. The abbreviations are further explained in Table 5-1.

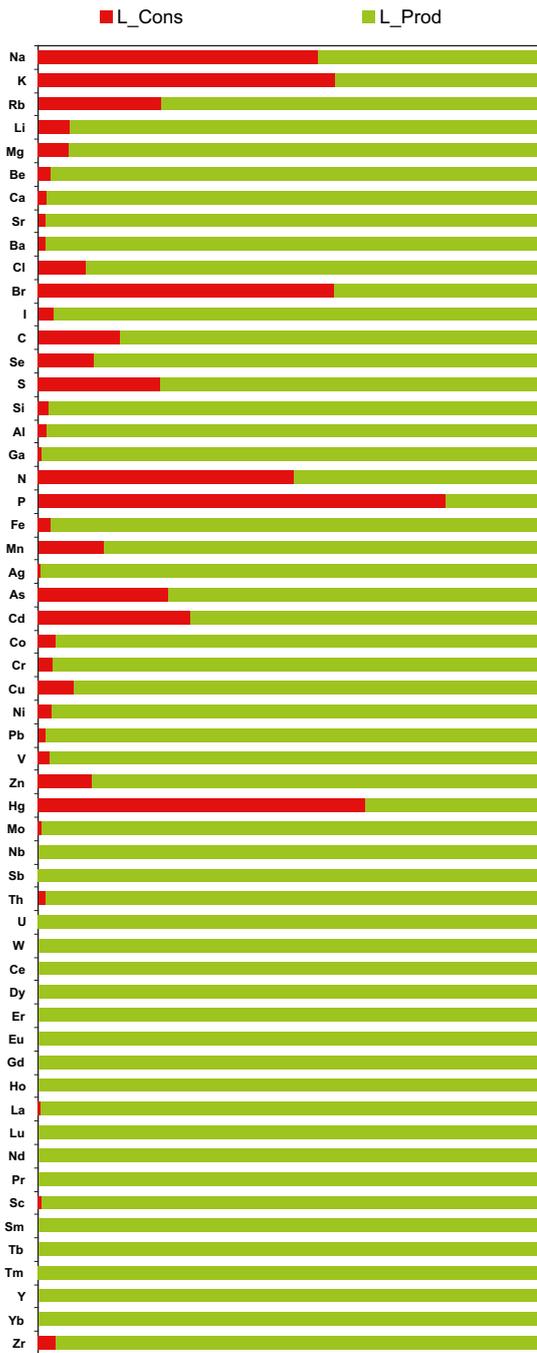


**Figure 5-11.** Relative distribution of elements among the different pools of the terrestrial ecosystem in the Forsmark area. Producers (*T\_Prod*), consumers (*T\_Cons*), upper regolith layer (*T\_RegoUp*) and lower regolith layer (*T\_RegoLow*). In the left panel, the lower regolith layer (*T\_RegoLow*) is for most elements totally dominating and the biotic pools are for some elements not visible. In the right panel, the *T\_RegoLow* layer has been excluded and the pools represent the amounts available in short term. The abbreviations are further explained in Table 5-1.



**Figure 5-12.** Relative distribution of elements among the different pools of the limnic ecosystem in the Forsmark area. Producers (*L\_Prod*), consumers (*L\_Cons*), upper regolith layer (*L\_RegoUp*), lower regolith layer (*L\_RegoLow*), particulate matter in lake water (*L\_Part*) and dissolved elements in lake water (*L\_Diss*). In the left panel, the lower regolith layer (*L\_RegoLow*) is for most elements totally dominating and some of the biotic pools are often not visible. In the right panel, the *L\_RegoLow* layer has been excluded and the pools represent the amounts available in short term. The abbreviations are further explained in Table 5-1.

### Limnic producers and consumers



**Figure 5-13.** Relative comparison between limnic producer (L\_Prod) and consumer (L\_Cons) pools. The abbreviations are further explained in Table 5-1.

The particulate pools are usually negligible compared to the dissolved pools as well as the other pools in the limnic system according to right panel of Figure 5-12. The particulate fraction comprises usually less than 20% of the total amount in the water phase. P, Pb, Th, La (REE) constitute exceptions where the particulate fraction may comprise up to 50% of the total amount in the water phase. In the limnic ecosystem, dissolved P is usually a limiting nutrient which is readily taken up by biota and particulate species of P are formed through co precipitation with calcite in the oligotrophic hard water lakes area /Brunberg and Blomkvist 2003/.

Compared to the distribution among the pools in the terrestrial system the limnic consumer pools (mainly fish) are not negligible for P, N and Hg (Figure 5-12). The two former elements are nutrients incorporated into biomass, whereas the latter, Hg, is bio-accumulated in the food web. If the limnic producer and consumer pools are compared in Figure 5-13, some elements seem to be accumulated in the consumer pool compared to the producer pool, e.g. nutrients such as Na, K, S, essential trace elements such as Zn, Mn, Se and also Cd, As and Rb. The three latter elements have no known biochemical function and the accumulation of these elements is probably an effect of co transport via active uptake pathways. Rb, for example, is supposed to follow the same pathway as K.

Among the halogen pools in the limnic system, Br and I are, in comparison with the very mobile and conservative element Cl, to a higher degree accumulated in the limnic biota pools or the L\_RegoUp pool in relation to the total amounts available in the limnic system (Figure 5-12). At landscape level the atmospheric input seems to be an important source for all these elements according to the mass balances in Section 5.2.1, and the overall landscape retention is higher for I than Br and Cl (cf. Section 5.2.4).

## 5.4 Estimation of time scales

In this section fluxes (mass transfer per unit time) are compared to the estimations of the total contents of different pools. Mass estimations divided by estimations of mass transfers per unit time results in estimations of time scales giving indications of the past and future evolution of the chemical environment in the Forsmark area. It should be noted that these estimations assumes future conditions not significantly altered from the situation in the present Forsmark area. Alternative climate cases as Global Warming or Permafrost might significantly alter governing processes and export rates making these estimations less valid (cf. Section 3.4.3).

If the pools in previous Section 5.3 are divided with yearly fluxes in Section 5.2, an estimation of the time needed for a total depletion at constant export rate is obtained. This time span is a minimum estimate and in practice the export rate is reduced over time proportional to the remaining storage resulting in longer time spans for total depletion. As the decay constant is not known for e.g. the weathering reactions investigated here, constant export rates are assumed as a base case. If exponentially decreasing export rates are assumed and the half life period is estimated from the initial export rate and the initial pools, 1 percent of the initial amounts remain at a time span corresponding to 3.3 times the time span assuming constant export rate. As described in Section 5.1.2 the terrestrial lower regolith pool (T\_RegoLow) is underestimated 8 times due to the underlying assumption of an active zone of 0.5 m. In these estimations the entire profile should be included, which means that the estimated time scales regarding the T\_RegoLow layer should be increased 8 times. If both these reservations are taken into account, the time spans regarding the T\_RegoLow layer should be extended up to 25 times.

Depending on the combinations of pools and fluxes different estimates are obtained:

- If the terrestrial regolith pools are related to the yearly export via surface water, this estimation gives a measure of how long time the pools will last at the present export rate (left panel in Figure 5-14). If the main source of the element is weathering reactions in the regolith and bedrock, this estimation gives a rough measure of the time span until the terrestrial pool is consumed by weathering at present rate. If the main input of the element is atmospheric deposition the terrestrial regolith/discharge ratio gives no indication of the time span of the actual mineral weathering at the site. The regolith/discharge rate reflects in such case the hypothetical time the pool will last at present discharge rate with no further contributions from atmospheric deposition.

In Figure 5-15 the regolith pools are related to the actual weathering rate, i.e. the weathering rate compensated for the atmospheric input, thus giving an alternative estimation of the time scales of flushing and weathering processes for some elements.

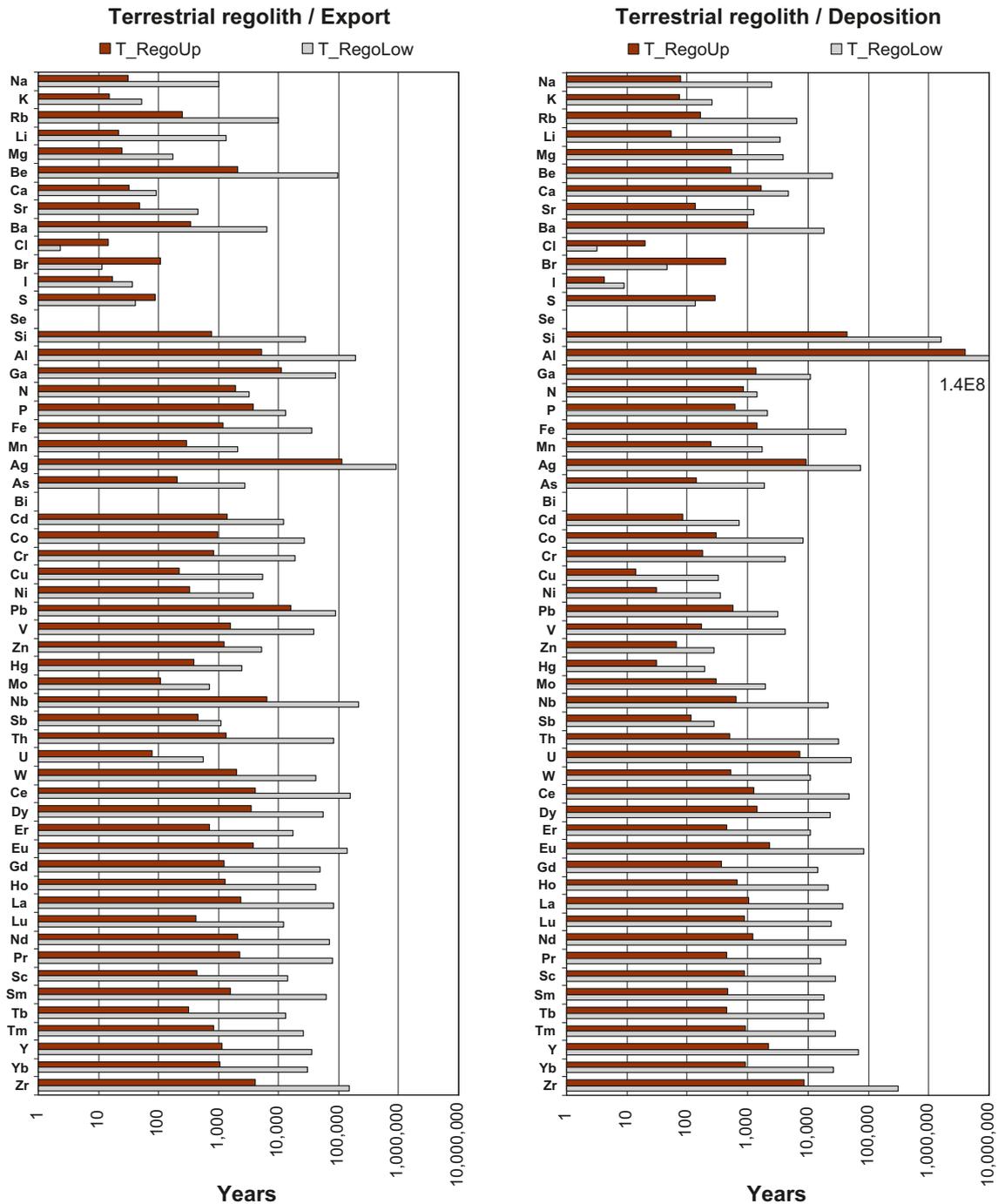
- If the terrestrial regolith pools are related to the atmospheric deposition input, the estimated time span gives a rough measure of the residence time in the regolith (right panel in Figure 5-14). Or, if the export is negligible compared to the input, the time span for accumulation in the regolith. This estimation is only valid if the atmospheric deposition is the main input to the system and input from mineral sources are negligible.

Si and Al are released to the soil solution as a result of weathering reactions in the regolith and bedrock. These elements originate from dissolution of more or less hard-weathered minerals as e.g. micas, feldspars and quartz, and the atmospheric input of these elements are negligible compared to the flux from the local mineral supply. At present weathering rate, the pools of these elements in the upper regolith layer (T\_RegoUp) will last for 1,000–10,000 years. The larger T\_RegoLow pools will last for 10,000–100,000 years at present rate according to Figure 5-14. Due to the underestimation of the T\_RegoLow pool the actual amount of regolith available in the Forsmark area is probably a magnitude larger (cf. Section 5.1.2), which means that the time span is extended to at least 100,000–1,000,000 years. Minerals based on Si and Al in the regolith layer will probably resist chemical weathering and mechanical erosion with margin until the next glaciation when a new regolith layer is deposited in the Forsmark area.

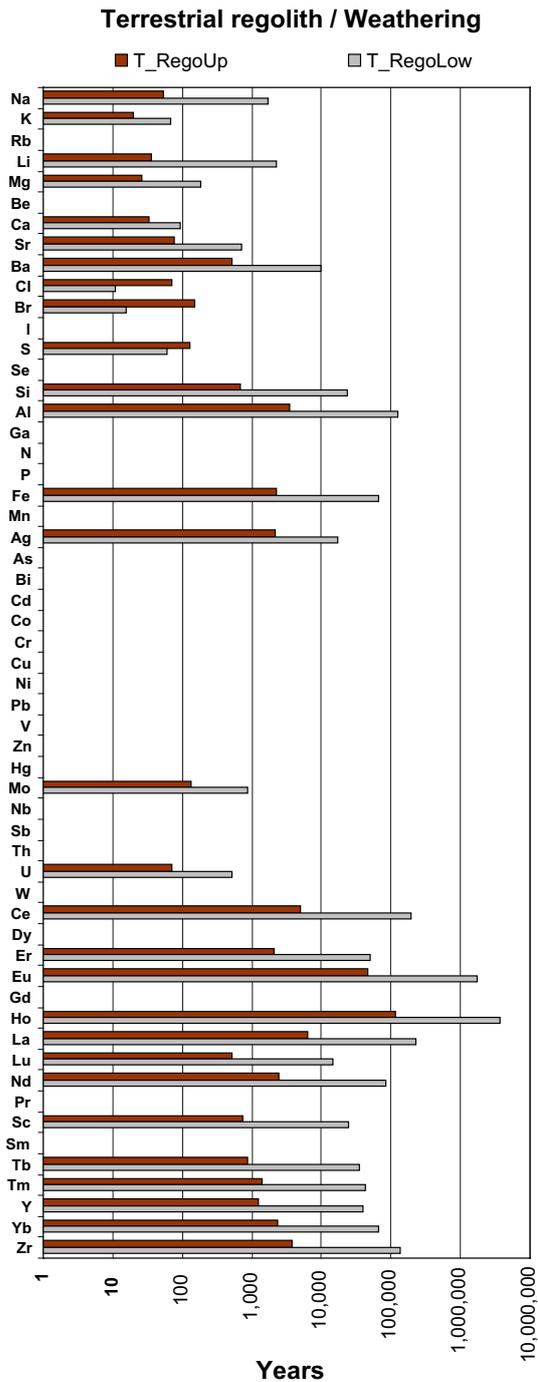
In the Forsmark area calcites in the regolith have a profound effect on the hydrochemistry in the surface system, cf. Section 3.2. According to the estimation in Figure 5-14 the Ca pool in the regolith layers will only last for 10–100 years at present weathering rate. This means that the calcite in the till will be consumed and that the most important prerequisite for the deviating hydrochemistry in the Forsmark area will change in a relatively near future. Although uncertainties regarding future weathering rates and the total amount of calcite available in the regolith, this estimation show that the calcite is a chemical factor that will change significantly in at least a 3,000 year perspective if the total amounts of regolith and exponentially decreasing weathering rates are accounted for (100 years·8·3.3). If the weathering of a mineral is driven by a constant supply of biogenic H<sup>+</sup>, the weathering rate could be expected to be nearly constant until weathering of other minerals competes with the dissolution front advancing downwards in the soil profile.

For many marine ions, there is a significant contribution to the surface system from marine remnants in the regolith since the area was covered by sea water /Tröjbom et al. 2007/. If the present atmospheric input rate is withdrawn, ongoing flushing of e.g. relict Cl and Br will theoretically drain the T\_RegoLow pool within 10 years and the T\_RegoUp pool within 100 years according to Figure 5-15. Atmospheric deposition will maintain the Cl export at a slightly lower level compared to the present rate as long as the present atmospheric deposition rate continues (cf. Section 3.4). Other ions of marine origin, such as Na, Mg and K, are also supplied via weathering of minerals in the regolith and bedrock. When the relict marine supply of these ions is reduced, the discharge rate will stabilise at a level corresponding to weathering of e.g. silicates and the future deposition rate (cf. the area specific transport of e.g. Na and Mg in relation to Si in the Storbergsbäcken catchment in Figure 5-7).

For several metals (e.g. As, Cd, Co, Cr, Cu, Ni, Pb, V, Zn, Hg, Mo, Sb, Nb, W) the total supply in the T\_RegoUp pool corresponds to approximately 100 years of atmospheric deposition at the present rate (with a span of 10–1,000 years depending on metal). The pool in the T\_RegoLow layer is usually a magnitude larger (cf. Section 5.3). According to the mass balances in Section 5.2.1 a significant fraction of the atmospheric input of these elements seem to be retained in the terrestrial system, either in the biota pools, the organic T\_RegoUp layer or in the mineral soil (T\_RegoLow). The estimations of the hypothetical flux from the T\_RegoLow layer in Section 5.2.3 show that groundwater discharge from this level could only explain a minor fraction of the total export via surface water, although the total amounts of these metals are significantly larger in the T\_RegoLow layer. This pattern could be interpreted as if mainly the upper soil layers are participating in the retention of the air transported heavy metals and that the relatively large pool in the T\_RegoLow layer is more or less immobile.



**Figure 5-14.** The terrestrial regolith pools divided by the annual mass export via surface water discharge (left) and the annual input via atmospheric deposition (right) give estimations of time scales which have different meanings depending on the element of interest. If exponentially decreasing export rates are assumed 1 percent of the initial amounts remain at a time span corresponding to 3.3 times the time span assuming constant export rate. As described in Section 5.1.2 the terrestrial lower regolith pool (T\_RegoLow) might be underestimated 8 times due to the underlying assumption of an active zone of 0.5 m, which means that the estimated time scales regarding the T\_RegoLow layer should be increased 8 times to account for the whole vertical extent of the T\_RegoLow layer. If both these reservations are taken into account, the time span regarding the T\_RegoLow layer might be extended 25 times.



**Figure 5-15.** The terrestrial regolith pools divided by the estimated annual amounts released to the surface water by weathering in the terrestrial system, i.e. the rate compensated for the atmospheric input, thus giving an alternative estimation of the time scales of flushing and weathering processes for some elements. If exponentially decreasing export rates are assumed 1 percent of the initial amounts remain at a time span corresponding to 3.3 times the time span assuming constant export rate. As described in Section 5.1.2 the terrestrial lower regolith pool (T\_RegoLow) might be underestimated 8 times due to the underlying assumption of an active zone of 0.5 m, which means that the estimated time scales regarding the T\_RegoLow layer should be increased 8 times to account for the whole vertical extent of the T\_RegoLow layer. If both these reservations are taken into account, the time span regarding the T\_RegoLow layer might be extended 25 times.

The patterns for the rare earth elements, REE, are hard to interpret and somewhat contradictory. Uncertainties seem to be large for this group of elements and interpretations should be done with this in mind (cf. Appendix B, Section B.2.8). According to the mass balances the retention of this group of elements in the limnic system is larger compared to e.g. Al and Si, and at the same time, atmospheric deposition is not negligible compared to the total flux out of the system via surface water discharge according to these balances. The overall pattern of REE in Figure 5-14, which resembles Al and Si, could be interpreted as if the release of this element group is mainly related to long term weathering reactions in the regolith, which is in line with the presumed geogenic origin of this element group. In this respect, the atmospheric input corresponding to only 10,000–100,000 years deposition in relation to the T\_RegoLow pool seems to be overestimated if compared with Al and Si.

The main conclusions from this exercise are that medium and hard weathered minerals in the regolith will last with margin until the next deglaciation, whereas easily weathered minerals as calcite will be consumed in a relatively near future (1,000–10,000 years). Marine remnants in the regolith will probably also be flushed out in a 100 year perspective. The build up of several metals in the upper parts of the soil horizon corresponds to approximately 100 years of atmospheric deposition at the present rate (with a span of 10–1,000 years depending on metal). Consequences of these changes are further discussed in Chapter 6.

## 5.5 Conclusions and discussion of uncertainties

The estimations described in this section are associated with several uncertainties. The broad range of elements studied show that there are general patterns for the distribution and behaviour in the landscape of different groups of elements. Mass balances reveals major sources and sinks, pool estimations show where elements are accumulated in the landscape and estimations of time-scales give indications of the future development. This general knowledge could be transferred to the radionuclides described in Chapter 6 in order to describe their behaviour and distribution in the landscape. Despite the large number of uncertainties associated to a specific element, the overall pattern reflecting the behaviour of the whole range of elements is most probable significant. Unexpected deviations from these general patterns either reflect unique properties of specific elements important for the understanding of their behaviour in the Forsmark area, or specific uncertainties related to this element. Specific uncertainties regarding input data and model assumptions are discussed in Appendix B, Section B.2, whereas some general uncertainties affecting the overall results are discussed below.

In the mass balances described in Section 5.2, estimations of atmospheric deposition seem to be one of the major uncertainties (cf. Appendix B, Section B.2.8). This is not exclusively a question of choosing the best method for estimating dry and wet deposition, but to correctly interpret what these measures represent. There are several pitfalls related to this topic: Compared to estimations of deposition, emissions to the atmosphere are much harder to estimate, and neglecting the emissions in the mass balances implicates that they are included in the mass balance term. This term is interpreted depending on the sign either as release or accumulation of elements in the terrestrial system and if the emissions are not negligible the net input from the atmosphere is exaggerated in the balances. For elements with a bidirectional gas exchange with the atmosphere, e.g. C and N, only the net input of dissolved substances is accounted for in the mass balances, which means that these models not necessarily describe the overall patterns for these elements correctly. For other elements the potential problems are less obvious. Deposition measurements might for example contain particles both originating from the near surroundings and areas outside the subcatchment (which is the system border for the mass balance). If the deposition data include significant amounts of matter originating from the same catchment, this implies that the actual net deposition is overestimated. This might be applicable on geogenic elements (e.g. Fe, Mn, Zr, Sc, Y, and REE) as well as nutrients (e.g. P) transported via for example pollen. If there is also a significant export of matter from the catchment via emissions to the atmosphere, the mass balances might be even more biased. The net exchange between two areas is determined by differences in properties and composition of the areas. The apparent net exchange via the atmosphere between two similar catchments attributed to similar processes could therefore be assumed zero. If the studied landscape adjoins an area with pronounced differing properties, e.g. the Baltic Sea in case of Forsmark, the net exchange between these areas

will be determined by this difference. Deposition or aerosols of sea water is an example in the Forsmark area of a directional net influence from the surroundings affecting elements such as Na, Mg, K, Cl and Br.

The estimations of the pools in the landscape described in Section 5.3 are attributed to several uncertainties described in Appendix B, Sections B.2.5 and B.2.6. Both the estimations of the total extent of the pools as well as the assigning of representative concentrations for specific elements are impaired with uncertainties; cf. the T\_RegoLow pool definition in Section 5.1.2. When these data are further used in the estimations of time scales, the total extent of the pools are not necessarily relevant for the question at issue. In this report no attempt has been made to further differentiate these pools, but potential effects are discussed.

The estimations of time-scales in Section 5.4 depend on several assumptions and uncertainties which influence the results. Uncertainties behind the pool and flux estimations described above and the selection of the pools relevant for the question at issue are of course important, but also the assumptions of the future decay rates, i.e. in what manner the for example weathering rates will change over time. Uncertainties regarding the time for depletion of the calcite in the Forsmark area couples to this question. Estimations based on the total contents in the regolith and present export rates via surface water discharge implies that the calcite will be depleted within approximately 1,000 years assuming constant export rate. If an exponentially decreasing weathering rate is assumed the time interval is extended 3 times to 3,000 years, compared to the constant weathering case, until 1% of the initial amounts remain. As the weathering rate of the calcite is governed by the supply of  $H^+$  mainly derived from decomposition of organic matter, it could be assumed that the rate will be approximately constant until the calcite is significantly depleted in the soil horizon or until weathering of other minerals consumes significant amounts of  $H^+$  at the expense on the calcite dissolution. This scenario implicates that the effects on the chemical composition of the surface water will be nearly constant until a significant depletion of the calcite has occurred. Investigations by /Ingmar and Moreborg 1976/ show that calcite has been depleted down to considerable depths in the soil profile in Northern Uppland since the withdrawal of the sea less than 10,000 years ago. Another question is which area specific export rates of Ca is needed to maintain a significant influence on the ecosystems in the future Forsmark area. If the decrease of the Ca export rate follows a more extended course it could not be ruled out that the effects on the ecosystems might be significantly longer compared to the estimations above, even if a significant fraction of the total amounts of calcite in the regolith have been depleted.

## 6 Future development of the chemical conditions at Forsmark and implications for selected radionuclides

In this section the presumed future chemical development in the Forsmark area is outlined in a 10,000 year perspective and the implications of a changing chemical environment are discussed for a selection of radionuclides. This interpretation is based on the present distribution of stable elements described in Chapter 5, the previous development of the Forsmark landscape in Section 3.3 and the factors forming the future chemical environment described in Section 3.4. The intention with this chapter is to put the pieces together and, with focus on the selected radionuclides, point at possible implications of the future chemical development for parameters used in the radionuclide model in the Safety Assessment.

### 6.1 The chemical development in a 10,000 years perspective

If the factors and driving forces described in Section 3.4 change, or if for example limited resources are depleted by ongoing processes, chemical conditions in the surface system will be altered in the future. In this section probable responses and potentially changing conditions are outlined. General implications of the altered conditions are also discussed for the selected radionuclides (cf. Section 4.1).

#### 6.1.1 Diminishing influence of the calcite rich till

Calcite bearing till deposited during the last glaciation has had profound effects on the hydrochemistry in the Forsmark area. Weathering reactions driven by biogenic  $H^+$  started to consume this calcite when the sediments emerged from the sea c. 2,500 years ago and meteoric recharge altered the hydrological flow patterns /Tröjbom et al. 2007/. Weathering of calcite releases  $Ca^{2+}$  ions and increases alkalinity in the shallow groundwater and fresh surface water. The supply of  $Ca^{2+}$  ions also mediates cation exchange processes in the regolith which in turn release cations as  $Na^+$ .

The ample supply of  $Ca^{2+}$  ions and the high alkalinity gives premises for precipitation of calcite in wetlands and lakes forming calcareous sediments /Hedenström and Sohlenius 2008/. During precipitation other elements co precipitate and accumulate in the sediments as well. This is the case for the essential nutrient P, which is withdrawn from the water phase leading to reduced primary production and development of the oligotrophic hard water lake type found close to the calcite influenced coast of northern Uppland /Andersson 2010, Ingmar and Moreborg 1976/. The limnic ecosystems in the Forsmark area are hence heavily affected by these specific hydrochemical conditions. Besides P, other elements could theoretically also be co precipitated in this hydrochemical environment, e.g. Sr and perhaps also elements associated to particles.

In the terrestrial ecosystem, the effects of the calcite are perhaps less pronounced compared to the limnic ecosystems and mainly manifested by the occurrence of some lime favoured species and the formation of some specific soil types /Löfgren 2010/. At smaller scale, the rich supply of the essential nutrient Ca could potentially affect primary production by the formation of fertile soils as well as biochemical characteristics of the plants. As an example could the 10 times elevated Cl contents of plant tissues in the Forsmark area be a consequence of a surplus uptake of Ca, where Cl perhaps acts as a counter ion balancing  $Ca^{2+}$  in the vacuoles of the plants /Tröjbom and Nordén 2010/.

Estimations based on pool estimates and present weathering rates suggests that the calcite in the regolith layers might be consumed by weathering reactions in a relatively near future, perhaps within a time span of some 1,000 years depending on the assumptions (cf. Section 5.4). This conclusion could perhaps seem contradicted by the fact that the area specific transports of Ca in the larger catchment of Forsmarksån nearby are comparable or even slightly higher than in the Forsmark area. It could be presumed that calcite in this older landscape should to some extent have been

consumed and that area specific transport should be lower than in the younger Forsmark area. On the other hand, if the calcite dissolution is mainly controlled by the available amounts of  $H^+$  driving the weathering reactions, area specific transports will not be reduced until the amounts of calcite is significantly depleted in the active soil layers. Studies of /Ingmar and Moreborg 1976/ showed that the calcite in the soil horizon has been consumed down to considerable depths further inland from Forsmark and that there is clear gradient towards the coast. The observation that the oligotrophic hardwater stage usually persists a few thousands years perhaps supports the diminishing calcite influence, although other factors in the catchment could have influence on the presence of this lake type /Nordén et al. 2008/.

These estimations implicate that the calcite influence will diminish in a relatively near future, at least in a 10,000 year perspective. The present deviating conditions of the Forsmark area will therefore change towards the hydrochemical environment seen in most other parts of Sweden (cf. Section 5.2.2). In the prolongation this has implications of the magnitude of processes as co precipitation and primary production in the limnic ecosystem. Dissolved Ca and P, as well as pH and alkalinity will change. The surface water quality in the Forsmark area will approach the conditions found in the brown-water lakes and streams further inland today. This probably means higher concentrations of total organic carbon (TOC) as well as concentrations of total P.

Empiric, site specific constants determined in the present Forsmark landscape, e.g. CR and  $K_d$  ratios, may hypothetically be less valid in the altered future chemical environment where other processes becomes more important. In case of site specific  $K_d$  values, the occurrence of calcite and co precipitation of related elements could lead to overestimated  $K_d$  values, i.e. a larger fraction seems to be adsorbed to the solid phase when these calcite precipitates are included. Site specific CR values, where concentrations in biota are related to concentrations in soil, could by the same reason be slightly underestimated due to the overestimation of the fraction adsorbed to the solid phase in the data from Forsmark.

This will potentially affect parameter estimates of radionuclides of Ca and also Sr. Sr behaves geochemically in many respects similar to Ca and is incorporated in the calcite. Parameter estimates of other elements of the same group of the periodic system, e.g. Ra, which is indirectly coupled to the calcite system, e.g. via co precipitation, could also be affected /Sena et al. 2008/.

### **6.1.2 Increased weathering of Si and Al minerals**

Chemical weathering of minerals more hard-weathered than calcite is also driven by the supply of  $H^+$  mainly generated during decomposition of organic matter and the formation of carbonic acid. Strong acids dissolved in the precipitation or formed by oxidation of sulphides also contribute with  $H^+$ . The maximum weathering potential is limited by the available amounts of  $H^+$ . Different minerals are persistent to weathering to a varying degree and if there are more easily weathered minerals present, the dissolution of these minerals consume  $H^+$  at the expense of the weathering of more hard weathered minerals. This means that weathering rates of e.g. silicates are expected to be lower in the regolith in the Forsmark area today compared to similar sites with no calcite present in the regolith. Area specific transports of Si and Al are also lower in the Forsmark region (both in the Forsmark area and in the catchment of Forsmarksån) compared to e.g. the Laxemar-Simpevarp area and Svartberget/Krycklan catchment (cf. Section 5.2.2).

It could therefore be presumed that weathering rates of hard weathered minerals as silicates will increase in the future when the calcite in the regolith is consumed. This in turn implicates that the time span estimated for the weathering of e.g. Si in the regolith might be overestimated in Section 5.4. Also weathering of other major constituents originating from this type of minerals, e.g. Al and Na, Mg, Ca, are supposed to be enhanced in the future chemical environment, as well as release of trace elements incorporated in the bulk minerals, e.g. U, Th, as well as e.g. Zr and REE.

The implications of the increased weathering rates of hard weathered minerals are possibly small for parameter estimations and the results of the radionuclide model. Increasing concentrations of geogenic elements, such as U, Th and Zr, in the environment do not necessarily implicate that relative parameters as  $K_d$  and CR are influenced if the major processes controlling the behaviour of the radionuclide are still unchanged.

### 6.1.3 Reduced marine influences

Surface water in the Forsmark area is characterised by relatively high concentrations of marine ions as Cl, Na and Mg. These elements either originate from relict marine sea water since the area was covered by the Baltic Sea, or from ongoing atmospheric deposition (sea spray). According to the mass balance in Section 5.2.1 approximately 1/3 of the total export of Cl could be explained by relict marine sources. The recent withdrawal of the Baltic Sea has left marine remnants in the bedrock as well as in the regolith in the Forsmark area /Tröjbom et al. 2007/. These relicts that are generally present below the highest coast line along the Baltic coast will consequently be flushed by recharging meteoric water and finally reach steady state corresponding to the atmospheric deposition. In the lower elevated areas at Forsmark, this process is ongoing, whereas it is almost completed in the higher parts. In fact there are still marine intrusions directly influencing the lakes close to the sea level (e.g. Lake Bolundsfjärden) /Tröjbom et al. 2007/.

Within approximately 100 years, this relict marine Cl-pool will be flushed according to the estimation in Section 5.4 (cf. Figure 5-15, which have been compensated for the present atmospheric input). This estimation is of course heavily dependent on assumptions of the vertical extent of the regolith as well as the typical concentrations in this pool. Despite these uncertainties, this estimation however shows that the relict marine influences will decrease in a relatively short time perspective (cf. /Johansson 2008/).

If steady state has been reached, the discharge of marine ions (e.g. Cl) is mainly determined by concentrations in the atmospheric deposition. In the future the deposition of elements originating from sea water will potentially decrease when the Baltic coast moves further east due to the land rise. When the brackish Sea Bottenhavet turns into a fresh water lake, the marine influence will be further reduced and the Cl export from the area will finally approach a level of 0.1 g/m<sup>2</sup>/yr corresponding to atmospheric deposition in the region.

The global warming climate case (cf. Section 3.4.3) could counteract or reverse the development towards reduced marine influences in the Forsmark area if the sea water level rises. If the Forsmark area again is covered by sea water, marine ions will be transported into regolith and bedrock by density driven recharge (cf. Section 3.3).

The main inputs of Cl and I in the present ecosystems are from marine sources. A large fraction of the total amount of Cl available in the surface system is incorporated into living matter as an essential element, and, at the same time Cl behaves conservatively with negligible retention in the soils of the surface system. I is also an essential element for biota, but with higher tendency for retention in the soils compared to Cl. As these elements are essential elements, the uptake in plants is not necessarily proportional to the available amounts in soil. This means that CR ratios for Cl and I, determined in the current marine influenced environment, could theoretically be underestimated in the future environment when the availability of these elements is reduced.

Lowered levels of sulphate in shallow groundwater and fresh surface waters could also be expected to appear in the future when marine remnants and pyrites in the regolith have been depleted. Also decreasing anthropogenic deposition will cause reduced sulphate levels in the shallow groundwater. This might have effect on the mobility and retention of Ba as well as Ra that co precipitates with this mineral during the prevailing conditions /Sena et al. 2008/. In the future when the sulphate concentrations probably are significantly lower due to less marine input, possibly lower atmospheric sulphate deposition and depletion of the pyrites, the influence from this reaction is perhaps reduced with increasing mobility of Ra as consequence.

### 6.1.4 Altered metal mobility

Mass balances in Section 5.2.1 show that many metals in the surface system mainly originate from atmospheric deposition and that a significant fraction of these metals seems to be accumulated in the terrestrial system. Isotope studies at other sites have for example shown that there is a slowly advancing Pb front in the soil horizon that will reach the discharging groundwater first after several thousands years /Klaminder et al. 2006/. There is consequently a potential for increasing area specific discharge of some metals in the future Forsmark area, either when the system has reached steady state or due to the advance of the metal front.

The potential decrease in pH, alkalinity and Ca concentrations due to the depletion of the calcite in the regolith will alter the hydrochemical environment in a direction that might affect metal complexation and metal mobility. As the mobility of many metals increases when pH is lowered, retention of these metals will potentially decrease in the future chemical environment. The mobility of other metals seems to be enhanced under present hydrochemical conditions and the presumed future changes could lead to reduced mobility of e.g. U (cf. /Smellie et al. 2008/ and area specific discharge in Section 5.2.2). In addition to these factors, organic matter contents and the form of particulate matter could also significantly govern metal mobility in either direction depending on element.

Many metals, e.g. heavy metals such as Cu, Zn, Pb, show enhanced mobility at lower pH which implicates that site specific parameters determined in the current chemical environment of the Forsmark area could be less valid when the conditions change. If the mobility is enhanced for these elements in the future chemical environment,  $K_d$  might be overestimated for radionuclides such as Ni, Nb, Pb, and Sn. Ni is an essential trace element taken up actively by plants and if the availability of this element changes, CR ratios determined during the current chemical conditions could be less valid in the future environment.

Atmospheric deposition of anthropogenic origin is the major source for many trace metals in the surface system at present date. Anthropogenic emissions of many metals have increased significantly in modern time and the conditions measured at the Forsmark site does not necessarily reflect a steady state in the soil profile. Relative parameters as  $K_d$  could nevertheless correctly reflect the partitioning between liquid and adsorbed fractions, whereas CR estimations could be biased by direct deposition of metals on the foliage.

### **6.1.5 Changing landscape morphology and physical properties**

The land rise process will in combination with the morphology of the future landscape create new catchments and new lakes in the land emerging from the Baltic Sea. This areas will contain larger areas of more fine grained sediments compared to the present Forsmark area /Hedenström and Sohlenius 2008/. Weathering rates and area specific mass discharge of some elements from these areas could be significantly higher compared to the present Forsmark land area which is dominated by till. This is especially evident if these soils are drained and cultivated similar to comparable arable soils further inland. Area specific losses of e.g. phosphorus is at least an order of magnitude higher for arable land compared to forest land /Tröjbom et al. 2007/, and a greater proportion of arable land compared the present land-use distribution could significantly increase the nutrient status and primary production in the fresh water recipients.

Lakes within the catchments formed downstream the present Forsmark area will receive water from larger catchments, e.g. Forsmarksån and Olandsån compared to the small drainage area in the present Forsmark area. This might have great effects on the lake ontogeny and the oligotrophic hardwater stage will probably be omitted due to the input of nutrients and coloured substances from upstream catchments. The chemical characteristics of these future lakes will probably more resemble the present lakes of Forsmarksån, e.g. the lakes “Norra Åsjön” and “Södra Åsjön”.

In a few thousands years, a number of relatively large and deep lakes will be formed in the deeper parts of Öregrundsgrepen /Lindborg 2010/. These lakes will possibly develop into the eutrophic lake type with a hydrochemistry more resembling e.g. Lake Erken further south along the coast. This type of lake is characterised by nutrient rich conditions, high primary production and a great potential for retention due to the large depth and long residence time.

Increasing primary production in future lakes due to higher availability of phosphorus together with longer residence times in the large and deep lakes might enhance the accumulation and retention in the limnic ecosystem. Also transport and retention of metals associated to organic matter could be expected to change when the surface water becomes more nutrient rich. Area specific transports of several metals are for example higher in the catchment of Forsmarksån compared to the Forsmark area, which might be explained by higher mobility in this chemical environment characterised by higher contents of organic carbon and nutrients.

The composition and properties of particulate matter as well as retention processes could therefore be significantly altered which in turn could affect  $K_d$  values for particulate matter, e.g. metals with high affinity for organic matter, such as Pb, Th as well as REE and some actinides.

### **6.1.6 General effects of alternative climate cases**

The above described development of the future chemical environment in the Forsmark area and the potential effects on the behaviour of the selected radionuclides assumes climate conditions not significantly deviating from the present conditions, i.e. temperate climate. During the next 10,000 years alternative climate cases are also possible: the global warming case leading to a warmer and wetter climate with rising sea levels, and the Permafrost case characterised by cold and dry conditions (cf. Section 3.4.3).

The Global Warming case implies higher temperatures, increased discharge and higher sea level. These factors will change the structures of the ecosystems as well as the magnitude of primary production and weathering rates, but if the underlying processes are unchanged compared to the temperate case, parameters that describe the behaviour of radionuclides in the ecosystems are not necessarily changed.

The Permafrost case implies a significantly colder and dryer climate compared to the present temperate conditions. During permafrost, discharge of groundwater is directed to vertical structures in the landscape called "Taliks", which probably coincide with the present lakes and other depressions in the landscape /Hartikainen et al. 2010/. During these conditions, characterised by less mobile water and lower temperatures, reaction rates are presumed to be lower, influencing e.g. weathering reactions. Primary production is probably also reduced implicating less influence from biota, e.g. uptake of nutrients, retention and presence of organic matter, which all are factors that might affect the behaviour of radionuclides in the landscape, as described in the sections above.

Beyond the time perspective of 10,000 years, the conditions at the surface will change radically when a new glacial cycle starts and the area is again covered by kilometres of ice. When the ice cover retreats the again submerged Forsmark area will be covered by fresh glacial deposits. These are exposed during the following land rise and the cycle will start all over again. Chemical signs of previous glacial periods and interglacials are mainly left in the bedrock as relict marine traces or isotopic signatures of the water reflecting other climates /Söderbäck 2008/.

## **6.2 Implications for selected radionuclides**

In this section, implications of the present chemical environment in Forsmark and the presumed future development are discussed for the selected radionuclides. These were selected because they contribute most to the dose to humans according to the SR-Site Safety Assessment and previous safety assessments (cf. Section 4.1).

### **6.2.1 Caesium ( $^{135}\text{C}$ )**

As there are few reliable measurements of stable Cs in the site investigation /Tröjbom and Söderbäck 2006a/, no conclusions could be drawn for this element from direct measurements. If the distribution and behaviour of Cs in the landscape resembles the related element Rb (cf. Section 4.1 and Appendix A – Description of the selected radionuclides), atmospheric deposition, probably originating from sea water, seems to be the most important input of this element in the surface system according to the mass balance in Section 5.2.1. If the pattern of Cs instead resembles K, sources in the terrestrial system are probably most important. Pool estimations in Section 5.3 show that most of especially Rb and also K are found in the regolith and that both these elements seem to be accumulated in biota compared to non-essential elements.

The future chemical environment of the Forsmark area, which is characterised by less input of marine ions as K, might theoretically result in greater uptake of Cs in plants (cf. Section 6.1.3). According to /Sheppard et al. 2009/, pH and clay contents are the main controlling factors of the mobility of Cs in the soils at Forsmark. Higher contents of fine-grained sediments and clay in the

future emerging land will probably lead to less mobility of Cs in these soils due to sorption to clay particles, probably resulting in higher  $K_d$  and lower CR in plant uptake. Increasing retention of Cs in the limnic system could also be expected in the future brown-water and the eutrophic lake types compared to the present clear oligotrophic hard water lakes, leading to increasing aquatic  $K_d$ . These retaining processes might be counteracted by a decrease in pH (cf. Section 6.1.1), which enhances the mobility of Cs.

### 6.2.2 Chlorine ( $^{36}\text{Cl}$ )

The major source of Cl in the present Forsmark landscape is atmospheric deposition of marine origin (cf. Section 5.2.1). About 25% of the Cl input originates from marine remnants since the latest deglaciation /Tröjbom et al. 2007/. In the terrestrial system, the absolute main part of the Cl is incorporated in the biota pool and in the organic soil layer (> 90% of the total amounts in the landscape). Only a minor fraction is found in the inorganic soil layer (cf. Section 5.3 and Appendix A – Description of the selected radionuclides).

In plants tissues, Cl concentrations relative to carbon are almost ten times elevated in the Forsmark area compared to the Laxemar-Simpevarp area. This difference is most pronounced for green parts of the plants but could be seen also among root and wood samples from higher plants. There is no difference regarding Cl between moss samples which have no root uptake. If not an artefact, the discrepancy between the sites could perhaps be explained by surplus uptake of Ca in the Forsmark area with Cl functioning as a counter ion (cf. /Tröjbom and Nordén 2010/).

Uptake by terrestrial biota is probably the major process that might affect Cl behavior in the future chemical environment. Diminishing atmospheric input of Cl and less Ca available for plant uptake due to the depletion of the calcite, could probably change CR ratios for Cl uptake in plants compared to present conditions. As several processes work in different directions the net effect on the CR is however not easily elucidated for Cl.

### 6.2.3 Iodine ( $^{129}\text{I}$ )

According to the mass balance in Section 5.2.1, atmospheric deposition of marine origin comprises the major input of I in the Forsmark landscape. Compared to the other halogens (Cl and Br) a greater fraction of I is incorporated into biota and retained in the landscape (cf. Section 5.2.4). The mobility of I is significantly lower compared to both Br and Cl, and the present chemical environment characterized by high alkalinity potentially contributes to the low mobility (cf. Appendix A – Description of the selected radionuclides). /Sheppard et al. 2009/ found that the mobility of I decreases at increasing organic matter contents, pH and clay contents. I concentrations are also significantly higher in plant tissues and fresh surface water in the Laxemar-Simpevarp area compared to the Forsmark area, a fact that perhaps supports the lower availability and greater retention of I in the chemical environment of the present Forsmark area /Tröjbom and Nordén 2010/.

In the future landscape with less influence from the calcite, the mobility of I could therefore theoretically be enhanced compared to the present situation and consequently have influence on parameters as  $K_d$  and CR ratios.  $K_d$  might decrease and CR perhaps increase. Diminishing marine input will also significantly reduce the amounts of I available in the ecosystems.

### 6.2.4 Molybdenum ( $^{93}\text{Mo}$ )

In the Forsmark area, Mo enters the landscape via atmospheric deposition and weathering of regolith and bedrock (cf. Section 5.2.1). The mobility of this element is relatively high, but a significant fraction is also retained in the limnic system (cf. Section 5.2.4). As an essential element, Mo is accumulated in biota as well as in organic soils (cf. Section 5.3 and Appendix A – Description of the selected radionuclides). /Sheppard et al. 2009/ found that the mobility of Mo decreases at increasing content of clay and organic matter, whereas decreasing pH enhances mobility. As the consequences of the presumed future chemical environment work in different directions, the net effect on Mo is not easily elucidated.

### 6.2.5 Nickel (<sup>59</sup>Ni)

Similar to other heavy metals, the major source of Ni in the present Forsmark landscape is atmospheric deposition, probably of anthropogenic origin. A very large fraction (c. 90%) seems to be retained in the terrestrial system and only minute amounts reach the limnic system (cf. Section 5.2.1). According to /Sheppard et al. 2009/ the mobility of Ni decrease with increasing pH and increasing contents of organic matter (cf. Appendix A – Description of the selected radionuclides). The high anthropogenic deposition input of Ni is most probably a relatively modern phenomena coupled to the generally increasing emissions of several metals during the last century. The largest Ni pools in the landscape are found within the mineral regolith pools, probably incorporated in minerals and not available in short term. As an essential element, Ni is accumulated in biota, but perhaps at a lower extent compared to e.g. Cu and Cd (cf. Section 5.3).

The future chemical conditions in the Forsmark area could probably increase the mobility of Ni and many other metals due to lowered pH when the calcite is depleted. As stated above, the present site specific data of Ni probably represent a situation biased by the dominating deposition of anthropogenic origin. Direct foliage uptake could affect the present site specific parameters by for example overestimating CR ratios, compared to a future environment with less deposition input. This potential problem is also applicable to several other metals, e.g. Pb and Cd.

### 6.2.6 Niobium (<sup>94</sup>Nb)

The distribution pattern of Nb in the Forsmark landscape is somewhat contradictory. According to the literature, cf. Appendix A – Description of the selected radionuclides, the behaviour of Niobium in water and in soils probably resembles the geogenic element Zr. Mass balances in Section 5.2.1 show that weathering in the terrestrial system is the main input of Nb, but also that atmospheric deposition is not a negligible source. The use of non-site specific deposition data (cf. Appendix B, Section B.1.2), could partly explain the perhaps unrealistic importance of atmospheric deposition of Nb in this estimation, if this rate is overestimated for the Forsmark area. Inclusion of deposition from very local sources as well as neglecting emissions to the atmosphere may also be responsible for this pattern, cf. Section 5.5. Compared to most other metals, e.g. Cu and Cd, a greater fraction is retained in the limnic system which could be explained by a higher degree of sedimentation of particulate bound Nb. /Sheppard et al. 2009/ suggests that the mobility of Nb is negatively correlated to clay content and pH in the soils of the Forsmark area.

The more nutrient rich characteristics of the future lakes of the Forsmark area, in combination with the presence of more fine-grained soils in the emerging land give premises for increasing retention of Nb in the limnic system if concentrations of organic and mineral particles increase in the fresh surface waters. This is also applicable on other particulate bound elements as Pb, Cd, Th and several actinides (see below).

As the consequences of the presumed future chemical environment work in different directions, the net effect on Nb is not easily elucidated; increasing retention due to increasing particulate matter in the surface water leads to higher  $K_d$ , whereas lower pH works in the opposite direction with higher mobility and lower  $K_d$  as a consequence.

### 6.2.7 Radium (<sup>226</sup>Ra)

There are fewer site specific measurements of Ra in the Forsmark area compared to most other elements /Tröjbom and Söderbäck 2006a/. Available measurements show that <sup>226</sup>Ra-activities are generally elevated in the Forsmark area compared to other parts of Sweden (cf. Section 4.2). This condition mainly reflects the elevated contents of U in the Forsmark area, of which <sup>226</sup>Ra is a decay product with a relatively short half-life.

As an analogue to Ca, Ra should also be absorbed by plants and accumulated in the calcified tissues of animals (cf. Appendix A – Description of the selected radionuclides). Although the analogous behavior in the geochemical cycles of the Ca, Sr, Ba, Ra element series, there are also chemical differences among these elements. Precipitation of calcite in lakes and wetlands is probably the major process governing the fate of Ca and Sr, but also to some extent affecting Ra via co precipitation.

The deviant behavior of Ba in the Forsmark area implicates that there are other processes controlling Ba, e.g. barite precipitation, which also could affect Ra /Sena et al. 2008/. Barite precipitation is controlled by sulfate, which is abundant in the marine influenced environment of the present Forsmark area.

In the future chemical environment of the Forsmark area, both the calcite influence (cf. Section 6.1.1) as well as the marine influence (cf. Section 6.1.3) might be reduced in a relatively near future of a few thousand years (cf. Section 5.4). This implicates that two important factors affecting Ra in the present Forsmark area also will change. If these solubility governed processes are included in the  $K_d$  values describing the present conditions, these parameter values are potentially overestimated in the future chemical environment.  $K_d$  for Ra might therefore decrease in the future chemical environment.

### 6.2.8 Selenium ( $^{79}\text{Se}$ )

Atmospheric deposition is the main input for Se in the Forsmark landscape according to the mass balance in Section 5.2.1, with sea water as the probable original source. Weathering of minerals in the regolith also releases Se in the landscape. Chemically, S could be regarded as an analogue to Se (cf. Appendix A – Description of the selected radionuclides).

If the distribution patterns of S and Se are compared in Figure 5-4, there is a profound difference between these elements: atmospheric deposition is the major input of Se and there is a substantial retention in the terrestrial system, whereas a significant portion of S is released from the terrestrial system, probably originating from marine remnants. Interpretation of S data is complicated by the radically changed atmospheric input during the last decades and previous deposition patterns could potentially affect the present mass balance. From this study is therefore not possible to distinguish if this discrepancy in distribution patterns reflects real differences in properties between these elements, or if uncertainties attributed to S and Se distort the picture.

/Sheppard et al. 2009/ suggested that there is a decreasing mobility of Se at higher pH in soils. This means that the mobility could be enhanced in the future chemical environment of the Forsmark area and parameters as  $K_d$  will be lower.

### 6.2.9 Silver ( $^{108m}\text{Ag}$ )

The mass balance for silver in the Forsmark landscape deviates from other heavy metals by showing a very large accumulation in the limnic system (cf. Section 5.2.1). This condition probably reflects the element specific property of Ag to form insoluble complexes with S and Cl (cf. Appendix A – Description of the selected radionuclides), which deviates other metals as Cu and Zn. Ag has no biological role in living organisms and there are no signs of accumulation in biota (cf. Section 5.3).

Uncertainties seem to be large for this element and the overall pattern is unrealistic compared to the other heavy metals, for example Cu. The observed pattern could however not entirely be disregarded as an artifact when original data is checked. Atmospheric deposition of anthropogenic origin is probably the major source for Ag, similar to most other heavy metals, which is also supported by the pool estimates in Section 5.3. A geogenic source exclusively for Ag in the Forsmark area and no other heavy metal as indicated by the mass balance is not probable.

In the future chemical environment of the Forsmark area, the reduced marine influence in combination with lower pH could lead to enhanced mobility of Ag in the surface system. This implicates that  $K_d$  values representing the present conditions might be overestimated in the future chemical environment and  $K_d$  for Ag might decrease in the future chemical environment.

### 6.2.10 Technetium ( $^{99}\text{Tc}$ )

In the site investigation, there are only a few measurements of the background activities of Tc (cf. Section 4.2). In the aquatic biota sample there are detectable amounts of Tc (1,500 mBq/kg), whereas the activity in the sediment sample is below the reporting limit (< 600 mBq/kg). This observation is in line with the presumed behaviour of Tc which acts conservative with low affinity to particulate matter and sediment, but with an extensive uptake in biota under aerobic conditions. Chemically

Tc is most similar to Re and to some extent to Mn, but due to the conservative behaviour of Tc, Cl perhaps could serve as an analogue at landscape level (cf. Appendix A – Description of the selected radionuclides). If the distribution pattern of Cl in the Forsmark landscape is applicable for Tc, only a minor fraction will be retained in the aerated parts of the regolith and in the lake sediments. If the release of Tc reaches the root zone, this element will also be readily taken up by the plants via the transpiration flow path in relation to the pore water concentration in the soil (cf. Section 5.3).

In the future chemical environment of the Forsmark area, the behaviour of Tc is expected to be less altered due to the conservative nature of this element. /Sheppard et al. 2009/ also concluded that Tc showed no significant correlation to any soil parameter.

#### **6.2.11 Thorium (Th), Uranium (U), Plutonium (Pu), Neptunium (Np) and Protactinium (Pa)**

Among the actinides regarded as important radionuclides in the safety assessment (cf. Section 4.1), Th and U occur in measurable amounts in the environment. These elements are also included in the extensive measurement program of the site investigation in the Forsmark area /Tröjbom and Söderbäck 2006a/. A smaller number of background activities have been measured for Pu, whereas there are no site specific measurements of Np or Pa.

If the distribution patterns of U and Th are compared in the Forsmark area, there is a profound difference. U is of geogenic origin with little retention in the surface system, whereas a significant fraction of the Th seems to be mainly supplied via atmospheric deposition and is readily retained in the limnic system according to the mass balance (cf. Section 5.2.1). These patterns reflect the different properties of U and Th, where U is very mobile in the present Forsmark chemical environment due to the formation of complexes with carbonates and Th is strongly associated to particles (cf. Appendix A – Description of the selected radionuclides). /Sheppard et al. 2009/ also concluded that the mobility of U in soil is governed by increasing pH (in soils with pH > 5.5).

The future chemical environment probably leads to lower mobility of U due to decreased formation of carbonates complexes. Retention of particulate bound elements as Th might perhaps increase in the future limnic systems characterised by higher contents of particulate matter and presumably higher retention (cf. Section 6.1.5). This probably leads to e.g. lower  $K_d$  for U and higher  $K_d$  for Th in the future environment.

If the behaviour of Pa is similar to Nb and Th (cf. Appendix A – Description of the selected radionuclides),  $K_d$  might also increase in the future chemical environment.

### **6.3 Summary and conclusions**

The future development of the chemical environment of the Forsmark landscape contributes to uncertainties in the estimated dose to humans. The future distribution in the surface system of radionuclides potentially released from the deep repository is modelled based on knowledge of present conditions and assumptions of the future development. As these results rely on a very large number of parameters describing conditions and processes in the current Forsmark area, the confidence in the results depends on if these parameter values also are valid in the future chemical environment /Nordén et al. 2010/. By using a deterministic approach based on best estimates, in combination with probabilistic simulations taking into account the spread of the input parameters /Avila et al. 2010/, uncertainties due to future conditions could be accounted for in the dose assessment. Conversely, if the future conditions deviate significantly from the conditions modelled, the model results could be less valid for some elements. The purpose with Chapter 6 is to discuss the qualitative implications of the presumed future chemical conditions of the distribution and behaviour of the radionuclides in general, and on site specific parameter values used in the radionuclide model in specific.

The presumed implications for the selected radionuclides are summarized in Table 6-1 and some general uncertainties are discussed below.

**Table 6-1. Summary of the effects of the future chemical conditions on the parameters describing the behaviour of radionuclides in the surface system compared to the present conditions.**

Radionuclide	Implications of the future chemical environment
Caesium ( <sup>135</sup> C)	K <sub>d</sub> might increase and CR decrease. Decreasing pH might counteract these effects
Chlorine ( <sup>36</sup> Cl)	No change
Iodine ( <sup>129</sup> I)	K <sub>d</sub> might decrease and CR for plant uptake perhaps increase
Molybdenum ( <sup>93</sup> Mo)	Possible changes of K <sub>d</sub> in either direction
Nickel ( <sup>59</sup> Ni)	K <sub>d</sub> might decrease
Niobium ( <sup>94</sup> Nb)	Possible changes of K <sub>d</sub> in either direction
Radium ( <sup>226</sup> Ra)	K <sub>d</sub> might decrease
Selenium ( <sup>79</sup> Se)	K <sub>d</sub> might decrease
Silver ( <sup>108m</sup> Ag)	K <sub>d</sub> might decrease
Technetium ( <sup>99</sup> Tc)	No change
Thorium ( <sup>230</sup> Th)	K <sub>d</sub> might increase
Uranium (U)	K <sub>d</sub> might increase
Plutonium ( <sup>239</sup> Pu, <sup>240</sup> Pu, <sup>242</sup> Pu)	No information
Neptunium ( <sup>237</sup> Np)	No information
Protactinium ( <sup>231</sup> Pa)	K <sub>d</sub> might increase

Modelling of the future landscapes, as well as experience from older inland catchments show that e.g. the limnic ecosystem of the investigated lakes in the Forsmark area deviates significantly from lakes inland and that new types of lakes will be formed in the future (cf. Section 6.1.5). Chemical parameters determined from site specific data from the present Forsmark area might therefore in some respects be less representative for the chemical conditions of the future chemical environment. The inclusion of data from the Laxemar-Simpevarp area, as well as generic data behind the parameter estimations could to some extent cover the discrepancy between present and future conditions. There are, however, still remaining uncertainties if the probability distribution functions of the radionuclide parameters described in /Nordén et al. 2010/ actually cover the future chemical environment at Forsmark, which affects the confidence of the radionuclide model results.

Uncertainties concerning the estimations of time-scales in Section 5.4 are not critical for the conclusions regarding the implications of the future chemical environment in the Forsmark area. There is no question that the calcite influence will diminish in a relatively near future compared to the long time spans modelled. In this respect it does not matter if the time span for significant depletion of the calcite is 1,000 or 10,000 years.

The alternative climate cases outlined in Section 3.4.3 affect the future development of the chemical environment in the Forsmark area at different levels. The magnitude of some of the processes discussed here will change due to altered temperatures and precipitation patterns (cf. Section 6.1.6). The Global Warming case might lead to higher primary production, increasing weathering rates and faster depletion of minerals e.g. calcite. Rising sea levels will again increase the marine influence in the lower located areas /Brydsten et al. 2009/. This means that both rates and spatial extent of some processes will change, but relative parameters such as K<sub>d</sub> and CR describing the nature of these processes might be unchanged.

The Permafrost case will significantly affect many processes as well as premises for the formation of ecosystems. Low temperatures, dryer conditions and reduced mobility of water probably lead to reduced rates of many processes in the landscape, e.g. weathering and mass transports. Low primary production and the permanently frozen soil layers might lead to conservation of the regolith and delayed depletion of e.g. the calcite. The influence on relative parameters such as K<sub>d</sub> and CR might be larger for the Permafrost case compared to the Global Warming case due to more pronounced differences from the present conditions regarding ecosystem structures and hydrological flow patterns.

It could be concluded that the future development of the chemical environment in the Forsmark area might affect element specific parameters used in the radionuclide model in different directions depending on element. The alternative climate cases, Global Warming and Permafrost, might have less influence on relative parameters such as K<sub>d</sub> and CR if the underlying processes are unchanged compared to the temperate case.

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## Description of the selected radionuclides

In this appendix a selection of radionuclides important regarding dose to humans are described. Site specific considerations and assumptions of which elements could serve as stable analogues are also accounted for in this compilation.

### **Caesium ( $^{135}\text{Cs}$ )**

Caesium behaves as a major cat ion in the environment where sorption increases with pH, leading to lower mobility in systems with high pH. Caesium has a strong affinity for sorption to clay minerals and is concentrated onto suspended particulate material and in bottom sediments. Both Rb and K are regarded as chemical analogues to caesium. The biochemical function of K as an essential element cycling in the biosphere mainly determines the uptake and cycling of caesium in the ecosystems, although there seems to be differences in the uptake mechanisms (cf. /Greger 2004/ and references therein).

### **Carbon ( $^{14}\text{C}$ )**

There are two stable forms of carbon,  $^{12}\text{C}$  and  $^{13}\text{C}$ , and one radioactive  $^{14}\text{C}$ . Carbon is fixed from the atmosphere by primary producers during photosynthesis and is the basis in food webs of all ecosystems. In the biosphere carbon is found in all biotic pools as well as in abiotic pools like soils, sediments, particulate matter and dissolved in water. The major part of carbon enters the food webs through the assimilation in plants from the atmosphere, which then is consumed by humans and other consumers. Stable carbon is the basis for the modelling of the ecosystems and this information is applicable on the radiogenic isotope  $^{14}\text{C}$ .

### **Chlorine ( $^{36}\text{Cl}$ )**

Chlorine is essential to almost all living organisms and very abundant in sea water. Chlorine occurs mainly as the  $\text{Cl}^-$  ion which is highly mobile in all chemical environments. In aquatic organisms the levels of chlorine present are subjected to strict homeostatic control /Coughtry et al. 1983/. The transfer from soil to plants of chlorine is extremely high, due to its high mobility in soils. Chlorine ions are mainly temporarily retained in the soil through sorption by vegetation. Following organic matter decomposition, chlorine passes into the soil solution and is thus, once again, available for plants /Colle et al. 2005/. Stable chlorine ( $^{35}\text{Cl}$  and  $^{37}\text{Cl}$ ) is regarded as an analogue to radiogenic  $^{36}\text{Cl}$ .

### **Iodine ( $^{129}\text{I}$ )**

Iodine is a biologically important element, required as a trace element for living organisms. Mobility and bioavailability are low under anoxic conditions, but significantly higher in e.g. aerated soils /Colle et al. 2005/. The iodine isotopes will generally have higher bioavailability than the stable form of the element under equivalent conditions. Root uptake of iodine occurs relatively easily /Coughtry et al. 1983/. Liming is found to decrease the availability of I to plants /Fuge and Johnson 1986/. Chemically, iodine is the least reactive of the halogens and hence differs in behaviour from Cl and Br. The stable isotope  $^{127}\text{I}$  is regarded as an analogue to radiogenic  $^{129}\text{I}$ .

### **Molybdenum ( $^{93}\text{Mo}$ )**

Molybdenum is a hard metallic transition element, chemically non-reactive but essential in trace amounts for most living organisms. Data concerning behaviour of molybdenum in soils and parental material is sparse; however, there seems to be a correlation between iron and molybdenum concentrations in soils. It is generally assumed that bone forms a long-term reservoir for molybdenum in living organisms. Stable Mo is measured in the site investigation and regarded as an analogue to the radiogenic isotopes.

### **Nickel (<sup>59</sup>Ni)**

Nickel is essential for some plants and there are also studies indicating that nickel is important for renal function among mammals. In soil Ni is rapidly adsorbed or occluded by the mineral lattice and is hence relatively unavailable for plant uptake. At anaerobic conditions, Ni is more available for plant uptake, whereas it is less mobile outside the pH-range of 6.5–7. The dominating transport phases of nickel in water are crystalline and particulate, presumably because of adsorption of nickel in metal hydroxide coatings of fine particles. With regard to aquatic animals it is suggested that, in the absence of specific data, reference should be made to the parameters determined for cobalt /Coughtry et al. 1983/. Stable Ni is measured in the site investigation and regarded as an analogue to the radiogenic isotope.

### **Niobium (<sup>94</sup>Nb)**

Niobium is a very rare metallic transition element. The behaviour of Niobium in water and in soils probably resembles that of Zr, but a higher proportion is probably bound to particles /Coughtry et al. 1983/. Nb has high affinity for sorption on particulate iron species which leads to an apparent correlation of these elements in surface water /Åström 2008/. Stable Nb is measured in the site investigation and regarded as an analogue to the radiogenic isotope. In addition, Zr and the related element Ti (and to some extent Si) could be regarded as analogues to Nb when the behaviour in the landscape is investigated. Zr occurs in the relatively hard weathered mineral zircon, which is abundant in e.g. granite rocks and Zr has no known biological role, similar to Ti and Nb.

### **Radium (<sup>226</sup>Ra)**

Radium is a decay product of uranium and is therefore found naturally in small amounts. As a homologue of calcium, which is an essential element in the plant metabolic system, radium is also adsorbed from the soil by plants. Radium preferentially accumulates in calcified tissue and skeletal parts. Bioaccumulation of Ra by fish is inversely proportional to the ambient concentrations of Ca in water as Ra probably utilizes the same uptake mechanism as for calcium. In the geochemical cycles, Ra are (similar to Sr) involved in solubilisation and precipitation reactions in parallel to CaCO<sub>3</sub> dissolution and precipitation (cf. /Sarkar 2002/). Co precipitation with other minerals, e.g. barite could also potentially affect the availability of Ra in the soil solution. Chemically Ba should be most similar to Ra. The biochemical role of Ca as an essential element of the ecosystems, together with the occurrence of calcites in the Forsmark area, probably governs the behaviour of Ra in the landscape.

### **Selenium (<sup>79</sup>Se)**

Selenium is present in all cells in living organisms, although at low concentrations. It tends to be easily taken up by organisms and is considered as essential to some plants and microbes and all vertebrates, where it has biochemical functions. Chemically selenium resembles sulphur /Coughtry et al. 1983/. Stable selenium is measured within the site investigation, but with lower sampling interval compared to most other elements. Sulphur is regarded as a chemical analogue to selenium when the distribution at landscape level is discussed.

### **Silver (<sup>108m</sup>Ag)**

Silver is a heavy metal with no biological role in living organism. The geochemical characteristics of Ag are usually similar to those of Cu, but at a 1,000 times lower level. Ag is easily released by weathering and then precipitated in media enriched in S compounds and halogens (e.g. AgCl). Ag complexes are immobile above pH 4, and MnO<sub>2</sub> has a significant affinity for Ag. Ag is taken up by plants and it seems that the concentration in plants follows the concentration in soil (cf. /Greger 2004/ and references therein). Stable Ag is measured in the site investigation and regarded as an analogue to the radiogenic isotope. In addition, other heavy metals could serve as analogues to Ag at landscape level, e.g. Cu, although the role as an essential element should affect the distribution of Cu to a higher degree than Ag.

### ***Technetium (<sup>99</sup>Tc)***

Technetium is a radioactive metallic transition element, in practice not naturally occurring in the environment as almost all technetium on Earth is produced synthetically. Chemical properties are intermediate between rhenium and manganese. Tc is very mobile in terrestrial environments and in general terms available to plants under aerobic conditions. Reducing environments decrease the mobility and availability of Tc to plants /García-León 2006/. Tc is very mobile in plants and readily transferred from roots to shoots and subsequently to developing fruit and seed tissues. Plants play an important role in the environmental cycling of Tc /Coughtry et al. 1983/. Tc is very soluble and mobile in seawater and behaves conservative by showing very low affinity to particulate matter and sediment. Reducing conditions in water and in the sediment, or in the sediment, facilitate its absorption on the sediment /García-León 2006/. Within the site investigation, there are only very few measurements of background activities of Tc in the Forsmark area. Depending on the conservative behaviour of Tc in most environments, the behaviour of Cl could serve as an analogue at landscape level.

### ***Thorium (Th), Uranium (U), Plutonium (Pu), Neptunium (Np) and Protactinium (Pa)***

Among the actinides, only Th and U occur naturally in the earth's crust in anything more than minute amounts. The actinide series displays less similarity in their chemical properties than the more homogenous lanthanide series. In some respects the elements of the lanthanide series are regarded as analogues to the corresponding elements of the actinide series. Chemically, U, Th and Pa show similarities with some of the transition metals. Th is most similar with the group 4 metals Ti, Zr and Hf. Pa is most similar to the group 5 metals V, Nb and Ta, whereas U is most similar to the group 6 metals Cr, Mo and W. Generally, Th is the actinide that has most matching properties with the lanthanides /Rayner-Canham 2000/. Th is similar to Pb and Po are easily adsorbed onto suspended particles and removed from the water column /Fisher et al. 1987/. Within the site investigation, stable isotopes of Th and U have been included together with a smaller number of measurements of the radiogenic isotopes (cf. Section 4.2).

## Detailed information regarding stable elements

### B.1 Detailed descriptions of input data

#### B.1.1 Estimated transports in streams

For three sampling points in the catchment of Norra Bassängen, annual export of elements via surface water discharge were estimated based on concomitant measurements of concentrations and discharge in streams during June 2004 to May 2006. Hydrochemical sampling and discharge measurements of the sampling sites PFM000069, PFM000070 and PFM000107 are described in detail in /Tröjbom et al. 2007, Johansson 2008/.

- Daily transports of major elements (Na, K, Ca, Mg, Cl, S, Br, F, Si, Sr, P, N) are calculated by multiplying total daily discharge with an linear interpolated concentration, and these daily estimations are summarised to yearly transports. Due to non-optimal spatial and temporal coverage of the runoff and hydrochemical sampling stations, interpolations and extrapolations were needed in order to make transport estimations possible /Tröjbom et al. 2007/.
- Transport estimations of trace elements (Ba, Cd, Ce, Co, Cr, Cu, Dy, Er, Eu, Fe, Gd, Hf, Hg, Ho, I, La, Li, Lu, Mn, Mo, Nd, Ni, Pb, Pr, Rb, Sb, Sm, Sr, Tb, Th, Tm, U, V, Y, Yb, Zn, Zr) were based on a correlation analysis between trace elements and major constituents. An estimated scale factor was multiplied with the yearly transport of the mostly correlated major constituent, thus giving a rough estimation of the transport of the trace element at interest. This estimation, which accordingly is based on several assumptions, may foremost be regarded an estimation of the order of magnitude of transports of trace elements in Forsmark watercourses /Tröjbom et al. 2007/.
- For a few elements, where estimations of transports were not available in /Tröjbom et al. 2007/, transports were estimated by multiplying the transport of an assumed analogue element with a scale factor derived from additional concentration measurements of lake water /Engdahl et al. 2008/. This applies to the following elements and the assumed analogue is shown within brackets: Ag (Cu) , As (Cu), Be (Mg), Bi (Cl), Ga (Si), Nb (V), Se (S), W (Mo), Al (Si), Sc (Y).

#### B.1.2 Atmospheric deposition

Data on atmospheric deposition originates from several sources described in detail in Table B-1. If available, site specific measurements of deposition are used to estimate the total yearly dry and wet deposition in the Forsmark area /Nilsson 2005/ and /Berg 2007/. If data from the Forsmark area are missing, deposition data from the Laxemar-Simpevarp site have been used instead (cf. /Tröjbom and Söderbäck 2006b/ and data from the SKB Sicada database not yet reported). In order to verify SKB site data, literature data from Sweden have been compiled and these data are also used in case of missing data from both the Forsmark and the Laxemar-Simpevarp sites.

#### B.1.3 Accumulation in lakes

The net accumulation per year in the lake sediments has been estimated by multiplying the net accumulation rate with site specific element concentrations of the sediments. The estimation is based on the assumptions that the average element concentration of the gyttja layer in Lake Eckarfjärden (c. 0 to 3 m) and Lake Stocksjön (c. 0 to 0.5 m) in the Forsmark area is assumed to represent the concentration in the net accumulation per year /Andersson 2010/. By selecting deeper layers and not only the top sediment layer, the estimation reflects the long term net accumulation. An average density of the gyttja layer is 72 kgDW/m<sup>3</sup> is assumed /Hedenström and Sohlenius 2008/. The net sedimentation rate (m/yr) is derived from the lake development model which describes the long term net accumulation rate, cf. /Brydsten 2004, 2006, Brydsten and Strömgren 2010, Hedenström and Risberg 2003, Andersson 2010/.

**Table B-1. Compilation of atmospheric deposition data from the Forsmark site, the Laxemar-Simpevarp site and Sweden (g/m<sup>2</sup>/yr). Data selected for mass balance calculations are marked in green and red colour denotes data below reporting limits.**

Element	SKB Forsmark data	SKB Laxemar-Simpevarp data	/Tyler and Olsson 2006/ <sup>A</sup>	/Land and Öhlander 2000/	Other ref 1 See notes below	Other ref 2 See notes below	Selected data
Ag			3.8E-06				3.8E-06
Al	1.8E-05	9.8E-03		3.84E-03			1.8E-05
As			5.3E-05	-1.20E-04	9.1E-05 <sup>B</sup>		5.3E-05
Ba		1.2E-03		2.82E-04			1.2E-03
Be			3.0E-06				3.0E-06
Bi			6.0E-06				6.0E-06
Br	-1.4E-03	-3.0E-01			1.7E-03 <sup>E</sup>		1.7E-03
Ca	1.7E-01	4.0E-01		2.40E-02	2.1E-01 <sup>D</sup>		1.7E-01
Cd		2.7E-05	4.5E-05	2.40E-05	4.3E-05 <sup>B</sup>		2.7E-05
Ce		4.8E-05	1.3E-04	1.20E-05			4.8E-05
Cl	5.1E-01	3.0E+00			1.3E-01 <sup>D</sup>		5.1E-01
Co		2.9E-05	2.9E-05	-1.80E-05	1.0E-05 <sup>B</sup>		2.9E-05
Cr		3.6E-04		8.40E-05	1.7E-04 <sup>B</sup>		3.6E-04
Cs		-6.0E-05					No data
Cu		2.5E-03	1.4E-03	2.22E-04	1.8E-03 <sup>B</sup>		2.5E-03
Dy		-1.0E-05	7.5E-06				7.5E-06
Er		-1.0E-05	3.8E-06				3.8E-06
Eu		-1.0E-05	9.8E-07				9.8E-07
F	-7.1E-02	-3.0E-01					No data
Fe	1.8E-02	1.9E-02		4.20E-03			1.8E-02
Ga			1.1E-05				1.1E-05
Gd		-1.0E-05	1.1E-05				1.1E-05
Ge			1.7E-06				1.7E-06
Hf		-1.0E-05					No data
Hg		-4.0E-06	1.6E-05	-1.20E-04			1.6E-05
Ho		-1.0E-05	1.5E-06				1.5E-06
I	-1.0E-03	4.9E-03			4.0E-04 <sup>G</sup>	5.9E-03 <sup>I</sup>	4.9E-03
K	9.8E-02	-8.0E-01		5.88E-02	1.8E-01 <sup>D</sup>		9.8E-02
La		2.9E-05	8.3E-05				2.9E-05
Li		2.0E-04	2.8E-05				2.0E-04
Lu		-1.0E-05	4.5E-07				4.5E-07
Mg	4.6E-02	2.9E-01		1.44E-02	1.1E-01 <sup>D</sup>		4.6E-02
Mn	-1.4E-02	7.8E-03		3.48E-04	1.9E-03 <sup>B</sup>	9.0E-03 <sup>D</sup>	7.8E-03
Mo		-1.0E-04	3.0E-05	-1.20E-04			3.0E-05
N					4.8E-01 <sup>D</sup>	3.6E-01 <sup>H</sup>	3.6E-01
Na	3.0E-01	7.7E-01		-3.60E-02	8.5E-01 <sup>D</sup>		3.0E-01
Nb			1.2E-05				1.2E-05
Nd		2.4E-05	5.6E-05				2.4E-05
Ni		8.4E-04		1.74E-04	1.5E-04 <sup>B</sup>		8.4E-04
P	1.2E-02	1.5E-02			3.0E-03 <sup>F</sup>		1.2E-02
Pb		4.2E-04		3.06E-04	1.2E-03 <sup>B</sup>		4.2E-04
Pr		-1.0E-05	1.7E-05				1.7E-05
Rb		6.4E-04					6.4E-04
S	2.8E-01	6.3E-01		1.74E-01	4.1E-01 <sup>D</sup>		2.8E-01
Sb		2.3E-04	3.2E-05		5.4E-05 <sup>C</sup>		5.4E-05
Sc		-1.0E-04	9.8E-06				9.8E-06
Se		2.1E-04	1.1E-04				2.1E-04
Si	8.4E-03	-6.0E-02		-6.00E-03			8.4E-03
Sm		-1.0E-05	1.1E-05				1.1E-05
Sr	5.1E-03	6.0E-03		6.00E-05			5.1E-03
Tb		-1.0E-05	1.5E-06				1.5E-06
Th		-4.0E-05	7.5E-06				7.5E-06
Tl		-2.0E-05					No data
Tm		-8.0E-06	5.3E-07				5.3E-07
U		4.3E-06	3.0E-06	-6.00E-06			4.3E-06
V		3.3E-04			3.1E-04 <sup>B</sup>		3.3E-04
W			1.1E-05				1.1E-05
Y		1.4E-05	4.3E-05				1.4E-05
Yb		-1.0E-05	3.0E-06	-1.20E-04			3.0E-06
Zn		3.6E-02	3.9E-03	3.78E-03	8.1E-03 <sup>B</sup>		8.1E-03
Zr		-6.0E-05	2.2E-05				2.2E-05

- A) Mid value of the reported range (mg/ha,year) during 6 vinter months recalculated to the unit g/m<sup>2</sup>,year. Estimations for the full year is probably not twice this figure due to lower deposition during the summer season. A factor of 1.5 was assumed to calculate yearly averages. Data from a beech stand in Håssleholm community, southern Sweden.
- B) IVL report B1420, mean for 1997–1999. Aspvreten at the east coast of Sweden.
- C) From IVL report B1473.
- D) From IVL report B1414. Mean for 98/99 and 99/00.
- E) Estimation based on the assumption that Br originates from sea water. Constant Cl/Br ratio estimated from Forsmark sea water.
- F) Phosphorus in precipitation. Results from measurements during the 1990's. IVL report B1442, Average Gårdssjön.
- G) From report No: 06819-REP-01200-10090 Nuclear waste management recommended biosphere model values for iodine. Ontario Power generation Minimum value.
- H) IVL 2003.
- I) From report No: 06819-REP-01200-10090 Nuclear waste management recommended biosphere model values for iodine. Ontario Power generation. Maximum value.
- J) SKB site deposition probably overestimated according to the "Sb" balance. IVL data from Aspvreten selected.
- K) Site specific deposition value probably 10 times elevated. Contamination? Data from IVL, Aspvreten was selected.

#### B.1.4 Area specific transports from other catchments

In order to validate the results of the mass balances for the Forsmark area, area specific transports from catchments nearby as well as from other parts of Sweden have been compiled for comparisons (cf. Table B-2). Data from four sites were selected:

- Forsmarksån, a medium sized watercourse that drains 375 km<sup>2</sup> of the inland area located close to the Forsmark site. Area specific transports have been calculated for 22 elements (Na, K, Mg, Ca, Cl, S, Si, Al, N, P, Fe, Mn, As, Cd, Co, Cr, Cu, Ni, Pb, V, Zn, Hg) /IMA 2009/. This catchment is comparable with the Forsmark site regarding soil composition and vegetation, but is much larger and perhaps represents a more mature landscape due to the slightly earlier withdrawal of the Baltic Sea /Andersson 2010/.
- The Laxemar-Simpevarp area which represents a landscape slightly larger than the Forsmark area, mainly representing average conditions in southern and middle Sweden. These data are comparable with the Forsmark dataset, both with respect to methods used and range of elements selected. See /Tröjbom et al. 2008/ further descriptions of the chemistry in the area and /Söderbäck and Lindborg 2009/ for a general description of the site.
- Another study represents the small catchment Storbergsbäcken (9.4 km<sup>2</sup>) located close to the Kalix River in northern Sweden /Land and Öhlander 2000/. This catchment is comparable in size with the Forsmark area but represents a different soil composition with e.g. no calcite in the regolith. Also atmospheric deposition differs for some elements due to the northern latitude. This study includes 17 elements (Na, Mg, Ca, Sr, Ba, Si, Al, Fe, Mn, Cd, Co, Cr, Ni, Zn, Mo, U, Ce).
- Svartberget/Krycklan/Svartberget, which is a small (67 km<sup>2</sup>), thoroughly studied catchment located in northern Sweden. This catchment, where parts are located both above and below the highest coast-line, is dominated by forest land but also contains open/arable lands as well as lakes and wetlands. This study includes 29 elements (Na, K, Rb, Mg, Ca, Sr, Ba, Cl, S, Se, Si, Al, Fe, Mn, As, Cd, Co, Cr, Cu, Ni, Pb, Zn, Sb, Th, U, La, Sc, Y, Zr), some described in /Björkvald et al. 2008, Cory et al. 2006, Bergknut et al. 2009/.

#### B.1.5 Estimated transports via groundwater discharge

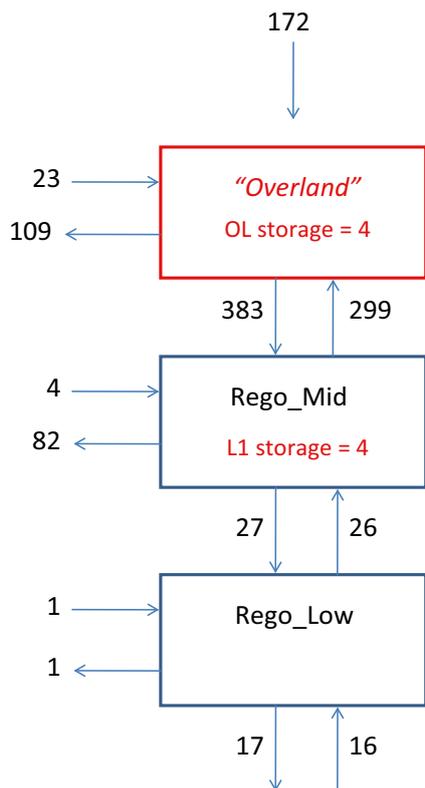
The content of elements in the groundwater is not estimated explicitly, but is to a varying degree included in the regolith pools (cf. 5.3). In order to estimate the potential of discharging groundwater from deeper levels of the regolith to influence concentrations observed in the surface water, a hypothetical mass transport from the T\_RegoLow layer was estimated. By multiplying a typical concentration of the T\_RegoLow layer by the modelled average groundwater flux from this level, a hypothetical mass transport, potentially reaching the surface, was obtained.

Typical element concentrations of the groundwater at deeper levels of the regolith were derived from the classification by the Ion Source Model (cf. /Tröjbom et al. 2007/). It was assumed that water types characterised by low marine contents in combination with a "mature" groundwater composition represent the modified groundwater type (Na-HCO<sub>3</sub>) found in the deeper parts of the regolith as well as in the upper parts of the bedrock (encircled in red in Figure B-1).

**Table B-2. Compilation of area specific transports from the Forsmark area, Forsmarksån, the Laxemar-Simpevarp area, Storbergsbäcken and Svartberget/Krycklan.**

Area specific transports (g/m <sup>2</sup> /year)					
	Forsmark	Forsmarksån	Laxemar-Simpevarp	Storbergsbäcken	Krycklan
Na	7.0E-01	1.4E+00	1.6E+00	1.7E-01	4.7E-01
K	4.4E-01	3.9E-01	2.7E-01		2.7E-01
Rb	3.9E-04		5.4E-04		1.2E-04
Li	4.5E-04		4.7E-04		
Mg	9.6E-01	5.7E-01	5.2E-01	1.2E-01	3.3E-01
Be	7.0E-07				
Ca	7.9E+00	8.8E+00	2.2E+00	5.2E-01	8.1E-01
Sr	1.3E-02		1.1E-02	2.2E-03	4.1E-03
Ba	3.3E-03		3.4E-03	5.9E-04	3.4E-03
Cl	6.5E-01	1.6E+00	1.9E+00		3.1E-01
Br	6.3E-03		2.5E-02		
I	1.2E-03		2.8E-03		
C	6.8E+00				
S	8.7E-01	1.5E+00	1.2E+00		3.9E-01
Se	1.7E-05				8.7E-06
Si	4.4E-01	8.5E-01	1.9E+00	5.7E-01	1.5E+00
Al	1.2E-02	3.3E-02	9.8E-02	1.6E-02	2.1E-01
Ga	1.3E-06				
N	1.5E-01	3.1E-01	3.0E-01		
P	1.8E-03	4.2E-03	8.7E-03		
Fe	2.0E-02	8.8E-02	2.4E-01	1.4E-01	5.1E-01
Mn	6.1E-03	8.0E-03		4.0E-03	1.4E-02
Ag	2.8E-07				
As	3.4E-05	1.2E-04			4.9E-05
Bi	4.5E-07				
Cd	1.2E-06	4.1E-06	1.4E-05	1.5E-05	9.9E-07
Co	8.3E-06	3.4E-05	1.7E-04	1.6E-05	2.0E-05
Cr	7.3E-05	1.2E-04	2.5E-04	7.0E-07	3.0E-05
Cu	1.4E-04	5.4E-04	7.1E-04		6.8E-05
Ni	7.3E-05	2.8E-04	8.1E-04	8.6E-05	9.1E-05
Pb	1.4E-05	5.0E-05			1.7E-05
V	3.4E-05	1.1E-04	3.1E-04		
Zn	4.0E-04	1.2E-03	2.1E-03	2.7E-03	4.0E-04
Hg	1.1E-06	1.0E-06	7.8E-07		
Mo	7.7E-05		1.5E-04	7.0E-05	
Nb	1.1E-06				
Sb	1.3E-05				1.8E-06
Th	2.7E-06		2.8E-05		4.8E-06
U	3.7E-04		1.5E-04	9.6E-06	6.5E-06
W	2.6E-06				
Ce	1.4E-05		5.8E-04	1.0E-04	
Dy	2.9E-06		3.8E-05		
Er	2.2E-06		2.5E-05		
Eu	5.5E-07		9.2E-06		
Gd	3.0E-06		5.0E-05		
Ho	7.2E-07		7.9E-06		
La	1.2E-05		3.1E-04		6.5E-05
Lu	8.5E-07		4.2E-06		
Nd	1.3E-05		3.3E-04		
Pr	3.1E-06		8.3E-05		
Sc	1.8E-05		1.7E-05		5.6E-05
Sm	2.9E-06		5.8E-05		
Tb	1.9E-06		1.1E-05		
Tm	5.3E-07		3.6E-06		
Y	2.5E-05		2.8E-04		4.0E-05
Yb	2.4E-06		2.6E-05		
Zr	4.2E-05		5.0E-04		5.7E-03





**Figure B-2.** Yearly water balance for the catchment of Lake Bolundsfjärden (all figures represent mm/yr). As a yearly average, approximately 1% of the total surface water discharge originates from the deeper levels of the terrestrial regolith according to the water balance.

Some elements have been sampled at several occasions with high accuracy, whereas others are impaired with large uncertainties regarding representativity, measurement accuracy or extrapolations. In case of elements strongly associated with particles, concentrations measured could for example be underestimated if high flow episodes are omitted, leading to underestimated representative concentrations of these elements. On the other hand, if concentrations fall below reporting limits, concentrations could be overestimated if extrapolation techniques are used. In case of missing site specific measurements, the use of comparable data from other sites implies varying uncertainties that could be hard to evaluate, e.g. atmospheric deposition.

### B.2.1 Transport estimations

Transport estimations used to estimate export via watercourses rely on both measurements of concentrations and water flow. Ideally, there are simultaneous measurements of concentrations and water flow at representative sampling interval behind these estimations. In practice, this is seldom fulfilled and in case of transports estimated for the Forsmark area, extrapolations in both time and space were needed as described in /Tröjbom et al. 2007/.

The most accurate transport estimations were probably made for the major elements which were sampled more frequent than most trace elements. Based on a correlation analysis among trace elements and major constituents an estimated scale factor was multiplied with the yearly transport of the mostly correlated major constituent, thus giving a coarse estimation of the transport of the trace element. The rationale for using this extrapolation technique is that the more frequent sampling interval of the major elements has the possibility to catch short episodes which e.g. could be important for trace elements associated to particulate or organic matter. For a few elements transports were estimated by proportionalisation based on measured concentrations in lakes (cf. Appendix B, Section B.1.1 for a detailed description of specific elements and /Tröjbom et al. 2007/ for the underlying assumptions).

This means that transport estimations of most major elements are probably relatively accurate, especially if concentrations show little variation over the year, whereas estimated transports of trace elements could be biased if the underlying assumption of the correlation to a major element is invalid (or only partially explains the variation of the concentrations of the trace element). The use of lake data for proportionalisation of transports also implies uncertainties if the ratios of the selected elements are not representative for concentrations in streaming water.

### **B.2.2 Atmospheric deposition**

In the mass balances, data on atmospheric deposition have been compiled from several sources. As could be seen in Table B-1, there is sometimes a substantial variation among the estimations for an element. Insufficient measurement accuracy leading to data below reporting limits is common in these dilute water samples.

When site specific deposition data have been used, representing the bulk deposition measured in field in the Forsmark area, these measurements represent the sum dry and wet deposition. If an element is predominantly trapped by vegetation, e.g. tree crowns, the total deposition input to the landscape could be underestimated compared to these bulk estimations.

When using data from the Laxemar-Simpevarp area deposition of long transported air borne pollutants, e.g. heavy metals, are probably overestimated. There is a general southwest to northeast gradient in Sweden implicating that the use of Laxemar-Simpevarp data overestimates atmospheric deposition in the Forsmark area.

For several trace elements, for which there were no data available from the Forsmark or Laxemar-Simpevarp sites, atmospheric deposition data from either IVL stations in eastern Sweden, or Hässleholm community in southern Sweden was used. The latter station is located inland possibly less influenced from marine sources, but with possibly higher deposition of long range transported pollutants from Europe.

In case of great discrepancies between the actual atmospheric deposition in the Forsmark area and these sites, the mass balance is more or less biased. This seems to be the case for e.g. Nb and Zr, two presumed geogenic elements where atmospheric deposition is the main source according to the mass balance, where atmospheric deposition data from the Hässleholm site in southern Sweden are used. These discrepancies could give an indication of the possibly great uncertainties attributed to the atmospheric deposition estimates. Also REE data indicates that there are substantial uncertainties associated to atmospheric deposition for this group of elements (cf. Appendix B, Section B.2.8).

### **B.2.3 Estimations of accumulation in lake sediments**

The yearly accumulation in lakes is estimated based on modelled, lake specific net sedimentation rates and measured concentrations of lake sediments. The net sedimentation rate is estimated from the Forsmark lake development model described in /Brydsten and Strömgren 2010, Brydsten 2004/ and lake specific data are described in /Nordén et al. 2008, Andersson 2010/.

A significant uncertainty associated to this estimation is the coupling of the net sedimentation rate and a concentration representative for the long term sedimentation process. The exclusion of the topmost layers assures that the estimations of the accumulation of elements attributed to resuspension and cycling are not overestimated /Nordén et al. 2008, Andersson 2010/.

### **B.2.4 Specific discharge reference data**

The compilation of area specific transports as a reference to the Forsmark data contains data from several sources with varying comparability. Data from the Laxemar-Simpevarp area are determined by similar methods as Forsmark data regarding sampling, chemical analyses, water discharge measurements and calculation. It could be presumed that there might be systematic differences between SKB and other data sources due to e.g. methodological differences, but this topic has not been investigated further in this study.

### **B.2.5 Pool estimations of the limnic system**

Site-specific measurements are available for all ecosystem components in the Forsmark lakes and confidence in the pool estimates are regarded as high. The estimated pools of different elements in particulate matter in lake water, in biota, and in sediments are associated with some uncertainties with regard to absolute numbers, but the orders of magnitude of the estimates are most probably correct /Nordén et al. 2008, Andersson 2010/.

The estimated pools of different elements dissolved in lake water are based on site specific data from several lakes and several years and can be considered reliable. Data on the pool of elements dissolved in water are available for 18 sampling sites for the period March 2002–June 2004, and for 9 of these sampling sites for the period July 2004–June 2006 as well, giving these estimates a relatively good resolution in both time and space. The pools of particulate carbon, nitrogen, and phosphorus are available for the same time period and sampling stations as the elements in the dissolved component and can be regarded as reliable estimates with good resolution in time and space. The estimated pools of other elements in particulate matter are, however, based on a single sampling performed in spring 2008. Using the results from this sampling to estimate the mean annual pool of different elements in particulate matter naturally entails relatively high uncertainty in the estimates. Moreover, the particulate fraction also includes the living biota. No estimate of living biota was made at the same occasion as suspended material and the living biota has not been subtracted from the particulate component in the result and thus the particulate fraction may be somewhat overestimated. However, as the data are site-specific they provide high confidence in the order of magnitude of the pools of elements in the particulate component.

The largest uncertainty in pools of elements in the biotic component stems from the lack of data on the chemical composition of bacteria for all elements except phosphorus, nitrogen, sulphur and carbon. This of course leads to uncertainties in the distribution of the elements within the biotic component. A relatively large share of biotic carbon, nitrogen, phosphorus and sulphur is present in bacteria. This is particularly true of phosphorus, as phosphorus concentrations in bacteria are higher than in other organisms. The concentrations of most elements in bacteria will probably be proportionate to the concentration of carbon, sulphur and nitrogen, but the possibility cannot be excluded that some trace elements will, like phosphorus, be highly concentrated in bacteria. Either way, it is clear that most elements are probably present in consumers to a considerably greater degree than presented. In the case of other organisms, the elemental composition data for biotic pools is a reliable estimate, since it is mainly site-specific. There are relatively few replicates, which cause some uncertainties, but the available replicates show small discrepancies for most functional groups.

The pools of elements in the sediment component are highly dependent on the estimated sediment volume. The volumes are based on average values of layer thickness from lake specific stratigraphical data measured in field. The representativity of these average values is dependent on the number of sampling points used and their spatial resolution. The number of sampling points in each lake was adjusted to the size of the lake basin (fewer points in smaller lakes, see /Hedenström 2004/). In the calculation of average sediment thickness, data points lacking organic sediments were excluded, probably resulting in overestimated sediment volumes. The elemental composition data for sediment is also site-specific with data from two lakes (Lake Eckarfjärden and Lake Stocksjön). It was assumed that the chemical composition of different sediment layers does not differ between lakes. Instead, a mean value of the different samples from the same sediment layer was calculated and used as a representative value for that sediment type in the area.

### **B.2.6 Pool estimations of the terrestrial system**

The pools have been estimated from a fairly small number of element estimates representing six ecosystems. This means that one or a few misleading samples would have a great impact on the final result. Generally many elements in the consumer compartment were found to be below the detection limit, which means that uncertainties are larger for this pool. See /Löfgren 2008, Löfgren 2010/ for further descriptions of assumptions and methods behind the terrestrial pool estimations.

## B.2.7 Spatial and temporal representativity of the mass balance model

The mass balance model presented in Section 5.1.1 describes average conditions during a year. When possible, input data were chosen to represent the period 2004–2006 which is the period available for transport estimations in the Forsmark area.

Data on atmospheric deposition represent other periods than 2004–2006 depending on element. Some data represent a mean of a time series reflecting seasonal fluctuations, whereas other represent only one sampling occasion. The temporal representativity of the model result is therefore more uncertain for some elements (especially some trace elements, cf. Table B-1). The estimated accumulation in lakes perhaps show less temporal uncertainties due to the integrating effects of the sediment sampling, although it is possible that the actual retention could vary between years due to e.g. climatic reasons and hence influence the uncertainties of the overall mass balance. In general, the short time series implicates that variations between years might have significant influence on the model results. Precipitation and hence mass transports show significant variation between years (cf. /Tröjbom et al. 2007/).

The spatial representativity of the mass balance model result could to some extent be quantified via the calibration result. This measure reflects how well the spatial variation was modelled among the subcatchments in the Forsmark area (cf. Section 5.1.1).

The calibration result is summarised in Table B-3, where the Nash-Sutcliffe model efficiency coefficient is compiled. This coefficient ranges from  $-\infty$  to 1. A value of 1 corresponds to a perfect match between modelled and observed data, 0 indicates that the predictions are as accurate as the mean of the observed data, whereas a value less than 0 indicates a more or less poor match. Most elements show an efficiency coefficient close to 1 which means that the variation among the three calibration points is modelled satisfactorily. This seemingly good match is a consequence of increasing transports when catchment size increases, and not too much attention should be laid on this apparently good match. If the sources of an element are uniformly distributed over the area, there will be

**Table B-3. Agreement between modelled and observed transports at three points in the catchment of Norra Bassängen. The Nash and Sutcliffe coefficient reflects the efficiency of the model. This coefficient ranges from  $-\infty$  to 1. A value of 1 corresponds to a perfect match between modelled and observed data, 0 indicates that the predictions are as accurate as the mean of the observed data, whereas a value less than 0 indicates a more or less poor match. The mass balance constant describes the annual loss (positive value), or the annual accumulation (negative value) in the terrestrial system, determined for each element during calibration.**

Element	Nash and Sutcliffe coefficient $-\infty$ to 1	Mass balance constant (ASP_const) g/m <sup>2</sup> /year	Element	Nash and Sutcliffe coefficient $-\infty$ to 1	Mass balance constant (ASP_const) g/m <sup>2</sup> /year	Element	Nash and Sutcliffe coefficient $-\infty$ to 1	Mass balance constant (ASP_const) g/m <sup>2</sup> /year
Na	-0.36	4.3E-01	Fe	0.98	1.1E-02	W	0.87	-6.2E-06
K	0.91	3.8E-01	Mn	0.99	-1.5E-03	Ce	-0.49	1.2E-05
Rb	0.92	-2.4E-04	Ag	-2,528	1.6E-05	Dy	0.79	-1.2E-06
Li	0.92	2.9E-04	As	0.97	-1.6E-05	Er	0.83	8.0E-07
Mg	0.88	9.9E-01	Bi	-248	-6.0E-06	Eu	0.71	4.8E-08
Be	0.89	-2.1E-06	Cd	0.89	-2.2E-05	Gd	0.98	-6.8E-06
Ca	0.97	8.4E+00	Co	0.99	-1.9E-05	Ho	0.83	8.6E-09
Sr	0.93	9.0E-03	Cr	0.98	-2.9E-04	La	0.53	4.9E-06
Ba	0.97	2.4E-03	Cu	0.36	-2.5E-03	Lu	0.77	7.6E-07
Cl	-0.49	1.5E-01	Ni	0.88	-8.1E-04	Nd	0.54	1.2E-05
Br	-0.34	5.0E-03	Pb	0.49	-4.1E-04	Pr	0.20	-6.3E-06
I	0.96	-4.1E-03	V	0.97	-3.0E-04	Sc	0.99	1.1E-05
S	0.90	6.5E-01	Zn	0.32	-8.2E-03	Sm	0.80	-4.4E-06
Se	0.82	-2.1E-04	Hg	-25	-1.6E-05	Tb	0.79	7.8E-07
Si	0.66	5.5E-01	Mo	0.91	6.7E-05	Tm	0.74	3.4E-07
Al	0.83	2.0E-02	Nb	-0.38	-7.7E-06	Y	0.97	2.5E-05
Ga	0.25	-1.0E-05	Sb	0.93	-4.4E-05	Yb	0.93	1.2E-06
N	0.99	-2.0E-01	Th	0.68	-9.7E-07	Zr	0.86	4.9E-05
P	0.89	-1.1E-02	U	0.98	4.4E-04			

necessity be little spatial variation and the efficiency coefficient will consequently indicate a good match. Some marine ions however, e.g. Na, Cl, and Br, show an efficiency coefficient less than 0 which means that there is a poor match. This discrepancy is mainly caused by sea water intrusions in Lake Bolundsfjärden adding significant amounts of marine ions to the lake, not accounted for in this modelling. This source was in practice excluded from the mass balance by omitting the calibration point representing export from Lake Bolundsfjärden, but is still included in the calculation of the calibration result (for Na, Cl, and Br). These mass balances thus represent the conditions upstream Lake Bolundsfjärden which were though to be most representative at landscape scale, cf. estimations in /Tröjbom et al. 2007/.

Some major elements, e.g. Ca and Sr mainly originating from calcite dissolution in the regolith, show good agreement according to the calibration result which reflects the satisfactory modelling of the spatial variation of these elements. Also many metals, mainly originating from atmospheric deposition, show a similarly good agreement. The REE group in the rightmost column shows varying agreement, despite the supposed uniform properties and sources for these elements. The varying sign of the mass balance constant, reflecting if the terrestrial system is a sink or a source, also suggests that uncertainties are considerable for REE (cf. further discussion in Appendix B, Section B.2.8).

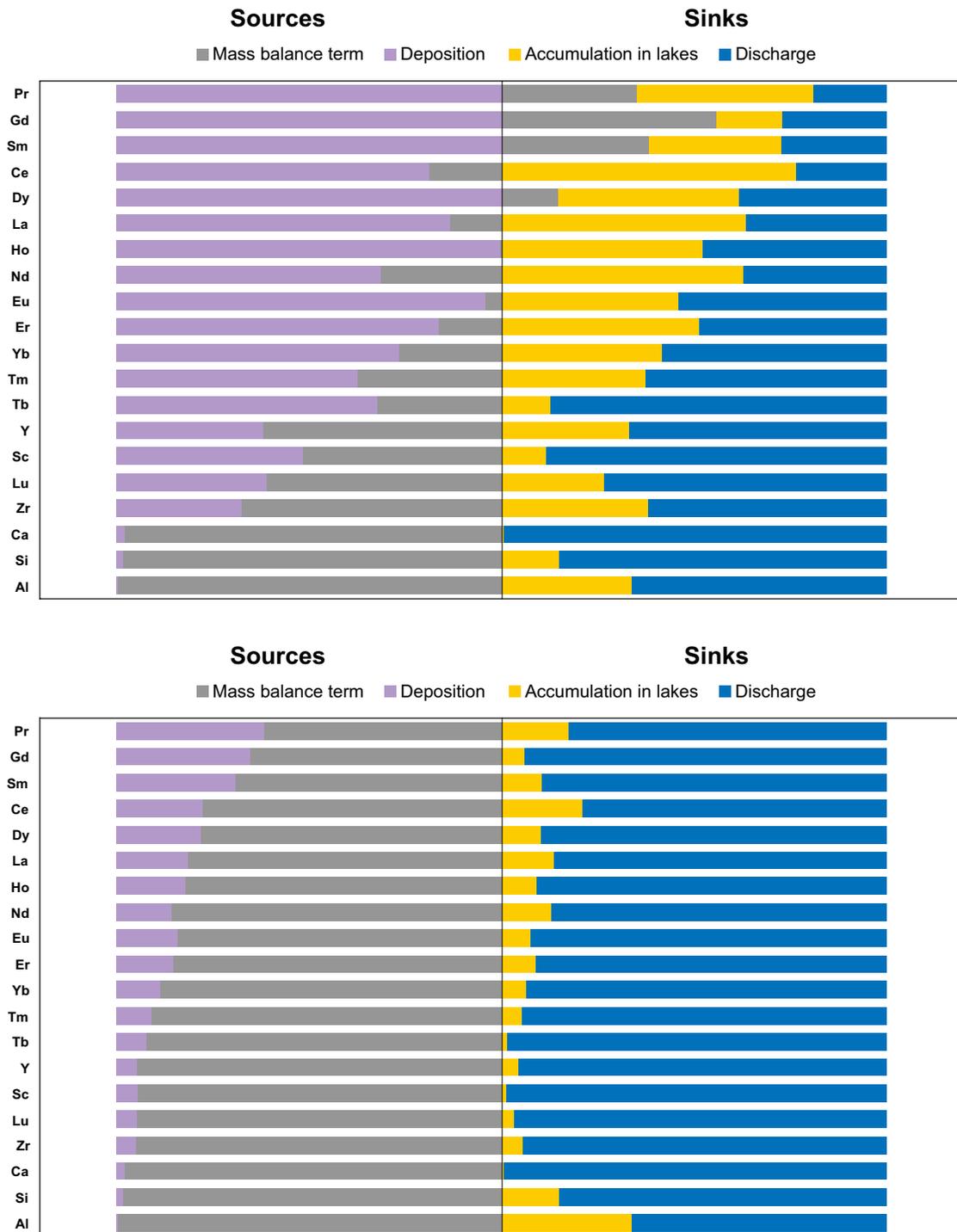
### **B.2.8 Overall uncertainties in flux estimations – REE example**

Mass balances and flux estimations are associated with highly varying uncertainties regarding the confidence in input data. This is particularly well illustrated by the rare earth element group for which the mass balances indicate very different behaviour in the landscape. Figure B-3, which includes all lanthanides together with Ca, Al, and Si, could serve as an example of the uncertainties associated with the mass balance calculations. The rare earth elements (abbreviated REE: comprising Sc, Y, and the lanthanides) usually show similar behaviour in all types of site data and concentrations are often closely correlated in the Forsmark area /Tröjbom and Söderbäck 2006a/. This behaviour could be interpreted as if all these elements originate from common sources and that they are influenced in a similar way by the same processes.

According to the balances in the upper panel of Figure B-3, sources of REE range from mostly atmospheric (Pr, Gd and Sm) to mineral sources (Sc, Lu and Zr). Sc and Zr are elements, that similar to Si and Al, usually have geogenic origin, and it could be expected that the relative distribution patterns for all REE should resemble the patterns for e.g. Sc and Zr. This pronounced discrepancy among the REE is most probably an artefact and there could be several explanations to the overall uncertainty of this group of elements:

- There are possibly considerable uncertainties associated with REE deposition data. There is no reliable site specific deposition data available and the atmospheric input could be overestimated in the Forsmark area.
- Some REE analyses are close to or below reporting limits. This is the case for Sc, Eu, Tb, Ho, Tm and Lu in fresh surface waters. The method of replacing values below the reporting limit with a value corresponding to half the reporting limit leads to an overestimated export via surface water if the actual value is significantly lower than the reporting limit. In such cases, the importance of atmospheric deposition is underestimated in the relative distribution of sources and sinks. This, however, further strengthens the relative importance of atmospheric deposition among the sources.
- Mass transports in discharge may also be underestimated due to difficulties in correctly capturing the particulate fraction. If a large fraction of these elements is particle bound and predominantly transported during high flow episodes, the sporadic sampling interval as well as filtration during sample preparation could contribute to an underestimation of the total yearly transports, and in turn increase the relative importance of atmospheric deposition. The importance of understanding particulate transport processes when evaluation REE elements could also be valid for other trace elements, for example metals as Pb and Th.
- The varying relative importance of atmospheric deposition in the REE balances could reflect varying anthropogenic emissions of these elements. The production and use of REE have increased dramatically during the last decades and it could not be ruled out that anthropogenic emissions might have an impact on the measured atmospheric deposition patterns of REE.

The mass balances for REE in the upper panel of Figure B-3 are most likely more or less unrealistic. The overall impression is that an unexpectedly high proportion of the REE input (including Sc and Y, as well as the related element Zr), seem to be explained by atmospheric deposition rather than weathering of minerals in the regolith and bedrock. If export via surface water is generally increased ten times, the resulting mass balances are shown in the lower panel of Figure B-3. This scenario does not rule out atmospheric deposition as an important source for REE, but leads to a more probable overall pattern with weathering as an important source, similar to geogenic elements as Al and Si. See discussion on Section 5.5 where uncertainties regarding atmospheric deposition are discussed.



**Figure B-3.** Relative comparisons of element fluxes at landscape level in the Forsmark area for rare earth elements and Ca, Si and Al. This figure includes two scenarios: the upper panel show balances based on original data and the lower panel a scenario where the export via surface water discharge has been increased ten times (not for Ca, Si and Al). See Figure 5-4 for further descriptions of this figure.

### B.3 Tabulated fluxes and pools from the terrestrial and limnic systems

Tabulated fluxes and pools from the terrestrial and limnic ecosystem described in Chapter 5.

Table B-4. Fluxes and pools in the terrestrial system. Fluxes are given in g/year and pools in g.

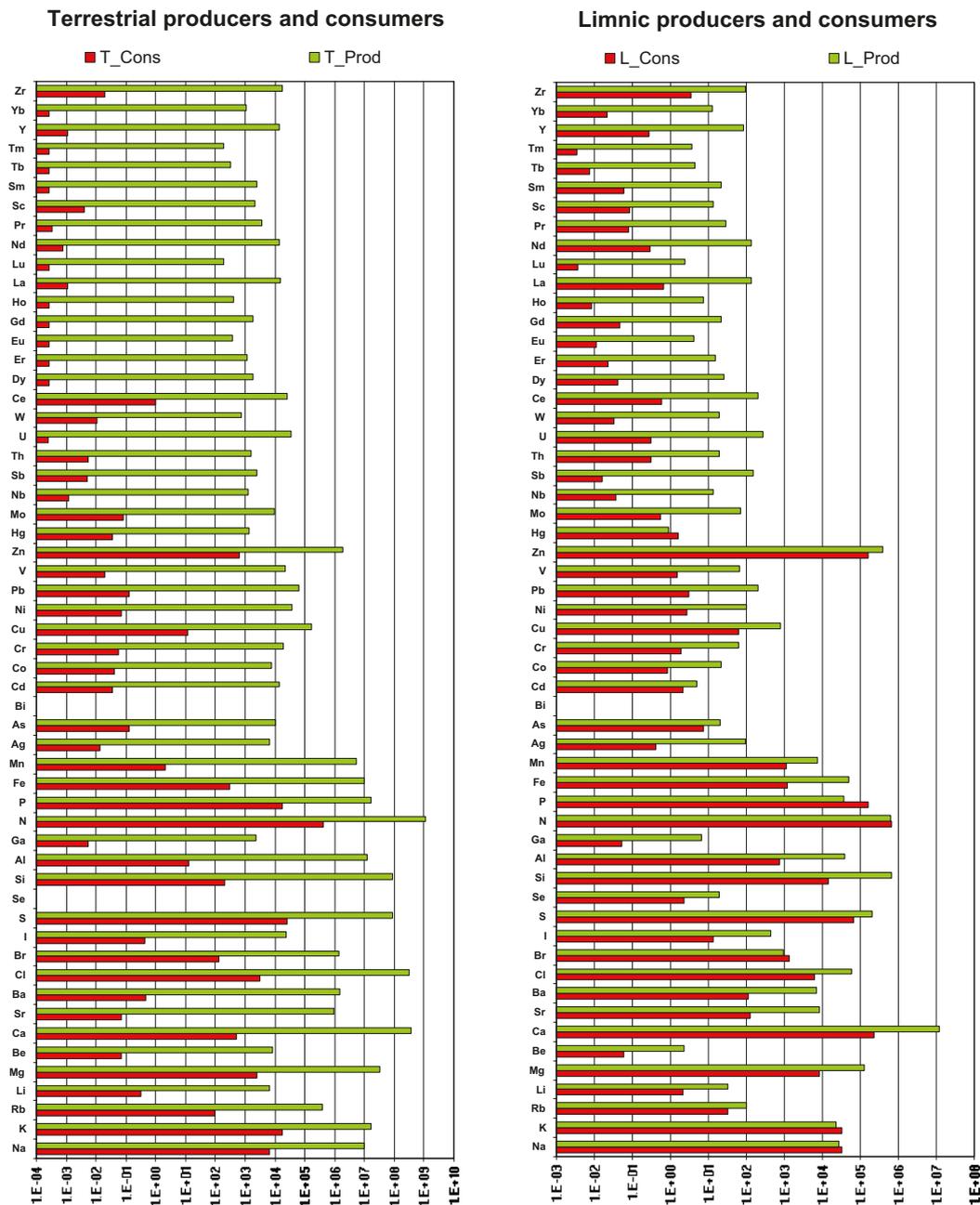
Terrestrial ecosystem									
Element	Fluxes				Pools				
	T <sub>InAir</sub>	Mass balance term T <sub>Acc/Wea</sub>	Deep discharge T <sub>OutRegoLow</sub>	Discharge T <sub>OutWater</sub>	Producers T <sub>Prod</sub>	Consumers T <sub>Cons</sub>	Upper regolith T <sub>RegoUp</sub>	Lower regolith T <sub>RegoLow</sub>	
Na	2.38E+06	3.42E+06	6.83E+05	5.80E+06	9.88E+06	6.78E+03	1.86E+08	5.95E+09	
K	7.72E+05	2.99E+06	1.44E+05	3.76E+06	1.62E+07	1.73E+04	5.80E+07	2.01E+08	
Rb	5.07E+03	-1.89E+03	2.56E+01	3.18E+03	3.87E+05	9.83E+01	8.50E+05	3.24E+07	
Li	1.54E+03	2.30E+03	1.59E+02	3.85E+03	6.77E+03	3.23E-01	8.28E+04	5.21E+06	
Mg	3.62E+05	7.80E+06	2.89E+05	8.16E+06	3.15E+07	2.42E+03	2.00E+08	1.41E+09	
Be	2.37E+01	-1.66E+01	0.00E+00	7.06E+00	8.47E+03	6.97E-02	1.26E+04	5.85E+05	
Ca	1.32E+06	6.63E+07	1.66E+06	6.77E+07	3.70E+08	4.97E+02	2.17E+09	6.17E+09	
Sr	3.98E+04	7.08E+04	4.77E+03	1.11E+05	9.18E+05	7.07E-02	5.49E+06	5.13E+07	
Ba	9.81E+03	1.86E+04	7.32E+02	2.84E+04	1.46E+06	4.57E-01	9.62E+06	1.80E+08	
Cl	4.06E+06	1.19E+06	7.18E+05	5.24E+06	3.22E+08	3.13E+03	8.22E+07	1.31E+07	
Br	1.36E+04	3.93E+04	3.02E+03	5.29E+04	1.38E+06	1.33E+02	5.90E+06	6.25E+05	
I	3.90E+04	-3.23E+04	7.80E+01	6.69E+03	2.45E+04	4.42E-01	1.66E+05	3.56E+05	
C	0.00E+00	6.11E+07	1.13E+06	6.11E+07	4.65E+10	1.60E+06	7.20E+10	3.55E+10	
S	2.21E+06	5.09E+06	4.02E+05	7.30E+06	8.65E+07	2.52E+04	6.49E+08	3.01E+08	
Se	1.66E+03	-1.65E+03	0.00E+00	1.34E+01	5.00E-10	5.00E-10	5.00E-10	5.00E-10	
Si	6.62E+04	4.36E+06	1.07E+05	4.42E+06	8.56E+07	2.00E+02	2.92E+09	1.04E+11	
Al	1.39E+02	1.60E+05	1.40E+02	1.60E+05	1.20E+07	1.25E+01	5.50E+08	1.99E+10	
Ga	8.88E+01	-8.02E+01	0.00E+00	8.52E+00	2.27E+03	5.23E-03	1.22E+05	9.78E+05	
N	2.84E+06	-1.60E+06	1.10E+04	1.24E+06	1.10E+09	4.29E+05	2.36E+09	4.09E+09	
P	9.59E+04	-8.44E+04	5.48E+02	1.15E+04	1.62E+07	1.73E+04	5.80E+07	2.01E+08	
Fe	1.42E+05	9.03E+04	1.91E+04	2.33E+05	9.55E+06	3.00E+02	2.02E+08	6.04E+09	
Mn	6.14E+04	-1.21E+04	2.83E+03	4.94E+04	5.24E+06	2.08E+00	1.52E+07	1.08E+08	
Ag	2.96E+01	1.26E+02	0.00E+00	1.55E+02	6.65E+03	1.38E-02	2.74E+05	2.17E+06	
As	4.14E+02	-1.30E+02	9.38E+00	2.84E+02	1.02E+04	1.26E-01	5.87E+04	7.94E+05	
Bi	4.73E+01	-4.74E+01	0.00E+00	-9.11E-02	5.00E-10	5.00E-10	5.00E-10	5.00E-10	
Cd	1.71E+02	-1.70E+02	9.58E-02	7.36E-01	1.36E+04	3.43E-02	1.44E+04	1.23E+05	
Co	2.26E+02	-1.53E+02	1.76E+00	7.26E+01	7.62E+03	4.20E-02	6.76E+04	1.90E+06	
Cr	2.81E+03	-2.31E+03	1.14E+00	5.02E+02	1.96E+04	5.46E-02	5.07E+05	1.17E+07	
Cu	1.95E+04	-1.98E+04	5.73E+00	-2.23E+02	1.69E+05	1.19E+01	2.67E+05	6.41E+06	
Ni	6.59E+03	-6.40E+03	1.13E+01	1.85E+02	3.73E+04	7.19E-02	2.04E+05	2.33E+06	
Pb	3.30E+03	-3.23E+03	7.50E-01	6.95E+01	6.31E+04	1.29E-01	1.88E+06	1.06E+07	
V	2.59E+03	-2.40E+03	5.43E+00	1.88E+02	2.26E+04	1.93E-02	4.48E+05	1.10E+07	
Zn	6.39E+04	-6.46E+04	1.10E+01	-7.89E+02	1.88E+06	6.53E+02	4.24E+06	1.75E+07	
Hg	1.24E+02	-1.24E+02	-2.74E-02	8.30E-13	1.37E+03	3.51E-02	3.87E+03	2.41E+04	
Mo	2.37E+02	5.31E+02	2.50E+01	7.67E+02	9.91E+03	8.29E-02	7.01E+04	4.61E+05	
Nb	9.47E+01	-6.08E+01	1.38E+00	3.39E+01	1.26E+03	1.20E-03	6.00E+04	2.07E+06	
Sb	4.26E+02	-3.49E+02	9.03E-01	7.75E+01	2.39E+03	4.89E-03	4.90E+04	1.17E+05	
Th	5.92E+01	-7.68E+00	-2.74E-01	5.15E+01	1.54E+03	5.51E-03	3.01E+04	1.86E+06	
U	3.40E+01	3.44E+03	9.88E+01	3.47E+03	3.48E+04	2.47E-04	2.47E+05	1.79E+06	
W	8.28E+01	-4.92E+01	0.00E+00	3.36E+01	7.45E+02	1.08E-02	4.41E+04	9.12E+05	
Ce	3.77E+02	9.47E+01	1.15E+01	4.72E+02	2.59E+04	0.00E+00	4.73E+05	1.81E+07	
Dy	5.92E+01	-9.42E+00	3.15E+00	4.98E+01	1.87E+03	2.75E-04	8.44E+04	1.36E+06	
Er	2.96E+01	6.29E+00	2.16E+00	3.59E+01	1.19E+03	2.75E-04	1.30E+04	3.17E+05	
Eu	7.69E+00	3.81E-01	3.46E-01	8.07E+00	3.84E+02	2.75E-04	1.79E+04	6.50E+05	
Gd	8.88E+01	-5.38E+01	3.81E+00	3.49E+01	1.85E+03	2.75E-04	3.19E+04	1.28E+06	
Ho	1.18E+01	6.79E-02	7.62E-01	1.19E+01	4.16E+02	2.75E-04	7.87E+03	2.53E+05	
La	2.30E+02	3.83E+01	2.01E+01	2.69E+02	1.49E+04	1.15E-03	2.42E+05	8.68E+06	
Lu	3.55E+00	6.03E+00	3.20E-01	9.58E+00	1.91E+02	2.75E-04	3.07E+03	8.69E+04	
Nd	1.86E+02	9.16E+01	1.84E+01	2.77E+02	1.39E+04	7.90E-04	2.23E+05	7.75E+06	
Pr	1.30E+02	-4.97E+01	4.37E+00	8.05E+01	3.60E+03	3.24E-04	5.93E+04	2.09E+06	
Sc	7.69E+01	8.98E+01	7.02E-01	1.67E+02	2.06E+03	3.91E-03	6.71E+04	2.17E+06	
Sm	8.28E+01	-3.44E+01	2.98E+00	4.84E+01	2.43E+03	2.75E-04	3.88E+04	1.51E+06	
Tb	1.18E+01	6.14E+00	4.43E-01	1.80E+01	3.29E+02	2.75E-04	5.31E+03	2.15E+05	
Tm	4.14E+00	2.69E+00	3.26E-01	6.84E+00	1.92E+02	2.75E-04	3.76E+03	1.17E+05	
Y	1.12E+02	1.99E+02	3.47E+01	3.11E+02	1.37E+04	1.13E-03	2.44E+05	7.73E+06	
Yb	2.37E+01	9.31E+00	1.93E+00	3.30E+01	1.10E+03	2.75E-04	2.17E+04	6.25E+05	
Zr	1.72E+02	3.89E+02	1.02E+01	5.60E+02	1.70E+04	2.01E-02	1.47E+06	5.23E+07	

Table B-5. Fluxes and pools in the limnic system. Fluxes are given in g/year and pools in g.

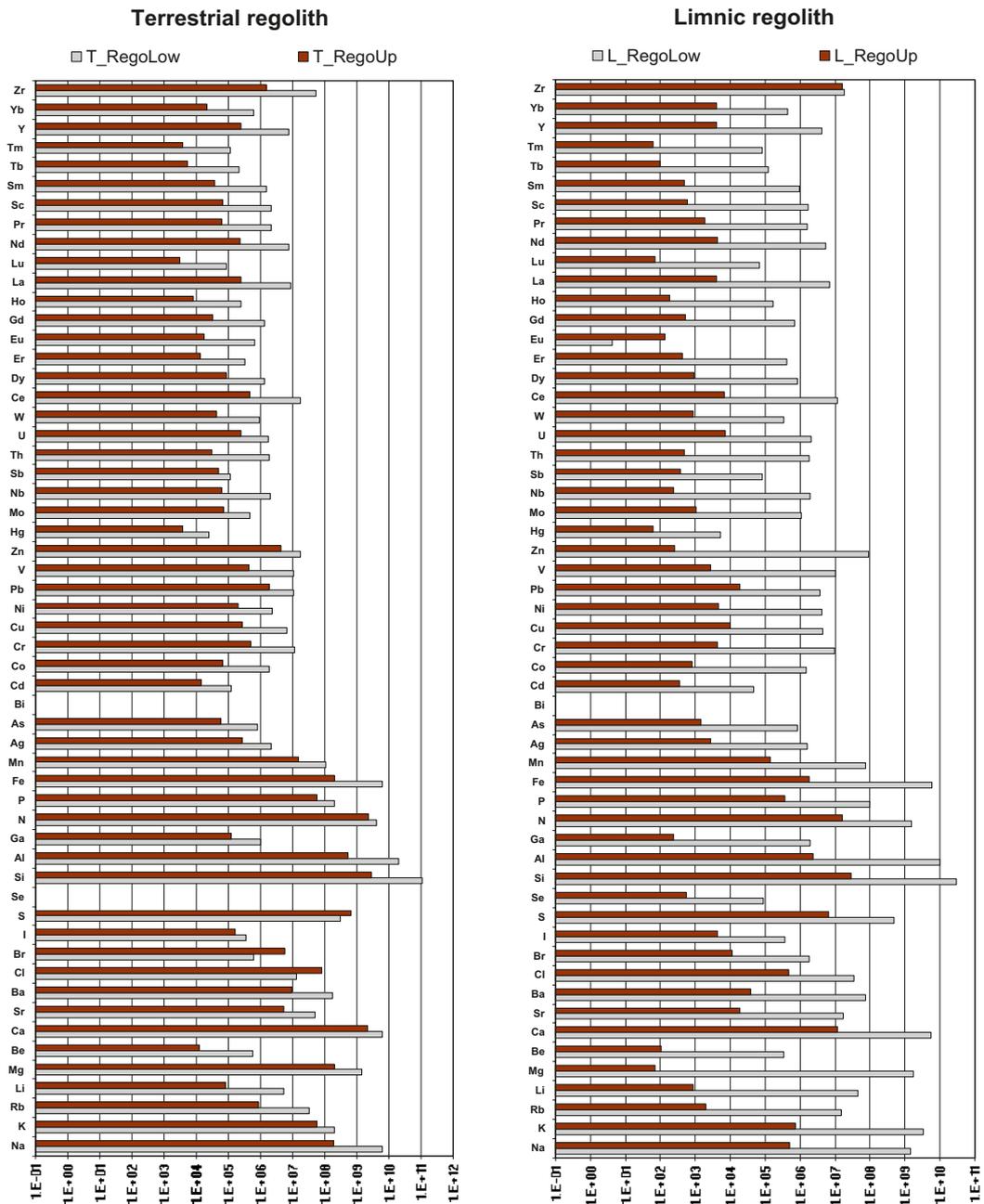
Limnic ecosystem										
Element	Fluxes				Pools					
	Deposition L_InAir	Import L_InWater	Accumulation L_Acc	Export L_OutWater	Producers L_Prod	Consumers L_Cons	Upper regolith L_RegoUp	Lower regolith L_RegoLow	Particulate L_Part	Dissolved L_Diss
Na	1.99E+05	1.49E+07	1.85E+04	5.98E+06	2.65E+04	3.34E+04	4.94E+05	1.43E+09	3.52E+03	4.01E+07
K	6.46E+04	9.47E+06	3.17E+04	3.79E+06	2.27E+04	3.31E+04	7.34E+05	3.39E+09	3.61E+03	2.61E+06
Rb	4.24E+02	8.52E+03	2.64E+02	3.34E+03	1.00E+02	3.28E+01	2.01E+03	1.53E+07	7.92E+00	1.87E+03
Li	1.29E+02	9.75E+03	9.56E+01	3.88E+03	3.20E+01	2.18E+00	8.75E+02	4.62E+07	2.23E+00	2.30E+03
Mg	3.03E+04	2.04E+07	2.07E+04	8.17E+06	1.28E+05	8.26E+03	6.63E+05	1.77E+09	1.86E+03	6.12E+06
Be	1.98E+00	1.78E+01	3.09E+00	5.95E+00	2.31E+00	5.83E-02	1.00E+02	3.29E+05	1.16E-01	4.41E+00
Ca	1.11E+05	1.68E+08	5.34E+05	6.72E+07	1.16E+07	2.27E+05	1.13E+07	5.55E+09	2.32E+04	3.06E+07
Sr	3.33E+03	2.83E+05	5.26E+02	1.13E+05	8.25E+03	1.26E+02	1.89E+04	1.75E+07	3.36E+01	3.27E+04
Ba	8.21E+02	7.15E+04	9.25E+02	2.83E+04	7.18E+03	1.11E+02	3.87E+04	7.50E+07	4.74E+01	1.27E+04
Cl	3.39E+05	1.38E+07	6.44E+03	5.58E+06	6.03E+04	6.41E+03	4.83E+05	3.37E+07	0.00E+00	6.26E+07
Br	1.14E+03	1.34E+05	2.28E+02	5.38E+04	9.32E+02	1.34E+03	1.12E+04	1.82E+06	2.16E+02	2.40E+05
I	3.26E+03	2.39E+04	7.27E+01	9.88E+03	4.33E+02	1.38E+01	4.24E+03	3.54E+05	4.10E+01	3.82E+03
C	0.00E+00	1.48E+08	2.94E+06	5.82E+07	1.72E+07	3.39E+06	1.67E+08	1.22E+10	2.14E+05	2.47E+07
S	1.84E+05	1.85E+07	7.01E+04	7.41E+06	2.07E+05	6.71E+04	6.33E+06	4.95E+08	8.05E+03	4.36E+06
Se	1.39E+02	3.35E+02	8.78E+00	1.44E+02	1.88E+01	2.35E+00	5.59E+02	8.66E+04	5.22E-01	7.12E+01
Si	5.53E+03	1.01E+07	6.63E+05	3.77E+06	6.42E+05	1.39E+04	2.89E+07	2.87E+10	1.97E+04	1.11E+06
Al	1.16E+01	3.26E+05	5.42E+04	1.06E+05	3.92E+04	7.27E+02	2.28E+06	9.70E+09	2.31E+03	3.01E+04
Ga	7.43E+00	3.11E+01	5.00E+00	1.09E+01	6.61E+00	5.14E-02	2.39E+02	1.94E+06	6.59E-01	2.73E+00
N	2.38E+05	3.31E+06	2.37E+05	1.24E+06	6.30E+05	6.54E+05	1.60E+07	1.50E+09	3.31E+04	6.87E+05
P	8.02E+03	4.13E+04	3.94E+03	1.56E+04	3.66E+04	1.58E+05	3.73E+05	9.95E+07	3.18E+03	4.37E+03
Fe	1.19E+04	5.05E+05	7.51E+04	1.70E+05	4.82E+04	1.19E+03	1.77E+06	5.94E+09	4.70E+03	5.69E+04
Mn	5.14E+03	1.32E+05	2.02E+03	5.25E+04	7.49E+03	1.15E+03	1.38E+05	7.43E+07	1.43E+03	2.68E+04
Ag	2.48E+00	1.81E+02	1.55E+02	2.42E+00	9.53E+01	4.08E-01	2.82E+03	1.60E+06	1.16E-01	5.93E-01
As	3.47E+01	7.48E+02	2.89E+01	2.90E+02	2.07E+01	7.29E+00	1.44E+03	8.57E+05	2.41E+00	1.15E+02
Bi	3.96E+00	8.69E+00	0.00E+00	3.87E+00	5.00E-10	5.00E-10	5.00E-10	5.00E-10	5.00E-10	5.00E-10
Cd	1.43E+01	2.74E+01	4.80E+00	1.02E+01	4.85E+00	2.10E+00	3.54E+02	4.66E+04	6.46E-01	3.83E+00
Co	1.89E+01	1.95E+02	2.09E+01	7.06E+01	2.18E+01	8.37E-01	8.17E+02	1.53E+06	9.89E-01	3.63E+01
Cr	2.35E+02	1.63E+03	1.13E+02	6.24E+02	6.36E+01	1.88E+00	4.25E+03	9.60E+06	2.42E+00	4.55E+01
Cu	1.63E+03	2.82E+03	2.19E+02	1.19E+03	8.12E+02	6.21E+01	1.01E+04	4.56E+06	2.15E+01	3.72E+02
Ni	5.51E+02	1.55E+03	1.13E+02	6.23E+02	9.79E+01	2.71E+00	4.48E+03	4.28E+06	9.20E+00	2.29E+02
Pb	2.76E+02	4.82E+02	2.29E+02	1.16E+02	2.02E+02	3.10E+00	1.94E+04	3.65E+06	3.06E+01	7.15E+01
V	2.17E+02	8.00E+02	1.14E+02	2.90E+02	6.71E+01	1.54E+00	2.75E+03	1.03E+07	5.02E+00	1.34E+02
Zn	5.34E+03	8.47E+03	1.16E+03	3.39E+03	1.92E+03	2.33E+02	3.79E+04	8.87E+07	4.68E+01	6.78E+02
Hg	1.04E+01	2.26E+01	5.69E-01	9.83E+00	8.67E-01	1.63E+00	6.21E+01	5.21E+03	1.12E-01	1.20E+00
Mo	1.98E+01	1.79E+03	1.25E+02	6.62E+02	6.99E+01	5.51E-01	1.07E+03	1.07E+06	1.82E+00	2.59E+02
Nb	7.92E+00	5.83E+01	3.23E+01	9.48E+00	1.34E+01	3.75E-02	2.40E+02	1.89E+06	5.39E-01	5.70E+00
Sb	3.56E+01	2.67E+02	4.50E+00	1.09E+02	1.56E+02	1.59E-02	3.68E+02	8.28E+04	8.73E-01	5.88E+01
Th	4.95E+00	9.36E+01	3.37E+01	2.27E+01	1.93E+01	3.12E-01	4.71E+02	1.81E+06	1.71E+00	5.93E+00
U	2.84E+00	8.28E+03	2.86E+02	3.19E+03	2.81E+02	3.02E-01	6.98E+03	2.04E+06	1.11E+00	1.03E+03
W	6.93E+00	7.40E+01	1.87E+01	2.19E+01	1.90E+01	3.19E-02	8.75E+02	3.32E+05	5.77E-01	8.67E+00
Ce	3.16E+01	7.22E+02	3.86E+02	1.17E+02	1.99E+02	5.92E-01	6.83E+03	1.18E+07	1.15E+01	6.04E+01
Dy	4.95E+00	9.42E+01	3.01E+01	2.46E+01	2.63E+01	4.02E-02	9.10E+02	8.23E+05	6.24E-01	7.93E+00
Er	2.48E+00	6.82E+01	1.97E+01	1.86E+01	1.50E+01	2.26E-02	4.25E+02	4.24E+05	3.91E-01	5.99E+00
Eu	6.44E-01	1.61E+01	4.02E+00	4.70E+00	4.14E+00	1.16E-02	1.30E+02	1.52E+05	9.03E-02	4.57E+00
Gd	7.43E+00	8.13E+01	1.65E+01	2.58E+01	2.11E+01	4.76E-02	5.19E+02	7.09E+05	3.94E-01	8.76E+00
Ho	9.90E-01	2.27E+01	6.75E+00	6.14E+00	7.44E+00	8.55E-03	1.82E+02	1.63E+05	1.35E-01	3.93E+00
La	1.93E+01	4.63E+02	1.83E+02	1.05E+02	1.33E+02	6.68E-01	4.13E+03	6.85E+06	7.98E+00	8.40E+00
Lu	2.97E-01	2.10E+01	2.64E+00	7.24E+00	2.49E+00	3.63E-03	7.02E+01	6.72E+04	4.62E-02	1.67E+01
Nd	1.55E+01	4.76E+02	1.84E+02	1.09E+02	1.35E+02	2.96E-01	4.42E+03	5.45E+06	4.39E+00	3.29E+01
Pr	1.09E+01	1.37E+02	6.48E+01	2.65E+01	2.90E+01	7.76E-02	1.82E+03	1.60E+06	1.28E+00	1.10E+01
Sc	6.44E+00	4.03E+02	2.03E+01	1.53E+02	1.34E+01	8.62E-02	5.69E+02	1.68E+06	8.61E-01	1.48E+01
Sm	6.93E+00	9.44E+01	3.08E+01	2.45E+01	2.11E+01	5.71E-02	4.92E+02	9.33E+05	8.31E-01	8.88E+00
Tb	9.90E-01	4.37E+01	2.43E+00	1.65E+01	4.50E+00	7.30E-03	9.63E+01	1.19E+05	9.97E-02	1.02E+01
Tm	3.47E-01	1.42E+01	2.69E+00	4.49E+00	3.56E+00	3.49E-03	6.33E+01	8.02E+04	5.44E-02	3.65E+00
Y	9.38E+00	6.51E+02	1.06E+02	2.14E+02	8.42E+01	2.75E-01	4.10E+03	4.10E+06	3.72E+00	2.14E+01
Yb	1.98E+00	6.68E+01	1.46E+01	2.03E+01	1.29E+01	2.09E-02	2.63E+02	4.51E+05	2.74E-01	5.72E+00
Zr	1.44E+01	1.13E+03	2.19E+02	3.55E+02	9.19E+01	3.37E+00	5.25E+03	1.86E+07	8.25E+00	9.48E+01

## B.4 Additional figures of absolute pool estimations

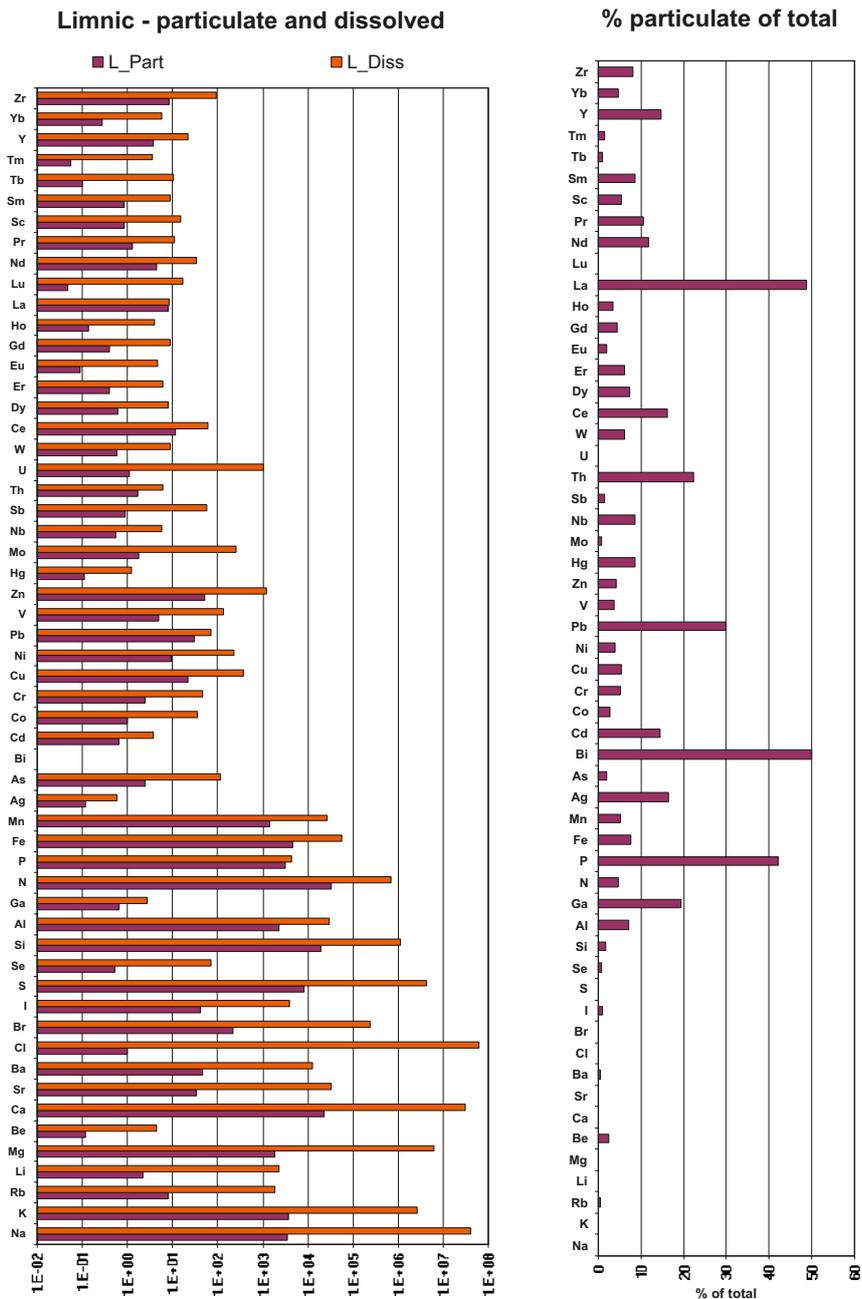
In this section additional figures showing the absolute size of the pools in the terrestrial and limnic systems.



**Figure B-4.** Absolute representation of the producer and consumer pools in the terrestrial (left) and the limnic (right) system in the Forsmark area. The figures denote the total contents expressed in g in the Norra Bassängen catchment. The total catchment area is 8.5 km<sup>2</sup>, of which the lakes comprise 0.66 km<sup>2</sup>.



**Figure B-5.** Absolute representation of the lower (RegoLow) and upper (RegoUp) regolith pools in the terrestrial (left panel) and the limnic (right panel) system in the Forsmark area. The figures denote the total contents expressed in g in the Norra Bassängen catchment. The total catchment area is 8.5 km<sup>2</sup>, of which the lakes comprise 0.66 km<sup>2</sup>.



**Figure B-6.** Absolute representation of the particulate and dissolved fractions in the limnic system in the Forsmark area (left panel). The figures denote the total contents in all lakes in the Norra Bassängen catchment expressed in g. The total catchment area is 8.5 km<sup>2</sup>, of which the lakes comprise 0.66 km<sup>2</sup>. The right-hand figure show the particulate fraction in relation to the total contents in the water phase.