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Hydrogeology and groundwater quality in the Nordic and Baltic countries

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ABSTRACT

Groundwater utilization and groundwater quality vary in the Baltic and Nordic countries mainly because of different geological settings. Based on the geology, the countries were treated in the following three groups: (1) Fennoscandian countries (Finland, Sweden, and Norway), (2) Denmark and Baltic countries (Estonia, Latvia, and Lithuania), and (3) Iceland. Most of the utilized groundwater resources are taken from Quaternary deposits, but Denmark and the Baltic countries have in addition, important resources in Phanerozoic rocks. The groundwater quality reflects the residence time of water in the subsurface and the chemical composition of the geological formations. Concentrations of ions in the Fennoscandian bedrock are elevated compared to Iceland, but lower than in Denmark and the Baltic countries. Compared to groundwater in the bedrock, groundwater in Quaternary deposits has usually lower concentrations of dissolved minerals. Unconfined Quaternary aquifers are vulnerable to contamination. Examples from Denmark and the Baltic countries illustrate challenges and successful effects of mitigation strategies for such aquifers related to agricultural application and management of nitrogen. Confined and deeper groundwater is better protected against anthropogenic contamination, but water quality may be affected by harmful compounds caused by geogenic processes (*viz*, sulfide, arsenic, fluoride, and radon).

Key words: Baltic and Nordic countries, geological framework, groundwater quality, groundwater pollution, groundwater utilization

HIGHLIGHTS

- The paper reviews groundwater quality, and groundwater utilization in the Baltic and Nordic countries.
- Chemical data from all involved countries have been compiled and the data is summarized in the Supplementary Material.
- The mineral composition of the underlying bedrock governs to a large extent the chemical character of groundwater in the Quaternary deposits.
- Denmark and the Baltic countries have large and complex aquifers, whereas the aquifers in Iceland and the Fennoscandian countries are relatively small and with less complexity.
- The groundwater in the region is largely of high quality, but local challenges exist related to geogenic (*viz* radon, arsenic, fluoride) and anthropogenic origin (*viz* nitrate, pesticides).

INTRODUCTION

In this paper, we focus on groundwater issues in the Nordic and Baltic countries (called the Nordic Region for short in the following). The region spans a geographical area of 56° East–West (24°W–32 °E) and 17° North–South (54°–71 °N). The total land area is close to 1.5×10^6 km^2 with a population of 33.5 million people. The climate and vegetation vary from arctic tundra in the north to boreal coniferous and deciduous forest to the south. In Iceland, the climate is characterized as tundra in the highlands, and sub-polar oceanic close to the coast. In Fennoscandia and the Baltic countries, the climate changes eastwards from coastal to continental, and specific runoff varies from around 300 mm/year in the eastern and southern part to more than 6,000–7,000 mm/year in the marine western and northern parts. Latitude, altitude, and distance to open ocean explain variation in temperature and seasonality. In general, precipitation exceeds the amounts of evapotranspiration in the region, and issues concerning climate-related scarcity of groundwater recharge is therefore not a main issue in this review article. The geological heterogeneity, on the other hand, is significant, and explains the main variation with respect to groundwater quality and groundwater utilization. Therefore, the focus of this article is the geological setting and the groundwater quality. The major fractions of mainland Norway, Sweden and Finland are characterized as non-aquiferous or with local aquifers of minor spatial extension (Figure 1). In Denmark and the Baltic countries, the geological history provided favorable conditions for storage and utilization of groundwater. The same is true for Iceland and the Faroe Islands, but for different geological reasons. The Nordic Region will therefore be divided in three separate groups: (1) The Fennoscandian countries (Finland, Sweden, and Norway), (2) Denmark and the Baltic countries, and (3) Iceland and the Faroe Islands.

Groundwater occurs in the void space of the subsurface. The connectivity of the void space determines the bulk permeability, which is decisive for the residence time of water in the subsurface. In addition to the bulk permeability, the residence time of water depends on the effective porosity, the liquid properties of the formation water (*viz*, viscosity and density), and recharge and discharge conditions for the formation water. Estimation of residence time of water in the subsurface requires a combination of numerical flow modeling, well testing, and large-scale estimates of the water balance. Observations

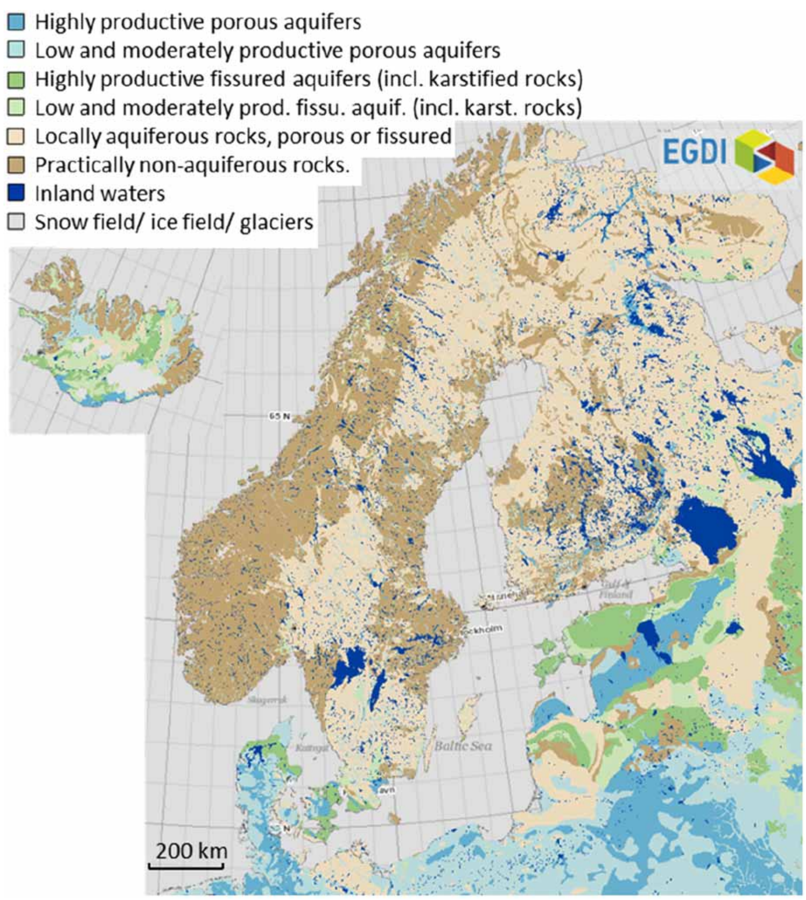

Figure 1 | Hydrogeological map of the Baltic and Nordic countries. The most productive aquifers are mainly located in Denmark and the Baltic countries and partly in Iceland, whereas most parts of Fennoscandia have low pervious rocks or locally aquiferous formations (EGDI 2022). Please refer to the online version of this paper to see this figure in color: http://dx.doi.org/10.2166/nh.2022.018.

of groundwater chemistry and water isotopes can be used as input for inverse modeling of critical flow parameters. In spite of the close relation between residence time of water and the groundwater chemistry, the topic of residence time is beyond the scope of this review article. The chemical composition of pore-water depends on the atmospheric input, soil processes, mineral solubility, and reaction kinetics. The H^+ concentrations of the pore-water and the redox potential (Eh) are critical for biogeochemical redox reactions. Temperature and pressure play a role in deep aquifers or in geothermally active areas. Hence, from a theoretical point of view, the driving forces behind groundwater quality are well known, and the governing equations are implemented in numerical models (e.g. PHREEQC, Parkhurst & Appelo 2013). These models often imply chemical equilibrium, and the spatial heterogeneity are usually unknown. The requested hydrogeochemical variables can therefore not be obtained by pure mathematical modeling alone. Thus, there is a need to describe the hydrogeochemical variance to provide robust decisions with respect to water management. The aim of the current review is to recapitulate existing knowledge that is relevant for the Nordic Region. Even though this review has a geogenic perspective, impact of human activity cannot be ignored. We therefore include examples from Denmark and the Baltic countries to illustrate this point. Other problems like arsenic in groundwater, are related to natural geogenic processes. Both cases illustrate the needs for monitoring and research to understand the groundwater quality.

Geological framework

Groundwater is mainly recharged by percolation of rain and snowmelt through soil and sediment deposits covering the bedrock. Since the chemical composition of soil and sediments are closely related to the local lithology, the geological framework is the key to hydrogeochemistry.

The Precambrian bedrock is the result of a series of mountain chain formation processes (orogenesis) which started for more than 3,500 million years ago (m.y.). The oldest rocks are in the northern part of Finland, Sweden, and Norway (Figure 2). In this area, the various rock types in the bedrock were formed during the Archean (3,500–2,500 m.y.) and Svecokarelien (2,500–1,950 m.y.) periods. The Precambrian bedrock in the southern parts of Finland and Northeastern parts of Sweden was formed during the Svecofennian orogeny (1,950–1,750 m.y.). This period was followed by granite and porphyry intrusions within the Trans Scandinavian Igneous Belt (1,810–1,650 m.y., Högdahl *et al.* 2004). The two following orogenesis were the Gothian (1,700–1,500 m.y.) and the Sveconorwegian (1,130–900 m.y.). These bedrock provinces are located in the southwestern part of Sweden and the southern part of Norway. Neoproterozoic and Phanerozoic rocks (1,000–250 m.y.) are found in the Oslo Graben in South Norway, western, central and southern Sweden (Scania), and in Denmark. Precambrian rocks were during Neoproterozoic times denudated to a low-relief surface, a platform, forming the Fennoscandian–Baltic shield. The next major orogenic event occurred in Cambrian–Silurian time (about 500–400 m.y.) when the Baltic plate collided with the Laurentian plate (Greenland–North American) and formed the Caledonian Mountain Chain. The Caledonian rocks occur in large 'nappes' or 'thrust sheets', up to several kilometer thick rock slabs that were pushed towards the east and southeast like giant blankets. The Caledonian nappe complexes contain a great variations of rock types, comprising Precambrian crystalline igneous rocks and gneisses, Neoproterozoic sandstone successions (sparagmites), and metamorphic Cambro-Silurian rocks like phyllite, schist, greenstone, gneisses, and intrusions as gabbro, diorite, and granite. Most of the Caledonian rocks in western Scandinavia were uplifted during Cenozoic time and followed by erosion and transport of sediments to the North Sea basin. These processes gave rise to the present high-relief Scandinavian Mountain Chain. In Denmark and the Baltic countries, the Precambrian platform was covered by sedimentary rocks from Neoproterozoic–Phanerozoic time. Most of these sedimentary rocks were formed in a shallow sea basin. In the Baltic region, the Precambrian platform was deformed into a wide synclinal form with an increasing width to the southwest and with a gentle dip in the same direction (Vaikmäe *et al.* 2021). In the Estonia territory, the depth to the Precambrian basement varies from 100 m below the surface in the north to 800 m in the south, while in the western part of Lithuania, the maximum depth is more than 2,000 m. As indicated in Figure 3, we find the oldest sedimentary cover rocks close to the surface in the northern parts of Estonia. Younger rocks are successively found in the south-westward direction because of the gentle dip (0.1°–0.3°) towards southwest (Raukas & Teedumäe 1997). This complex sedimentary basin constitutes the Baltic Artesian Basin which is explained in more detail below.

The geology of Iceland and the Faroe Islands is a result of the rifting of the Pangaea supercontinent which separated the Eurasian and the North American plate from each other. Both islands consist mainly of volcanic rocks. The Faroe Islands were formed during a few million years in Paleogen time (55 m.y.) by a series of volcanic eruptions. The eruptions formed basaltic layers with interbeds of volcanic ashes and some relatively thin sedimentary formations. Iceland is an anomaly on the Mid-Atlantic spreading axis. The oldest bedrocks are found most distant from the current active spreading axis, in the

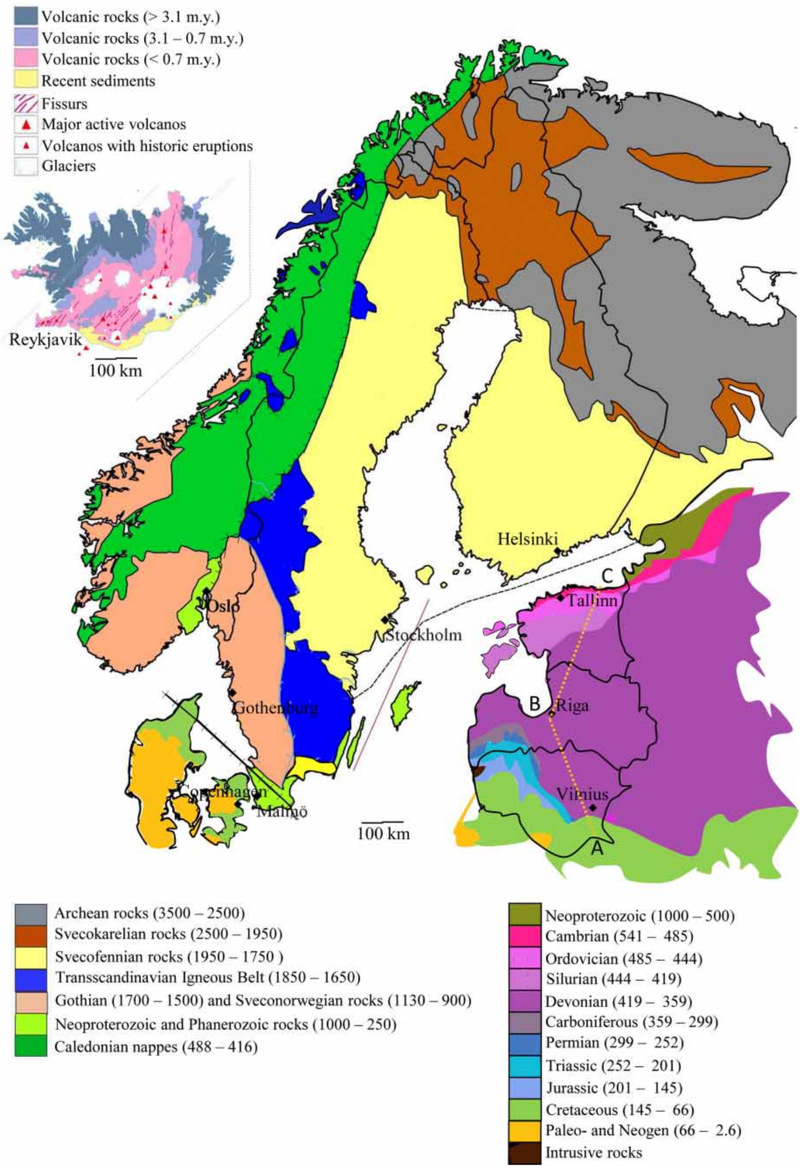

Figure 2 | Simplified geologic map of the Baltic and Nordic countries. The Precambrian crystalline basement in Fennoscandia consists of five mega blocks: The Archean block, the Svecokarelian block, the Svecofennian block (included the Transscandinavian Igneous Belt), the Sveconorwegian block (included the Gothian rocks), and the Caledonian nappes. The Precambrian platform below Denmark and the Baltic countries are overlaid by Phanerozoic rocks from the Paleozoic time: Cambrian, Ordovician, Silurian, Devonian, and Permian; Mesozoic time: Triassic, Jurassic, and Cretaceous; and the Cenozoic time: Paleogene and Neogene. A vertical cross-section through the Baltic countries – indicated by A–B–C, is given in Figure 3. Iceland is located on the Mid-Atlantic spreading ridge, and the bedrock consists of young volcanic rocks with ages increasing with the distance from the spreading axis. Sources of information: Ramberg *et al.* (2008); Fennoscandian platform (2021); Baltic geology (2021); Danish geology (2021); Icelandic geology (2021). Please refer to the online version of this paper to see this figure in color: http://dx.doi.org/10.2166/nh.2022.018.

western, northern, and eastern parts of Iceland. These rocks were generated during the Miocene period (from 23 m.y.), and have low permeability. The Pliocene (5.3–2.6 m.y.) and Pleistocene (2.6 m.y.–11,700 years ago) bedrocks, which border the volcanic belts, have higher permeability, larger springs, and spring-fed rivers. The Holocene lava fields, with their central volcanoes and associated fissure swarms, have exceptionally high permeability. The location on the spreading axis gives Iceland access to valuable geothermal energy.

The geological cross-section in Figure 3 indicates the complex stratigraphy and aquifer systems of the Baltic countries. The surface of the crystalline basement is covered by the Ediacaran (Vendian) to Cambrian sandstones and clays. These strata are

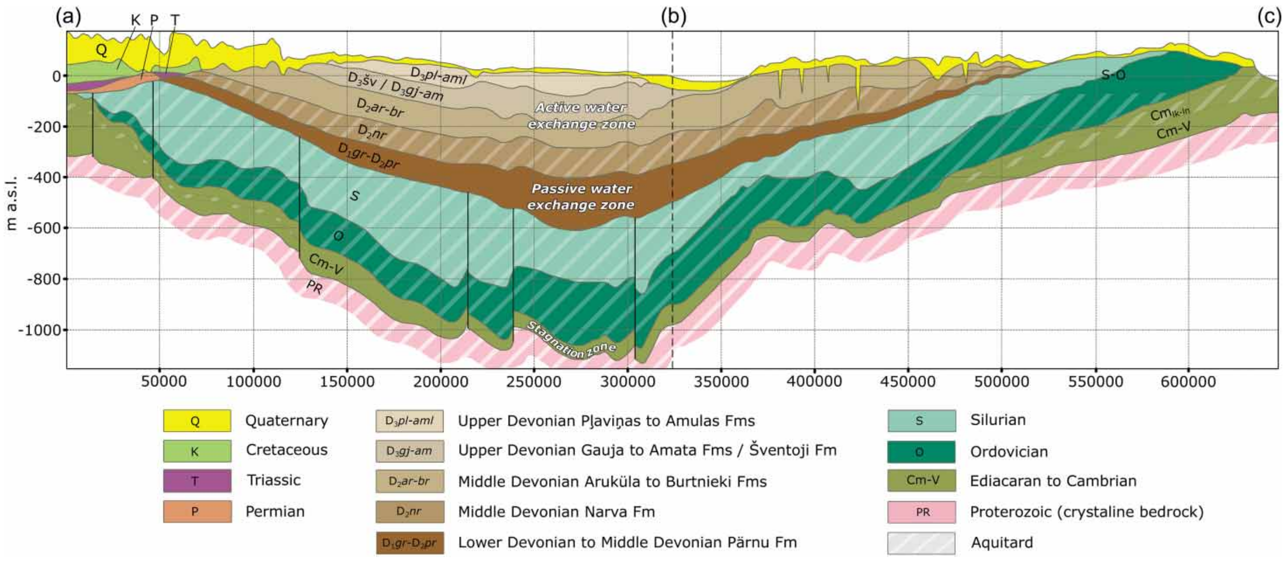

Figure 3 | Cross-section through the Phanerozoic rocks above the Precambrian crystalline basement for the Baltic countries (locations a–b–c are indicated in Figure 2). The cross-section shows the main geological units of the Baltic Artesian Basin based on the model created by Virbulis *et al.* (2013). Black vertical lines indicate faults. BAB consists of an active water exchange zone, a passive water exchange zone, and a stagnation zone. See text for more details. Please refer to the online version of this paper to see this figure in color: http://dx.doi.org/10.2166/nh.2022.018.

exposed at the bottom of the Gulf of Finland. The Cambrian–Vendian (Cm–V) sandstone holds fresh artesian groundwater used for water supply in Northern Estonia (Perens *et al.* 2001), while in the rest of BAB the salinity increases. In South Estonia, Latvia, and Lithuania, the Upper-Middle Devonian aquifer system are of major importance for water supply. More details of the BAB geology are given below in the Baltic hydrogeology section.

Quaternary deposits

The maximum extension of the Fennoscandian ice sheets in the Pleistocene (Quaternary) time occurred during the Elster (490,000–410,000 years ago) and Saale (240,000–130,000 years ago) glaciations, when the northwestern Europe, including the Nordic and Baltic regions were covered with glacial ice. During the last glaciation (Weichsel, 115,000–11,700 years ago), the Fennoscandian ice sheet had a maximum extent some 20,000 years ago. Whereas during the Elster and Saale glaciations, the ice sheets extended beyond Fennoscandia, Denmark, and the Baltic countries, the ice sheet during the Last Glacial Maximum did not cover the western parts of Denmark (Ehlers 2020). Advance and retreat of the glaciers left thick deposits of till (moraine) and glaciofluvial sediments in Denmark and the Baltic countries, but less glacial debris over the Fennoscandian territory where the deposits in general are thinner. Especially, in Norway, there are widespread areas with exposed bedrock or sparse and patchy deposits (Kitterød 2017), where thick fluvial and glaciofluvial deposits are restricted to the valleys. Nevertheless, Quaternary deposits hold important aquifers in all involved countries due to high permeability in combination with regular and substantial recharge from precipitation and snowmelt. In addition, artificial recharge or bank infiltration from surface water occur in aquifers with extensive water abstraction.

GROUNDWATER GEOCHEMISTRY

The chemical quality of groundwater is governed by several different processes. The composition of wet and dry precipitation plays a role. These inputs are determined by natural and anthropogenic sources (*viz*, marine and terrestrial aerosols, emissions from industry and other human activity). Plant growth and decay modify ion composition. Root respiration together with microbiological activity impact the soil water (i.e. high soil P_{CO_2}, carbon accumulation, and degradation). Chemical weather is an important modifying factor. It is particularly intense in the 'soil reactor', which involves carbonic acid, reactive minerals, and soil water flow (Garrels & Mackenzie 1967; Reardon *et al.* 1979; Jørgensen *et al.* 1991). Similar weathering reactions also take place in the saturated groundwater zone. Usually, the residence time of water increases with depth or with the length of the flow path. The time available for reaction will normally lead to more evolved (or mature) groundwater. Finally, mixing of different waters affects the groundwater (*viz*, intrusion of seawater, saline formation water, or surface water). The average chemical composition of groundwater for all countries in the Nordic Region is given in the Supplementary Material and details are discussed in the following text.

A comprehensive hydrogeochemical data material were compiled and made publicly available by the Northern Europe Geochemistry (NEG) Atlas project (NEG 2009; Salminen 2009). Even though groundwater samples were not included in NEG, it is interesting to review some of the main results from the project. Hydrogeochemical analyses were undertaken of samples taken from terrestrial moss (4,287), organic soil (6,475), mineral soil (20,502), stream sediments (15,421), stream water (2,723), and lake water (12,954, numbers of samples in parenthesis). Even though the spatial variability was overwhelming, it is possible to deduce some general conclusions: anthropogenic impact was most evident in samples taken from terrestrial moss. The chemical signal in the organic soil horizon was partly influenced by human activity and partly by the bedrock. Samples from the mineral soil horizon reflect the geochemical character of the local bedrock. Stream sediments were essentially determined by the geochemistry of the bedrock of the upstream catchment. The general chemical pattern of elements like Ca, Mg, SO_4^{2-}, Sr, and Ba is quite similar in organic soil samples and in surface water. The latter result indicates the influence of shallow groundwater on the chemical composition of streams and lakes. The chemical signal of surface water has a distinct geogenetic signature, which means that the hydrogeochemistry of groundwater and surface water mirrors the general chemical composition of the bedrock.

Figure 4 shows examples of results that can be downloaded from the NEG (2009) website. The two maps in Figure 4 illustrate arsenic concentrations in surface waters and mineral soils. Hotspots of arsenic concentrations in surface water are indicated in Sweden and in Norway with concentrations above 3 μg/L. The main chemical composition of mineral soils reflects the local bedrock, which in this case indicates similar arsenic content in the bedrock. The lack of overlap between arsenic concentrations in mineral soils and in surface waters is most probably caused by binding characteristics of As. The mineral soils are the primary source of As, while the release to the aqueous phase is determined by weathering reactions and soil conditions. The main mobilization mechanisms of As are either oxidation of sulfides or reduction of iron- and aluminum oxides whereby releasing sorbed/bound As (Bhattacharya *et al.* 2010).

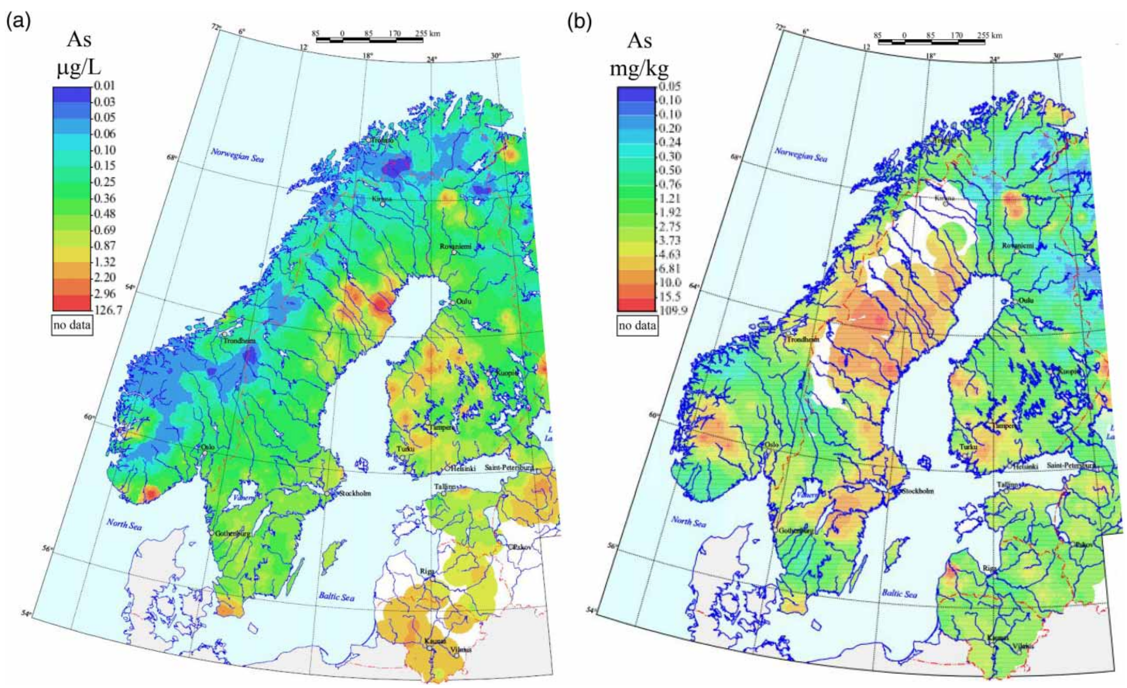

Figure 4 | Arsenic concentration in (a) surface water and in (b) mineral soil (C-horizon). The soil samples were analyzed by Aqua regia extraction from soil fraction with grain size less than 2 mm. The maps are taken from NEG (2009). Please refer to the online version of this paper to see this figure in color: http://dx.doi.org/10.2166/nh.2022.018.

Fennoscandian hydrogeochemistry

The most important aquifers in Fennoscandia are glacifluvial and fluvial deposits (*viz*, glacial deltas, eskers, kames, sandur plains, riverbanks). These high permeable aquifers are usually thin and with minor lateral extension – usually much less than 100 km^2 (igras-GGIS 2022). The residence time of water in such aquifers is therefore relatively short. Hence, the ion concentration is relatively low compared to ground water at greater depths (Figure 5). Currently, there are approximately 5,000 registered aquifers in Finland with size varying from less than a square kilometer to maximum of hundred square kilometers (Britschgi *et al.* 2009). In Sweden, the authorities estimate the number of groundwater bodies to approximately 3,000 (McCarthy & Gustafsson 2021). Approximately 1,400 groundwater bodies have been defined in Norway, but the areal extent of these aquifers is less than 0.1% of the total estimated aquifer area in the European Union (EU) (EEA 2021). These groundwater bodies are for the most part located in valleys and often bordered by rivers and lakes, and also partly recharged from surface water. The unconsolidated sediments have high permeability and are quickly recharged by precipitation and snowmelt.

The coastal areas of the Baltic Sea in Finland and Sweden were affected by the Littorina sea phase when the Baltic Sea covered a larger area than present. This resulted in relict sea salts and fine sediments covering aquifers and creating low oxygen conditions with increased concentrations of Cl, Na, Fe, Mn, and SO_4^{2-} in groundwater (Hatva 1989; Lahermo *et al.* 1990). Acid sulfate soils are also a problematic soil type in these regions, causing risks to surface waters (Sundström *et al.* 2002; Beucher *et al.* 2013; Mattbäck *et al.* 2017). Elevated Fe and Mn concentrations and low pH are also listed as common groundwater quality issues (Kløve *et al.* 2017).

A comprehensive study by Banks *et al.* (1998a, 1998b) revealed that groundwater in Quaternary deposits in Norway showed an evolution from Na–Cl dominated, immature waters which reflect marine salts in precipitation to Ca–HCO_3 dominated waters derived from calcite dissolution. In the same study, three main water types were distinguished in bedrock, Na–Cl, Ca–HCO_3, and Na–HCO_3 waters, where Ca–HCO_3 type dominated while Na–HCO_3 waters comprised 25% of the samples.

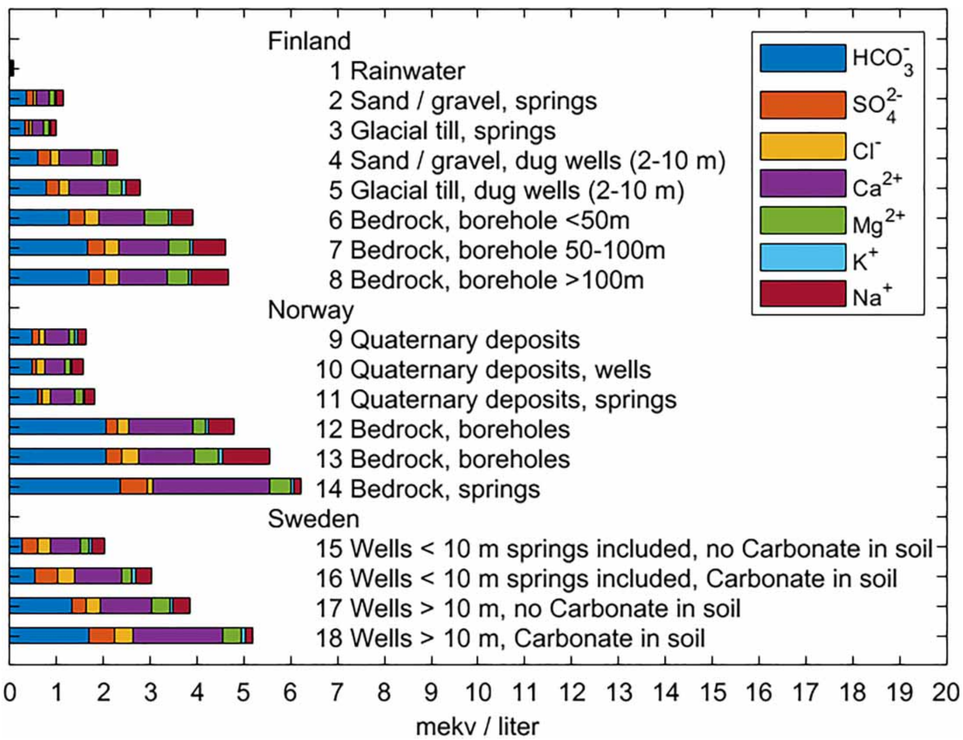

Figure 5 | Average ion concentrations in groundwater from Finland, Norway, and Sweden. The rainwater samples from Finland illustrate the significant difference in ion composition between precipitation and groundwater. Data sources from Finland (Karro & Lahermo 1999), Norway (Banks *et al.* 1998b; Frengstad 2002; Frengstad & Banks 2008; NGU 2022), and Sweden (Maxe 2001). Please refer to the online version of this paper to see this figure in color: http://dx.doi.org/10.2166/nh.2022.018.

The largest precipitation fed aquifer in Norway is the Gardermoen delta (~80 km^2) located north of Oslo. Jørgensen *et al.* (1991) made a chemical mass budget for the major solute constituents of the Gardermoen groundwater and evaluated the processes causing their uptake and release: Only 10% of the solutes in the groundwater discharge originates from precipitation input. As an example, 72% of sulfate comes from oxidation of pyrite (FeS_2). The remaining 28% came from precipitation. For all acid consuming processes (mainly chemical weathering), the H^+ in precipitation accounts for only 9%. Most acids (76%) originate from CO_2 produced in the soil, 14% from sulfide oxidation and ca. 1% from organic acids. There is an upper decalcification zone in the delta making the groundwater hard, and the deeper part of the aquifer is anoxic. Excess chloride comes probably from old (9,500 years) fossil sea water in the bottomset of the delta.

Contrary to the highly permeable Quaternary deposits, water in most of the Fennoscandian bedrock has long residence time. The effect can be detected in the increasing content of dissolved ions (Figure 5). For groundwater utilization, the matrix (primary) permeability in crystalline rocks in Fennoscandia is essentially close to zero. Interconnected fractures and joints make up a secondary permeability, which usually yields sufficient of water for private households and small waterworks. Studies by Rohr-Torp (1994) and Morland (1997) showed a significant spatial correlation between specific water yield and variations of postglacial isostatic rebound velocities in Norway. According to these studies, the specific water yield increased by a factor of 2 for boreholes drilled in similar type of rocks, if the increase in vertical uplift was about 1 mm/year. This was the case for brittle bedrock, while schists and softer rocks gave very little yield.

Karro & Lahermo (1999) pointed out that the composition of the bedrock groundwater is controlled, among other things, by the type of the bedrock, the density and hydraulic conductivity of fractures, the nature of fracture coatings and fillings (e.g. carbonates, iron sulfides, ferrihydroxides, clay minerals), and the geochemical and hydraulic characteristics of the overlying soil cover. Karro & Lahermo (1999) mentioned that in coastal areas of Finland, relict salts in bottom sediments and in fractured bedrock have a greater effect on the overall chemistry of the bedrock groundwater than structural or petrological peculiarities of the bedrock. Frengstad & Banks (2003) analyzed more than 1,600 samples from crystalline basement in Norway and found a trend towards high pH-$NaHCO_3$ at larger depth. Distance to the coast controlled the distribution of Cl, Br, and I in the bedrock groundwater, and the highest pH-values were found in boreholes close to the coast (Banks *et al.* 1998a). Frengstad (2002) detected, however, surprisingly little dependence of major ion chemistry on aquifer lithology. He pointed out that important factors for the ion chemistry might be the Quaternary weathering history, the initial CO_2 pressure of recharge water, the degree of openness or closure of the fracture system. Weathering of silica minerals is an important buffering mechanism. Carbon acid is consumed during the hydrolysis of silica minerals and transformed to bicarbonate: $CO_2 \rightarrow H_2CO_3 \rightarrow HCO_3^-$. The increased concentration of bicarbonate balances the released cations from the silica minerals. Aluminum and to some extent silica, is bounded in insoluble product minerals. The net effect is an excess of silica which increase the silica concentration in the water phase.

A Finnish study of deep groundwater revealed a density stratification of fresh water at the top to brackish or saline groundwater at the bottom. In the upper part, HCO_3 concentrations were relatively high, whereas deeper down the brackish and saline water were mainly of Na–Ca–Cl or Ca–Na–Cl-type and seldom of Na–Cl-type groundwater (Karro & Lahermo 1999). In cases Mg and SO_4 concentrations were high.

For deep groundwater with long residence time the chemical composition can be governed by mineral composition of the host rock, as demonstrated by a study from Finland (Karro & Lahermo 1999). Low values of bicarbonate were observed in hornblende gabbro and sandstone, while high values were found in serpentinite. High values of sulfate appeared in unmetamorphosed silt and shale while mica gneiss, black schist, serpentinite, skarn, and quartzite had low values. A similar chemical pattern could be recognized in groundwater in the glacial till on top of the bedrock. The glacial till had local origin and therefor reflected the composition of the bedrock (Karro & Lahermo 1999). More details on average chemical composition of groundwater are given in the Supplementary Material.

Groundwater concerns related to geogenic origin

Radon. Recording of radon in groundwater started in the Nordic countries before 1910 (Knutsson 2008). The perception was that radioactivity had a positive health effect. Today there are general restrictions on maximum levels of radioactivity, and radon is one of the main sources for radioactivity hazards in groundwater. According to Swedish legislation, radioactivity in groundwater should not exceed 1,000 Bq/L (Knutsson 2008). High levels of radioactivity are measured in Precambrian granites and pegmatites, and in Phanerozoic black shale in Sweden and Norway (Banks *et al.* 1998a; Knutsson & Olofsson 2002). According to Knutsson (2008), the highest recorded levels of radioactivity were measured in southern Finland

(77,500 Bq/L, Leveinen *et al.* 1998); in Southeastern Sweden (57,000 Bq/L, Åkerblom & Lindgren 1997), and Southeastern Norway (31,900 Bq/L, Banks *et al.* 1998a).

Fluoride. Groundwater in igneous rocks may contain fluoride concentrations above permissible levels for drinking water (1.5 mg/L). High fluoride concentrations are reported in rapakivi granite in Southeastern Finland (5.8 mg/L, Lahermo *et al.* 1990), and in mafic rocks in Norway (8.26 mg/L, Banks *et al.* 1998a). Knutsson (2008) reports that as much as 15–17% of the wells in rapakivi granite have fluoride concentrations above 1.5 mg/L.

Arsenic. There is an increasing concern on arsenic compounds in drinking water. In the Fennoscandian bedrock, arsenic is mainly associated with feldspars, apatite and sulfide minerals which occur in mafic and ultramafic rocks, and in Precambrian granites and anorthosites (Frengstad *et al.* 2000). Knutsson (2008) refers to investigations which indicate that 15% of the wells in southwestern part of Finland had arsenic concentrations above the permission level for drinking water (10 μg/L, Leveinen *et al.* 1998). In the Pirkanmaa region in Finland, Backman *et al.* (2006) found that 22.5% of the drilled wells had As concentration exceeding 10 μg/L. 1.5% of the investigated wells in Norway had arsenic concentrations above this level (Frengstad *et al.* 2000), while 5% of the investigated wells in Sweden exceeded the drinking water requirements (Socialstyrelsen 2006 in Knutsson 2008). However, in the Northeastern part of the Västerbotten county in Sweden, an area with considerable sulfide mineralizations, Bhattacharya *et al.* (2010) found frequent elevated As concentrations in groundwater wells (up to 350 μg/L). Here, 29% of the drilled wells had As concentration above 10 μg/L.

Baltic hydrogeochemistry

The Baltic territory is situated on the multi-layered sedimentary Baltic Artesian Basin (BAB, Figures 2 and 3), which holds vast amounts of groundwater with varying chemical composition. Igneous and metamorphic Paleoproterozoic rocks form the basement of the BAB, which contains saline groundwater in their uppermost weathered portion. The crystalline basement serves as an impermeable base layer for all the overlying aquifer systems (Vallner 1997). The most important aquifer systems and aquitards are formed by terrigenous and carbonate sedimentary rocks, which fills the whole BAB with Quaternary deposits on top (Figure 3). Average chemical compositions of groundwater related to the different BAB aquifers are given in the Appendix.

The Cambrian–Vendian aquifer system is a term with dual meaning among the Baltic countries. In Latvia the system comprises all Cambrian to Vendian formations between the crystalline basement and the Ordovician formations (Retiķe & Dēliņa 2018). In Estonia and Lithuania, the Cambrian–Vendian aquifer corresponds to layers from the crystalline basement up to the lower Cambrian Lontova clays, which forms a regional aquitard (Lukševičs *et al.* 2012), leaving the upper part of the Cambrian to another Ordovician-Cambrian aquifer system (Karro *et al.* 2009). The Cambrian–Vendian aquifer system lies on top of the crystalline bedrock and is distributed throughout BAB, except the Mõniste uplift area in Southern Estonia (Perens & Vallner 1997). The water-yielding portion of the aquifer consists of sandstones and siltstones with interlayers of clay. In the Eastern part of Estonia and Latvia, up to 53 m thick clays of the Kotlin Formation divide the aquifer system into two aquifers – Voronka and Gdov (Perens & Vallner 1997). The Cambrian–Vendian aquifer system is used as a major source for public water supply in Northern Estonia, where the water has a characteristic Cl-HCO_3–Na–Ca and HCO_3–Cl–Ca–Na composition with TDS mainly below 1.0 g/L (Perens *et al.* 2001). Recharge occurs in Northern Estonia where the overlying aquitard is penetrated by ancient-buried valleys (Karro *et al.* 2009). However, the salinity of the Cambrian–Vendian aquifer system rises with depth and reaches TDS values of up to 22 g/L in Southern Estonia, up to 140 g/L in Latvia (Levins *et al.* 1998; Raidla *et al.* 2009) and up to 200 g/L in western part of Lithuania (Zinevicius & Sliaupa 2010) where Cl^- and Na^+ ions dominate. According to dating tracers, these brines are likely the result of evaporative enrichment of seawater with an estimated age of 1–5 Ma (Gerber *et al.* 2017), while in the Northern part of the BAB, water of glacial origin from the last Ice Age has been found (Raidla *et al.* 2012; Vaikmäe *et al.* 2021). As a result, in Latvia and Lithuania this aquifer system is considered a stagnant water exchange zone.

In the 1970 and 1980s the deepest parts of the Cambrian–Vendian aquifer system were intensively prospected for oil and gas. The investigations brought up the potential for industrial bromide extraction and geothermal energy (Levins *et al.* 1998). The temperature of the aquifer system can reach 40–60 °C in Southwestern part of Latvia and 70–90 °C in western part of Lithuania (Levins *et al.* 1998; Zinevicius & Sliaupa 2010). The latest study emphasized the potential of the aquifer system for carbon dioxide storage (Krūmiņš *et al.* 2021).

The following Silurian–Ordovician sequence consists of marls and clays with occasional limestone and dolostone beds and forms a regional aquitard with a thickness up to 850–900 m in western parts of Latvia and Lithuania (Lukševičs *et al.* 2012). In Latvia, the aquitard separates the stagnant water exchange zone (Cambrian–Vendian aquifer system) from the overlying passive water exchange zone. The uppermost part of the Silurian–Ordovician sequence, however, is perceived as an aquifer system in central and Western Estonia, in islands of the West-Estonian Archipelago and SouthEast Lithuania where it is an important and often the sole source of drinking water (Perens & Vallner 1997; Lukševičs *et al.* 2012). The aquifer system has a characteristic HCO_3–Ca–Mg and HCO_3–Mg–Ca water type with TDS mainly below 0.6 g/L in its upper 30–50 m parts in Estonia. In coastal areas and greater depths, the content of Cl^- and Na^+ in groundwater increases and HCO_3–Cl–Na–Mg–Ca type water with TDS 0.3–1.5 g/L is widespread (Perens *et al.* 2001).

The Middle-Lower Devonian aquifer system consists of fine-grained weakly cemented sandstones and siltstones with interlayers of clayey and dolomitized sandstone. The aquifer system is distributed through the territory of Latvia with the maximum thickness of 200 m (Levins *et al.* 1998), while it dips out in the Northern and Southern part of the BAB. In the Southern Estonia and Northeastern part of Latvia, the Middle-Lower Devonian aquifer system (partly together with the underlying Silurian strata layers) is used for the public water supply in several cities. In this aquifer there is HCO_3–Mg–Ca and HCO_3–Cl–Na–Mg–Ca type groundwater with TDS ranging from 0.5 to 1.5 g/L. However, in Latvia the aquifer system is considered as a passive water exchange zone and typically contains Na–Cl (sometimes Na–Cl–SO_4) saline water with TDS usually ranging from 3 to 10 g/L. In addition, the water has elevated trace element content (*viz*, Br, Bo, Se, Pb, Rb (Retike *et al.* 2016a)). Locally around Riga and Carnikava, a sharp salinity increase can be observed (up to 40 g/L) due to upward saline water intrusion near faults (Levins *et al.* 1998; Babre *et al.* 2016). There is a hypothesis that the Middle-Lower Devonian aquifer system in Western and Southwestern parts of Estonia contains glacial paleo-groundwater, but further studies are needed for conclusive evidence (Vaikmäe *et al.* 2021). While there is no direct evidence of glacial paleo-groundwater in Latvian side of the aquifer system, extraordinary groundwater chemical composition are observed in the Northern part of Latvia, having untypically low chloride content (<3 mg/L) in a combination with high barium concentrations. With some precautions, these observations might be used to estimate the groundwater age (Retike *et al.* 2016a). Water stable isotope studies, however, seems to exclude glacial meltwater presence, but the spatial sampling coverage is limited and the possibility to find glacial meltwater in Latvia is still viable (Babre *et al.* 2016). The Middle-Lower Devonian aquifer system is covered by Narva regional aquitard, which is mainly formed of marl and clay that separates the passive water exchange zone from the active water exchange zone (Popovs *et al.* 2015).

The Upper-Middle Devonian aquifer system is a very important drinking water source in Latvia and Lithuania as well as in Southern part of Estonia (Perens & Vallner 1997; Mokrik *et al.* 2009; Retiķe & Dēliņa 2018). The thickness of the aquifer system is typically 200–300 m but reaches up to ca. 650 m in the Northern part of Latvia. The lower part of the aquifer system is represented by terrigenous rocks – sandstones and siltstones with clay interlayers, while the upper part is made of both carbonate and terrigenous rocks – dolomite, marl, sandstone, siltstone, clays, and gypsum. The Upper-Middle Devonian system in Latvia and Lithuania is subdivided into several smaller aquifer systems because of many individual aquifers and aquitards. In Latvia and Lithuania Upper-Middle Devonian Amata-Aruküla (Šventoji-Arukiula in Lithuania), Upper Devonian Amulas-Pļaviņas and Permian-Famennian (Permian-Upper Devonian in Lithuania) aquifer systems are recognized (Mokrik 2003; Retiķe & Dēliņa 2018). Fresh Ca–Mg-HCO_3 groundwater with TDS values of 0.2–0.6 g/L with often elevated iron content (up to 1.5–1.7 mg/L) dominates the aquifer system (Perens & Vallner 1997). Due to dissolution of gypsum and water mixing SO_4–Ca–Mg fresh groundwater and SO_4–Ca brackish groundwater with TDS up to 2 g/L can be observed in central and western parts of Latvia. Such groundwaters are often accompanied by elevated content of trace elements: fluorine, strontium, lithium, and copper (Levins & Gosk 2007; Retike *et al.* 2016a). The main aquifer systems for water supply are Permian – Upper Devonian (limestone and dolomite) in Lithuania and Amata-Aruküla (mostly sandstone) in Latvia (Klints & Dēliņa 2012).

A Cenozoic-Mesozoic aquifer system is only found in Lithuania, and it is separated from the Permian aquifer by a Lower-Triassic regional aquiclude, which consists of clays and clayey marls. The main Cenozoic-Mesozoic aquifers are Jurassic, Cenomanian-Lower Cretaceous, Upper Cretaceous and Paleogene and Neogene aquifers. Almost all the Permian aquifers and the Triassic aquiclude are dipping in SouthWestern direction and mineralization increases in that direction and with depth. Generally, freshwater is not found below 300 m depth. The Cretaceous and Jurassic aquifers (carbonate and terrigenous rocks) cover a wide area in the Western and Southwestern Lithuania territory. However, the Jurassic aquifer is only sporadically distributed approximately in the same area. The Cretaceous aquifer contains Ca–Mg-HCO_3 freshwater and is the main water supply in the Western part of Lithuania.

The terrigenous Paleogene aquifer is rather small, and it is only specific for Southern Lithuania. The Neogene aquifer is sporadically distributed in the Eastern part and forms a united hydraulic system with the Quaternary aquifers (Mokrik 2003).

The whole territory of BAB is covered by Quaternary, mostly glacial and marine sediments, and its structure is very heterogeneous (Popovs *et al.* 2015). The thickness of Quaternary deposits varies from a few meters up to 300 m in the areas of buried valleys (Popovs *et al.* 2022). Such valleys supply, for instance, drinking water to Daugavpils, which is the 3rd largest city in Latvia. Quaternary intermorainic aquifers that are used as groundwater sources in the Western and Southeastern part of Lithuania, where Quaternary aquifers consist mainly of glaciolacustrine sand and fluvioglacial gravel deposits. The main cities – Vilnius and Kaunas, are supplied from intermorainic and alluvial aquifers (Klimas & Plankis 2007). Due to the shallow occurrence, the Quaternary aquifers are used for groundwater supply in rural areas where inhabitants exploit groundwater from shallow wells (Klavins *et al.* 1996; Retike *et al.* 2016b).

In Baltezers vicinity in Latvia, the Quaternary aquifers contain exceptionally good quality HCO_3–Ca–Mg freshwater with TDS less than 200 mg/L and low iron content (<0.3 mg/L). This aquifer is therefore an important resource for the capital Riga (Retike *et al.* 2022). Around 35% of the water supply in Riga comes from the Quaternary aquifer which is used in combination with managed aquifer recharge from lake Mazais Baltezers (Eynard *et al.* 2000; Lace *et al.* 2017; Retiķe & Dēliņa 2018).

Concerns related to geogenic origin

Iron and manganese. Groundwater abstracted from Upper to Middle Devonian aquifers in Latvia and Lithuania, and from the Middle Devonian aquifer system in South Estonia, are naturally rich in iron (Retike *et al.* 2016a; Albrektienė *et al.* 2019; Karro *et al.* 2020). According to the EU Drinking Water Directive (European Communities 1998) the total iron (Fe_{tot}) content in drinking water should not exceed 0.2 mg/L. This threshold is often exceeded in the Baltic region. Total iron concentrations above the threshold have been reported in 81% of the analyzed samples in Estonia (Karro *et al.* 2020), in 88% of wellfields that reported the water quality and abstracted fresh groundwater in Latvia (Retiķe & Caune 2012), and in about 87% of all investigated groundwater sources in Lithuania (Diliūnas *et al.* 2006). The total iron content in Baltic freshwater aquifers varies. For instance, Fe_{tot} median concentrations in Latvian freshwater aquifers range from 0.2 to 3.3 mg/L (Retike *et al.* 2016a), while in Estonian Middle Devonian aquifer the median value is 0.6 mg/L (Karro *et al.* 2020). The lowest iron concentrations have been observed in recharge areas and some springs associated with oxic conditions, while the highest concentrations are in discharge areas and reducing groundwater conditions (Retike *et al.* 2016a; Karro *et al.* 2020). In South Estonia the Fe_{tot} concentrations can reach up to 26 mg/L and are associated with strongly reducing conditions. Leaching tests indicate that higher iron content is partly controlled by the granulometric composition of terrigenous rocks where large pores enhance water circulation and iron leaching (Karro *et al.* 2020).

Fluorine, boron, and barium. Health problems (dental fluorosis) arising from excess of fluorine (F) intake have been recorded in Estonia (Indermitte *et al.* 2009, 2014). The Silurian–Ordovician aquifer system in Western Estonia can be delineated as an anomaly where high F content is associated with HCO_3–Cl–Na groundwater type, deep wells and long residence time in water-bearing carbonate rocks (especially marlstones and domerites), and presence of K-bentonites and illite/illite-smectite (Karro & Rosentau 2005; Haamer & Karro 2006; Karro *et al.* 2006, 2009; Uppin & Karro 2012, 2013; Karro & Uppin 2013). In Western Estonia, natural F concentrations in groundwater can reach 7 mg/L (Karro *et al.* 2006), which exceeds the EU drinking water standards of 1.5 mg/L by a factor of almost five (European Communities 1998). The anomalies of F of natural origin are well known in Permian and Upper Devonian aquifers of Western Lithuania, where fissured limestone and dolomite contain HCO_3–Na (with low Ca^{2+} content) groundwater type and F concentrations can reach 6.5 mg/L, while average F concentration in Lithuanian freshwater aquifers is around 0.4 mg/L.

High concentrations of boron (B > 1 mg/L) are detected in Upper Devonian, Permian, and Jurassic aquifers in (mostly) Western Lithuania (Klimas & Mališauskas 2008), while sporadic B anomalies occur in Nemunas, Neris, and Šešupė valleys. The sources of high B concentration are buried salinities or saline water leaking from deep aquifers. In Latvia the highest boron concentrations (B > 0.2 mg/L) have been detected in Cl^- or SO_4^{2-} rich groundwater (Retike *et al.* 2016a).

High barium (Ba) concentrations (up to 6.37 mg/L) are detected in the Cambrian–Vendian aquifer system in Northeastern Estonia (Marandi *et al.* 2004). Even though Ba content in drinking water is not restricted according to national regulations, these anomalies should not be ignored. A potential source might be ion exchange in combination with low SO_4^{2-} content in places where groundwater comes from the upper weathered and fissured portion of the crystalline basement into the overlying Cambrian–Vendian aquifer system (Marandi *et al.* 2004). The highest Ba concentrations (>0.5 mg/L) in Latvia are associated with confined aquifers that contain groundwater with extremely low SO_4^{2-} and Cl^- concentrations (SO_4^{2-}<1.5 mg/L and Cl^-<4 mg/L, Retike *et al.* 2016a). Such anomalies can be observed in the Upper Devonian and Permian carbonate aquifers; in the Southeast part of Latvia, near Daugavpils where paleo valleys are present; and in the Middle Devonian aquifers in Northeastern Latvia (Babre *et al.* 2016; Vaikmäe *et al.* 2021).

Arsenic. Arsenic (As) concentration above EU drinking water standard of 10 μg/L (European Communities 1998) has been discovered in Southwestern, Western and Northern parts of Lithuanian aquifers at depths of 20–60 m (Figure 6). Research rules out the anthropogenic pollution and states that the main source of aqueous arsenic in groundwater could be the solid ferric oxides/hydroxides and possibly bonded As species on thiol (sulfhydryl) groups that are present in peat and gyttja material (Herath *et al.* 1997). There are also indirect indications of reductive leaching of As species to the aqueous phase in aquifers that are enriched with organic material (Pūtys *et al.* 2021). Slightly elevated As values (>5 μg/L) in Latvia have been observed in aquifers with strongly reducing conditions, in combination with the highest Ba, Fe_{tot}, Si and NH_4^+ values (Retike *et al.* 2016a). Arsenic values above the threshold have also been recorded in Southern Estonia (Middle Devonian aquifer system) in 2017 at depths around 50–60 m and likely have a natural origin.

Sulfates. In Northern Lithuania, shallow karstified Upper Devonian groundwater aquifers are naturally of low quality (SO_4^{2-}>1,000 mg/L, hardness >30 mg-eq/L) and requires pre-treatment. Due to economic reasons, groundwater abstraction from shallow aquifers is often the preferred option for the installation of new private wells. This situation leads to increased groundwater abstraction which may intensify the already active karstic processes in the region (Arustienė 2021). In the Central part of Latvia, groundwater quality of the Upper Devonian aquifers is also poor due to elevated SO_4^{2-} content (SO_4^{2-} 350–1,200 mg/L) and water hardness derived by gypsum dissolution in the water-bearing aquifers (Retike *et al.* 2016a). The karst processes in Latvia are less active than in Lithuania, probably due to the thick and continuous overlying Quaternary sediments in Latvia.

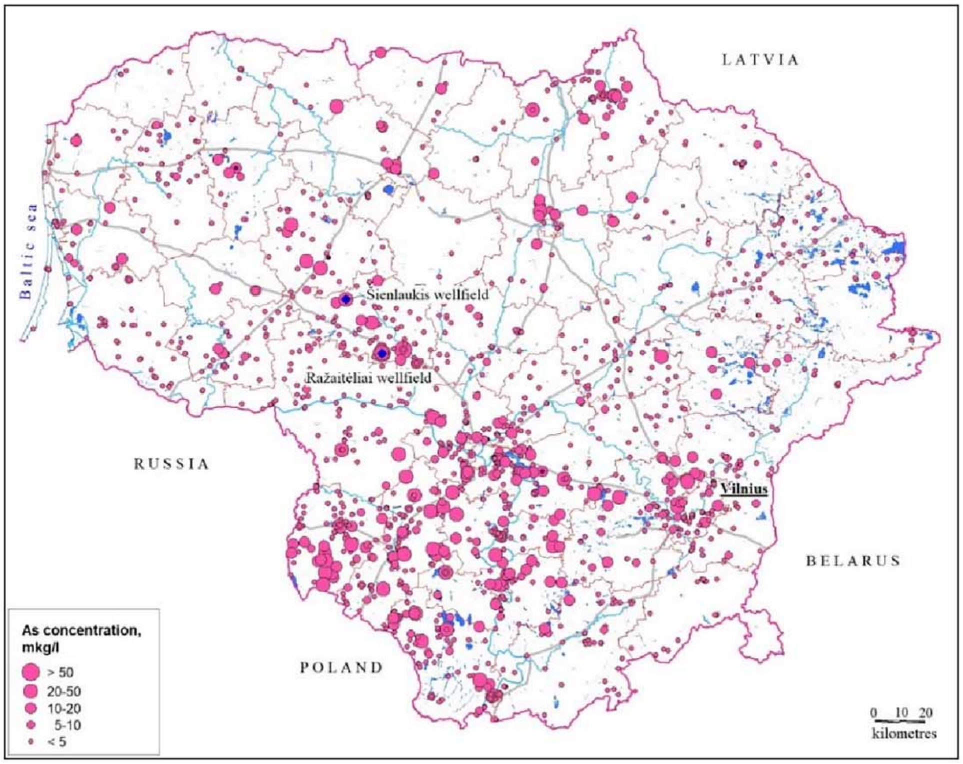

Figure 6 | Arsenic concentrations (μg/L) in drinking water wells of Lithuania (LGS 2021). Please refer to the online version of this paper to see this figure in color: http://dx.doi.org/10.2166/nh.2022.018.

Concerns related to intensive groundwater pumping

Saltwater intrusion. An intensive groundwater abstraction in Northern Estonia has led to the formation of regional depressions in potentiometric levels and consequent deterioration of groundwater quality (with TDS up to 1,500 mg/L) in the Cambrian–Vendian aquifer system (Perens & Vallner 1997). A geochemical study from the Kopli peninsula (Tallinn) ruled out the lateral seawater intrusion as a source for salinity, while the theory of saline deep-seated groundwater up-coning from underlying weathering core of the fractured crystalline basement, which hosts brackish Cl–Ca groundwater, was supported by isotope studies (Karro *et al.* 2004). An extensive depression cone was formed in the vicinity of the Latvian capital Riga during the 1970s due to overexploitation of major Upper Devonian Gauja-Amata freshwater aquifers (Klints & Dēliņa 2012). The over abstraction was followed by an increased groundwater salinity. Similar to Estonia, the source of salinity is most likely not related to seawater intrusion, but rather to upward migration of deeper located brackish aquifers through tectonic faults (Kalvāns 2012). Increased Cl and Na concentrations in the Cretaceous aquifer system can be found in Southwestern Lithuania and in the Nemunas valley in the Southern part of Lithuania (especially near Druskininkai and Birštonas towns) and are a result of saline water intrusion from deeper aquifers. Moreover, the groundwater abstraction could intensify the water mixing and increase groundwater salinity (Arustienė 2021).

Seawater intrusion. An increased groundwater salinity has been observed since the 1930s in the Upper Devonian Mūru-Žagares aquifer beneath the third largest city of Latvia, Liepaja. The aquifer outcrops at the bottom of the Baltic Sea, approximately 5 km from the coast, and the hydrogeological conditions are favorable to direct seawater intrusion (Bikše & Retike 2018). Cl content up to 1,800 mg/L are observed in the central part of the groundwater body (Retike & Bikše 2018; Pulido-Velazques *et al.* 2022).

Water mixing. Intensive groundwater pumping from the Cambrian–Vendian aquifer system in the central part of the Viimsi peninsula, located Northeast of Tallinn, has led to the lowering of potentiometric levels and the increase of Cl^- concentrations and enrichment of naturally light isotopic ($\delta^{18}O$) composition of glacial paleogroundwater in the aquifer system. Changes in groundwater quality are caused by the complex water mixing between glacial paleo-groundwater, water from the underlying crystalline basement and modern groundwater recharging from the buried valley in the Southwestern part of the peninsula (Raidla *et al.* 2019). The water mixing has led to an increased radium (Ra) activity. Raidla *et al.* (2019) propose two sources for these increased Ra activities in the aquifer system – the crystalline basement and the secondary uranium deposits in sedimentary sequence originating from paleo-redox-front in the southwestern part of the peninsula developed in the Pleistocene. The groundwater in the Cambrian–Vendian aquifer system on the Viimsi peninsula contains up to 600 mBq/L of ^{226}Ra and 800 mBq/L of ^{228}Ra. Such activity concentrations in drinking water lead to an effective dose above 0.5 mSv/year. The Viimsi water treatment facility was designed for Fe, Mn and Ra removal. However, in case of significant increase in Ra activity concentrations, its ability to decrease the total indicative dose below 0.10 mSv/year is questionable (Suursoo *et al.* 2017). An increased salinity (Cl^- >250 mg/L) has been recently observed in the Gdov aquifer, which is used for drinking water abstraction in Northeastern Estonia, that typically contained fresh meltwater from the last ice age more than 12,000 years ago. Due to increased groundwater abstraction, the limited amount of freshwater in the aquifer is being replaced by initial saline groundwater (formerly seawater) that was pushed away during the last ice age. The overlying Voronka aquifer is also overexploited and highlights the importance of conceptual understanding of baseline quality and governing of hydrogeochemical processes to support sustainable groundwater management (Marandi *et al.* 2012).

Concerns related to agriculture

The reducing conditions in the Baltic aquifers provide favorable conditions for denitrification. The fractured aquifers on the other hand, without thick and clayey overlying Quaternary sediments, are vulnerable to agricultural pollution (Højberg *et al.* 2017; Koit *et al.* 2020; Kalvāns *et al.* 2021). A total area of 3,250 km^2 was established in the Central Eastern part of Estonia (Pandivere and Adavere-Põltsamaa nitrate vulnerable zone, NVZ) to comply with the EU Nitrate Directive (91/676/EEC) and the Water Framework Directive (2000/60/EC). The average nitrate (NO_3^-) value above the 50 mg/L threshold was recorded in more than 15% of groundwater monitoring points in the NVZ area (Leisk 2021).

Highest NO_3^- concentrations in Latvian groundwater can be observed in shallow wells up to 5 meters and springs. Even though the NVZ has been set for the central part of Latvia and covers 12.8% of the country, elevated nitrate concentrations can be found in groundwater both inside and outside of the NVZ (Retike *et al.* 2016b; Nitrate report 2020). Nitrate concentrations in Latvian groundwater rarely exceed the EU threshold of 50 mg/L, but they are often above the natural baseline level of 4 mg/L according to Retike *et al.* (2016a), especially in springs outflowing from fractured aquifers. They can thus deteriorate vulnerable groundwater dependent ecosystems and be harmful for public health if consumed (Kalvāns *et al.* 2021).

The whole territory of Lithuania has been designated as NVZ and natural baseline levels are exceeded for NO_3^- (1.5 mg/L), NH_4^+ (0.33 mg/L) and phosphates, PO_4^{3-} (0.08 mg/L). In intensively cultivated arable lands and urban areas, high average concentrations are recorded of NO_3^- (28 mg/L), NH_4^+ (1.5 mg/L), and PO_4^{3-} (0.12 mg/L). Nitrates above EU drinking water standard of 50 mg/L (European Communities 1998) were observed in 10% of all groundwater monitoring stations in Lithuania. Deterioration of groundwater quality due to poorly treated wastewater infiltration into deeper productive aquifers has also been observed near the largest Lithuanian cities Vilnius, Kaunas, and Klaipėda.

Groundwater in Denmark

All drinking water in Denmark originates from groundwater. The tap water is not treated with chlorine or any other chemicals (Evlampidou *et al.* 2020; Thorling *et al.* 2021). The groundwater aquifers consist of either unconsolidated sands and gravels or fractured limestone and chalk, except on Bornholm south of Sweden, where the hydrogeology resembles the Swedish geology, due to the crystalline bedrock. The Quaternary deposits in Denmark are approximately 50–200 m thick. These unconsolidated sediments are underlain by Paleogene and Neogene deposits or Cretaceous limestone and chalk. The basement has a huge impact on the geochemical composition of the overlaying Quaternary deposits. The widespread chalk layers give rise to neutral groundwater with pH around 7.3–7.5, buffered from $CaCO_3$ molded into the Quaternary layers in most of the country. Only at the west of Denmark, groundwaters with low pH of 4.5–6.3 can be found. The reason is that this part of Denmark was not covered by ice during the last glaciation, and the pre-Quaternary layers consist of Miocene sands. This has great impact on the natural solubility of metals and the carbonate system of the groundwater. In all parts of the country saltwater can be found in deep groundwater, and the freshwater layers are mostly found in the upper 100 m or less. Average chemical composition of groundwater in Denmark is given in the Supplementary Material.

The most important natural substances that pose a threat to drinking water quality are geogen arsenic and fluoride, with a geochemistry that resembles that of the Baltic countries. At most waterworks the natural contents of iron, manganese, and sulfide in reduced groundwater can be removed by aeration and filtration, and these substances are thus not generally considered problematic. High levels of arsenic in groundwater are also removed and precipitated at the waterworks by use of natural or added iron compounds (Ramsay *et al.* 2021).

Besides nitrates, pesticides are the most widespread pollutant in Danish groundwater. Some pesticides have been regulated since the 1990s and signs of decreasing impact on the groundwater quality can now be found (Thorling *et al.* 2021). Pesticides and their metabolites were detected in 72% of the national monitoring wells and 51% of the water work wells in 2021 (Thorling *et al.* 2021). 39% of the wells and 15% of the water works had concentrations above 0.1 μg/L, which is the threshold value for drinking water and groundwater (EU 2006).

Nitrate abatements

Since the 1980s the nitrogen (N) application in Danish agriculture has been regulated. Time series from 2009 to 2016 in shallow oxic groundwater (down to 5 m below the ground surface) show deterioration in the last 8 years of the monitoring period (Figure 7). In 2016, catchments with mainly sandy sediments had nitrate concentrations in groundwater exceeding the regulation standard of 50 mg/L for approximately 70% of the monitoring points. In loamy catchments, approximately 8% on average, the monitoring points exceeded the regulation standard.

The nitrate observations in deep oxic groundwater (down to 50 m below the ground surface) show delayed effect of the N-regulation due to longer flow paths than near-surface groundwater. Infiltration time for deep groundwater may be up to 50 years. Results showed a nitrate trend reversal in the beginning of the 1980s (Figures 7 and 8). In 2016, approximately 40% of the monitoring points had exceedance of the groundwater standard of 50 mg/L nitrate. Significant downward nitrate trends of 72% (39% statistically significant) and 80% (49% statistically significant) are found in the monitoring points for the latest infiltration periods 1985–1998 and 1999–2014, respectively (Hansen *et al.* 2017).

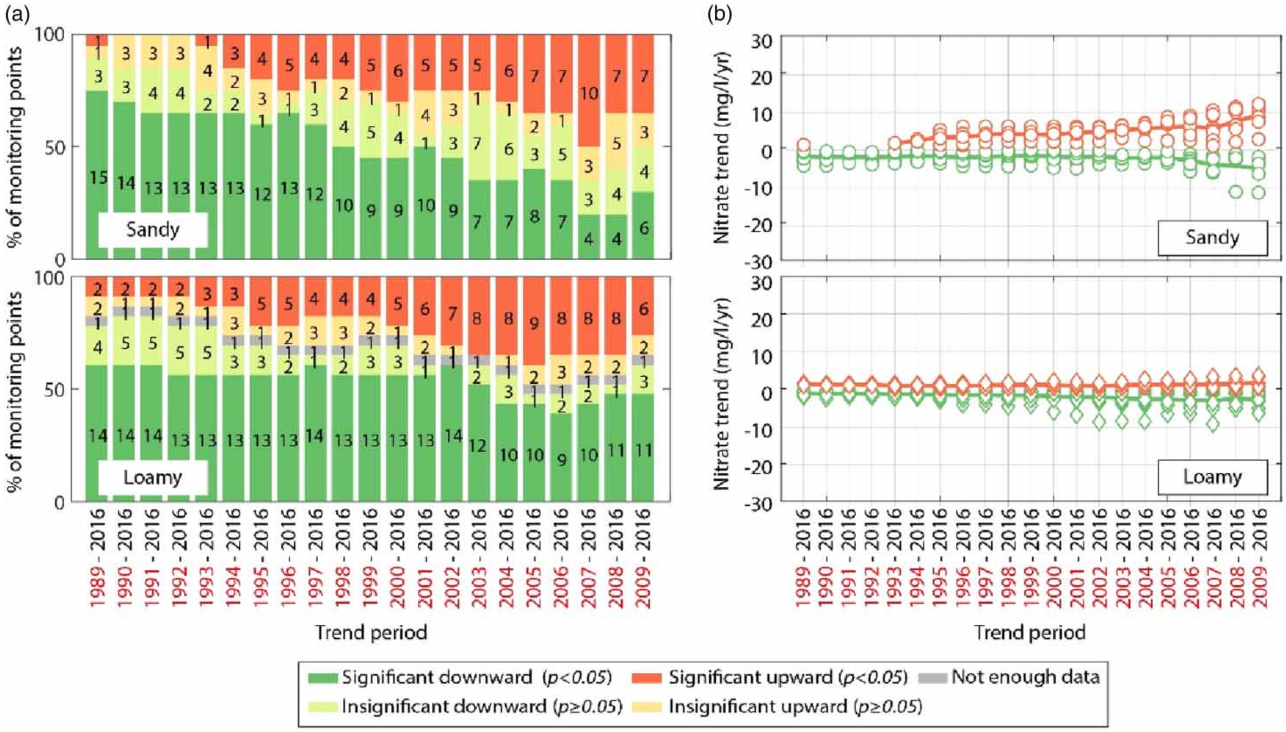

Figure 7 | Backward groundwater nitrate trends for 21 sampling periods from 2009–2016 to 1989–2016 in shallow oxic groundwater. The number in the columns shows the number of monitoring points (Hansen *et al.* 2019). Please refer to the online version of this paper to see this figure in color: http://dx.doi.org/10.2166/nh.2022.018.

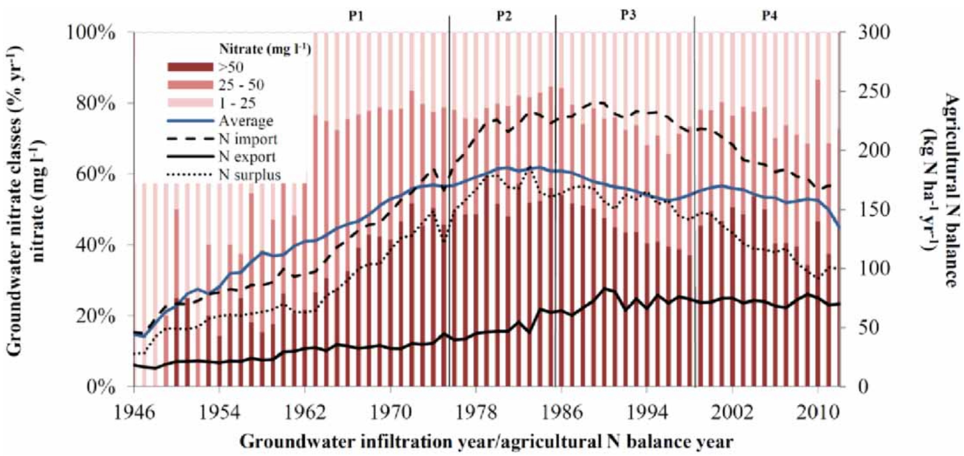

Figure 8 | The annual trends of nitrate in oxic groundwater taking infiltration time into account using groundwater dating, and agricultural N balances for Denmark. The 5-year moving average of the nitrate content in oxic groundwater (blue line) is based on 5,506 samples from 340 monitoring points (Hansen *et al.* 2017). Please refer to the online version of this paper to see this figure in color: http://dx.doi.org/10.2166/nh.2022.018.

On a national basis, the effect of agricultural N management on nitrate concentrations in oxic groundwater was also evaluated by analyzing the relationship between nitrate in oxic groundwater (5-year moving average) and annual N surplus, N-use efficiency (NUE) and the gross domestic product, GDP (Figure 9).

Even though these 'one-fit-all' regulations lowered the level of N pollution in groundwater and improved the aquatic environment, national goals and EU legislation for environmental standard have not yet been reached in all parts of Denmark. Cost-efficient and targeted N-regulations have therefore been suggested as a way forward (Jacobsen & Hansen 2016). The idea is that the local N-regulation of a field is adjusted to the natural denitrification potential of the subsurface.

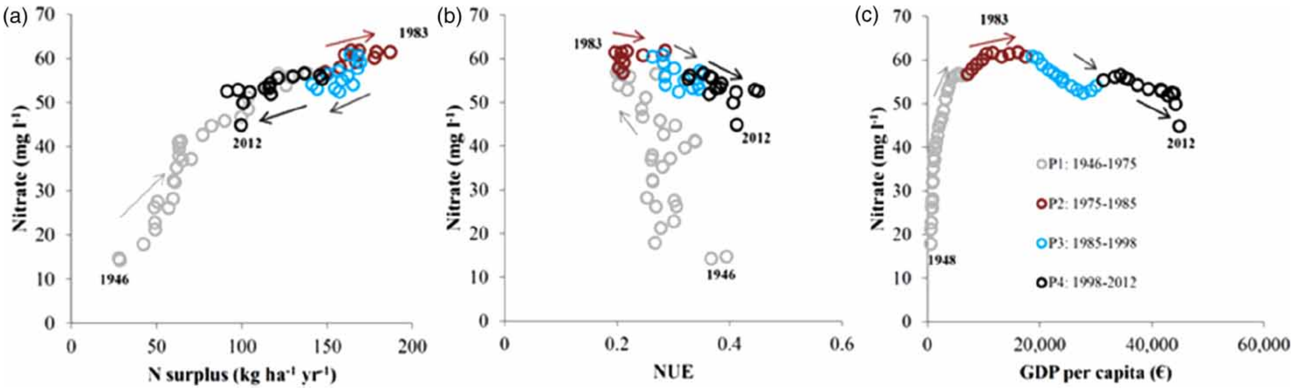

Figure 9 | Nitrate trend reversals in oxic groundwater in four groundwater infiltration periods. (a) Annual N surplus in Danish agriculture. (b) Annual NUE in Danish agriculture. (c) Annual Danish gross domestic product per capita (GDP per capita) in euro (€) (Hansen *et al.* 2017). Please refer to the online version of this paper to see this figure in color: http://dx.doi.org/10.2166/nh.2022.018.

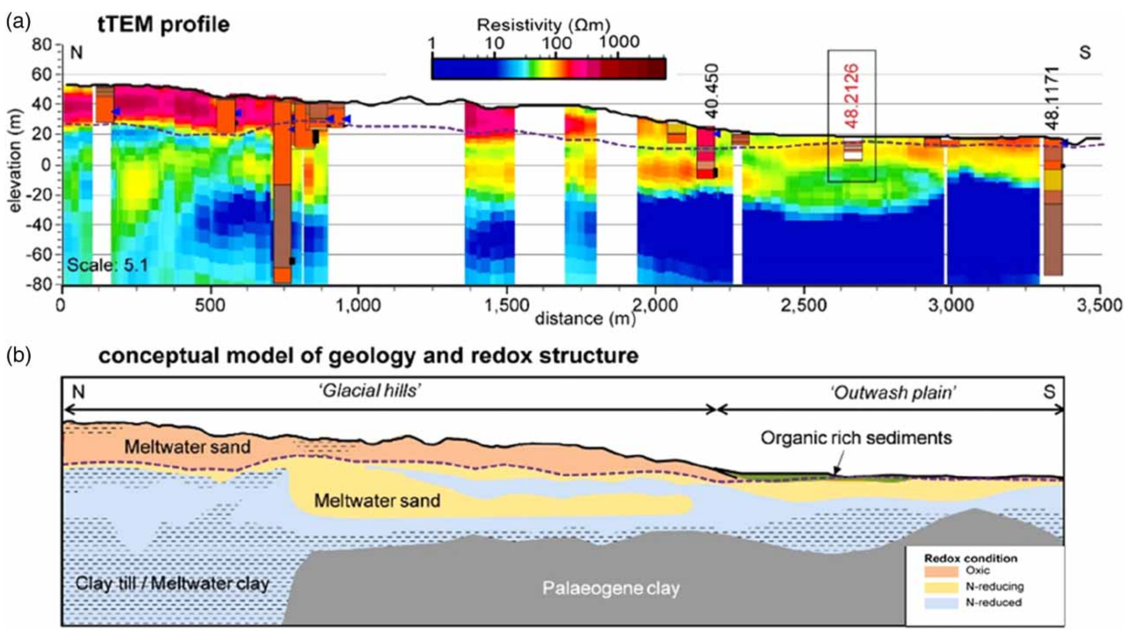

Figure 10 | Detail mapping of hydrogeochemical conditions for future targeted N-regulation of agriculture from the MapField (2021) project. (a) Profile with electrical resistivity data from the towed transient electromagnetic system (tTEM). (b) Geological structures and redox zones based on interpretation of data shown in (A) (Hansen *et al.* 2021). Please refer to the online version of this paper to see this figure in color: http://dx.doi.org/10.2166/nh.2022.018.

The denitrification potential depends on the water pathways, the N reduction rates, and the capacities of the subsurface media below the field. The new concept is based on detailed knowledge about local hydrogeochemical conditions (Hansen *et al.* 2021; MapField 2021). Geophysical surveys and geochemical investigation wells can be used to assess hydrogeological structures and redox zones (Figure 10, Auken *et al.* 2019). Such investigations give knowledge-based decisions for targeted N-regulations (Hansen *et al.* 2021).

Hydrogeology in Iceland

The Icelandic bedrock has different hydrogeological properties from one place to another, mainly dependent on age. The overall permeability reflects the relative age of the rock formation. The oldest bedrock is found in the Miocene regions in the west, north, and east of Iceland (Figure 2). These bedrocks have low permeability with fluctuating direct runoff to rivers and small springs. In this area, the richest aquifers belong to the unconsolidated overburden such as alluvial and glaciofluvial deposits and rockslides debris. The Pliocene and Pleistocene bedrock formations, which border the active volcanic belts, have higher permeability, larger springs, and spring-fed rivers. The Holocene lava fields, with their central volcanoes and associated fissure swarms, have exceptionally high permeability. The strong flow of groundwater emerges in large areas of springs at the borders of the lava fields or in connection with the fissure swarms.

The largest surficial deposits in Iceland are the sandur (glacial outwash plains) which border the major ice caps, especially Vatnajökull, Mýrdalsjökull, and Hofsjökull. High-permeability aquifers with large volumes of storage are fed partly by direct infiltration of liquid water at the glaciers bed and partly by infiltration from the complex network of glacier rivers which flow across the sandur. An overwhelming majority of Icelandic waterworks use pure groundwater, some from springs and others from boreholes. The tap water is not treated with chlorine or any other chemicals. 97% of the domestic water consumption is groundwater (Egilson & Steinþórsdóttir 2014).

Geothermal water and geothermal energy are vital for Iceland, and geothermal springs can be found in most districts in Iceland. The geothermal fields are often divided into high-temperature and low-temperature fields. Power plants rely on the high-temperature fields, while the residential space heating, greenhouse industry and fish farming rely on the low-temperature fields. About 85% of all houses in Iceland are heated with geothermal energy. In 2015, the total electricity consumption in Iceland was 18,798 GWh. Renewable energy provided almost 100% of the electricity production, with about 73% coming from hydropower and 27% from geothermal power (Ragnarsson *et al.* 2020).

The bedrock of Iceland is relatively young (0–16 Ma), and the lithology is rather uniform, entirely built up of volcanic rocks with interbeds of sedimentary layers of volcanic origin. Therefore, the water chemistry is also quite uniform. The main chemical characteristics of the cold groundwater in Iceland is the low content of dissolved solids. Analysis of water from 79 aquifers in Iceland demonstrated the average TDS as 75 mg/L (Gunnarsdottir *et al.* 2015, 2016). The concentrations of all elements in solution are well within the limits of European and American standards. Bulk water export from Iceland could be a profitable industry in the future, but currently only a minor production is carried out. Average geochemical composition of Icelandic groundwater is given in the Supplementary Material.

DISCUSSION AND CONCLUSIONS

Statistics of freshwater use and groundwater abstractions show significant differences in the Nordic Region (Table 1). In the following, we briefly discuss these differences in the light of water balance, demography, and geology.

Table 1 | Estimates of total freshwater withdrawal, Tfw (km^3/year)

	Tfw (km^3/year)	Grw (%)	Municipal (%)	Industrial (%)	Agricultural (%)	Estimated year
Denmark[a]	0.69	97	55	10	35	2020
Estonia[b]	1.82	12	25–56	39–73	2–5	2018
Finland[c]	1.87	13	14–21	76–84	3	1999
Iceland[d]	0.17	97	29–34	66–71	0–0,1	2003
Latvia[e]	0.20	53	37–55	33–35	12–28	2018
Lithuania[e]	0.29	57	50–78	15–27	7–23	2018
Norway[f]	3.05	2	12	87	1.4	2006
Sweden[g]	2.44	13	23	74	3	2015

Groundwater abstraction, Grw, municipal, industrial, and agricultural consumption given as the percentage of Tfw. Municipal consumptions for Norway and Sweden were specified as domestic consumption. Except for Denmark, Norway and Sweden, data for municipal, industrial, and agricultural freshwater consumption are taken from The World Factbook (2021) and Wikipedia (2021a).

[a]*Source:* GEUS (2021).

[b]*Source:* Statistics Estonia (2021).

[c]*Source:* Statistics Finland (2018); Wikipedia (2021a).

[d]*Source:* Egilson & Steinþórsdóttir (2014); Wikipedia (2021a).

[e]*Source:* Eurostat (2021a).

[f]*Source:* Statistics Norway (2008).

[g]*Source:* Statistics Sweden (2015).

Groundwater abstraction and total freshwater withdrawal

Iceland and Norway have the highest specific runoff in the Nordic Region, but the use of groundwater is quite different. For the Norwegian part, the groundwater abstraction as a fraction of domestic water consumption is between 10 and 15%, which is significantly higher than 2%. In spite of the uncertainties, Table 1 illustrates some geological differences between the countries. Iceland, for example, has unconsolidated deposits and young volcanic rocks with high permeability, while most part of mainland Norway has low permeable till and marine clay deposits above an old crystalline bedrock with a modest water yield. Groundwater abstraction is more important for Sweden and Finland compared to Norway (Table 1). Most of the groundwater is taken from high permeable Quaternary deposits which are more abundant in Sweden and Finland than in Norway, and groundwater abstraction for domestic water supply is important in Sweden and Finland, especially in rural areas.

The reported values indicate a groundwater fraction of total freshwater withdrawal of more than 50% for Latvia and Lithuanian (Table 1). The total freshwater withdrawal in Estonia has been reduced from the 1990s, and the fraction of groundwater usage is probably higher than indicated in Table 1. For Latvia and Lithuania, the total freshwater withdrawal is low compared to estimated runoff. The corresponding fraction for Estonia is the highest in the Nordic Region (Table 2), but the total freshwater abstraction for Estonia includes most likely cooling water for power plants, water used in the mining industry, and fish farming.

Denmark's geological setting and groundwater consumption are more similar to the Baltic countries than the Fennoscandian countries. The groundwater fraction of total freshwater consumption is estimated to about 97% (Table 1), and the total freshwater withdrawal is estimated to about 4% of total runoff (Table 2). Around 40% of Denmark's freshwater consumption is allocated to agriculture. The other countries allocate relatively more freshwater to industry (Table 2). This is particularly evident for Norway where hydropower is the dominant part of the energy consumption. It is necessary to emphasize that the values in Tables 1 and 2 were derived from different statistical sources. The reported numbers should therefore be interpreted with some precautions. The procedures for reporting water usage in the EU system strives towards consistent definitions, and the purpose with Tables 1 and 2 was to apply the specified definitions as far as possible. With the increasing pressure on water resources, it is important to apply consistent reporting procedures. The apparent discrepancies between countries in Tables 1 and 2 illustrate the need for consistent interpretations of the EU guidelines for reporting water consumption statistics.

Groundwater chemistry

Groundwater chemistry for all countries in the Nordic Region is compiled in a main table in the Supplementary Material. The median concentrations of major ions in groundwater were highest in the sedimentary aquifers in the Baltic Artesian Basin

Table 2 | Average specific runoff (R, mm/year), runoff volumes (Rv, km^3/year), fraction total freshwater withdrawal of average runoff (Tfw/Rv, %), and statistics of countries areal size (km^2) and population

Country	R mm/year	Rv km^3/year	Tfw/Rv[a] %	Area[a] km^2	Population[a] million
Denmark[b]	381	16.34	4.2	42,933	5.85
Estonia[c]	262	11.88	15.3	45,339	1.33
Finland[d]	300	101.53	1.8	338,424	5.50
Iceland[e]	1460	150.05	0.1	102,775	0.37
Latvia[b]	567	36.64	0.6	64,589	1.91
Lithuania[b]	341	22.27	1.3	65,300	2.78
Norway[f]	1140	367.77	0.8	322,601	5.40
Sweden[b]	434	195.33	1.3	450,295	10.40

Estimated total freshwater withdrawal (Tfw) is given in Table 1.
[a]Wikipedia (2021b).
[b]*Source:* Eurostat (2021b).
[c]*Source:* Vallner & Metslang (1970).
[d]*Source:* Vakkilainen (2016).
[e]*Source:* Jónsdóttir (2007).
[f]*Source:* Beldring *et al.* (2002).

and in chalk and limestone aquifers in Denmark. Fennoscandian bedrocks aquifers yielded groundwater with higher ion concentrations than found in Iceland. The compiled data indicate dominance of Ca–HCO_3 type groundwater in the Fennoscandian bedrock, whereas the sedimentary aquifers in the Baltic region have deep groundwater with high sulfate content and high salinity which indicate mixing of water with different geological origins. Groundwater in Iceland has low content of dissolved solids. This is probably an effect of Iceland's young rocks with high permeability combined with high annual precipitation, which give high throughflow of water and less contact between soil, bedrock, and water. The opposite is true for deep groundwater in Denmark and the Baltic Artesian Basin. Despite the great variability in the Nordic Region, some general conclusions can be drawn:

- Residence time of water in the aquifer and the mineral content of the solid aquifer material explain the main chemical character of groundwater in highly permeable Quaternary deposits in the Nordic Region. The chemical composition of precipitation contributes less but is important for marine input in coastal areas.
- Root respiration and decay of organic matter increase concentration of CO_2 in the soil atmosphere, which in turn reduce pH and makes the soil solution effective in mineral weathering and produce bicarbonate rich water. In addition, organic acids leach down in the soil profile.
- Weathering of silica minerals is important for water quality especially in the Fennoscandian countries where silica-rich rocks and sediments are abundant. In carbonate-sparse environment, dissolution of silica minerals is an important pH-buffer. Because weathering of silica minerals is slow, silica-rich environments are sensitive to acidification.
- The reduction and oxidation potential (redox) of the subsurface govern the natural concentrations of dissolved oxygen, nitrate, iron, sulfur, and methane in groundwater. The nitrogen management example from Denmark illustrates the importance of local knowledge of redox conditions to improve groundwater quality.
- Mixing of water with different origin occur in coastal aquifers and in geological formations with interface between porewater with high salinity and infiltration of freshwater. In areas with extensive groundwater extraction, mixing of water qualities may pose a risk for drinking water quality.
- The average chemical composition of groundwater in the Nordic Region depends on the geological setting, i.e. the mineral abundance, surface area, and reactivity. Groundwater in shallow Quaternary deposits has typically low concentrations of ions, but the general chemical character resembles the local bedrock. Carbonate dissolution is fast, so if these minerals are present, the groundwater will have a high content of HCO_3^- and Ca^{2+}. The same is true in bedrock aquifers of limestone and chalk. In deeper groundwater, the concentration of dissolved minerals normally increases. Examples from the Baltic Artesian Basin show that in most cases, the geochemical composition reflects the lithology of the aquifer, but the pattern is not always so evident in the Fennoscandian bedrock.
- Groundwater is the only alternative for drinking water supply for many residents in the Nordic Region. It is therefore important to protect groundwater from anthropogenic contamination or hazards due to geogenic processes.
- Human activity modifies the natural water chemistry. Agriculture and industry may cause local problems, especially for unconfined aquifers. Examples are given from Denmark and the Baltic countries, but the list is by no means complete.
- Finally, hydrogeochemical management requires cross-disciplinary teamwork which includes knowledge of hydrology and residence time of water in the subsurface, understanding of weathering and soil chemical processes, insight into the geological framework, and expertise on legislation and public dissemination. With such expertise involved, carefully designed monitoring programs can be implemented, and knowledge-based management action can be undertaken.

Future challenges

The Nordic Region comprises a large variation with respect to geological setting, climate conditions, and demographical situation. These factors affect groundwater utilization and groundwater quality in numerous ways from economic benefits to complex interactions with the ecosystem. Many of these challenges are incorporated in the EU legislation and to some extent implemented in national and regional research and monitoring programs. The Nordic Region has valuable resources in this context, and the Region offers interesting opportunities for future cooperation. As a final remark, we take this opportunity to highlight some research challenges that deserve more attention. Global warming and change in land-use will affect groundwater quantity and quality – directly or indirectly. Of that reason, groundwater flow and hydrogeochemistry needs to be integrated in large-scale Earth system modeling. Efforts are made to include impact of land-use and human activity in such large-scale ocean–atmosphere–land simulations, but there is a significant research challenge to include the spatial heterogeneity of the geosphere in such models. The specific challenge is to minimize the modeling uncertainties. To achieve conceptually meaningful results, empirical control is a cardinal point. Geochemical data and tracer experiments are examples of such empirical input. The big challenge is the coupling of processes on different scales, from large-scale climate systems to micropore processes. The demand for basic research, however, comes easily in the shadow of more urgent problems. Here, it is enough to mention the needs for more knowledge on utilization of geoenergy and aquifer thermal energy storage; geohazards (*viz*, landslides, quick clay); urban hydrogeology (*viz*, subsurface infrastructure; land subsidence); tunnel constructions (*viz*, water leakage and local water balance); contaminant transport and subsurface remediation capacity; water supply; and water quality issues. All these topics are examples of the great societal demand for more knowledge on groundwater and groundwater interaction with the environment.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The authors declare there is no conflict.

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