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The chemistry of bottled mineral and spring waters from Norway, Sweden, Finland and Iceland

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ABSTRACT

Twenty-two bottled mineral and spring waters from Norway, Sweden, Finland and Iceland have been analysed for 71 inorganic chemical parameters with low detection limits as a subset of a large European survey of bottled groundwater chemistry (N = 884). The Nordic bottled groundwaters comprise mainly Ca-Na-HCO₃-Cl water types, but more distinct Ca-HCO₃, Na HCO₃ and Na-Cl water types are also offered. The distributions for most elements fall between groundwater from Fennoscandian Quaternary unconsolidated aquifers and groundwater from Norwegian crystalline bedrock boreholes. Treated tap waters have slightly lower median values for many parameters, but elements associated with plumbing have significantly higher concentrations in tap waters than in bottled waters. The small dataset is able to show that excessive fluoride and uranium contents are potential drinking water problems in Fennoscandia. Nitrate and arsenic displayed low to moderate concentrations, but the number of samples from Finland and Northern Sweden was too low to detect that elevated concentrations of arsenic occur in bedrock boreholes in some regions. The data shows clearly that water sold in plastic bottles is contaminated with antimony. Antimony is toxic and suspected to be carcinogenic, but the levels are well below the EU drinking water limit. The study does not provide any health-based arguments for buying bottled mineral and spring waters for those who are served with drinking water from public waterworks. Drinking water from crystalline bedrock aquifers should be analysed. In case of elevated concentrations of fluoride, uranium or arsenic, most bottled waters, but not all, will be better alternatives when treatment of the well water is not practicable.

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1. Introduction

The Geochemistry Group of the European geological surveys (EuroGeoSurveys) has arranged a common European sampling campaign of bottled mineral and spring waters, analysing a selection of 884 samples for more than 70 chemical parameters in one laboratory. The results show large-scale geographical patterns for many elements and will be presented as a European mineral water atlas (Reimann and Birke, 2010).

A subset of this survey, representing bottled groundwater from Norway, Sweden, Finland, and Iceland are presented together due to a small number of brands. The "Nordic countries" is normally a term comprising Denmark, the Faroe Islands, Finland, Iceland, Norway and Sweden. In this paper we have chosen to use the term without Denmark which hydrogeologically is very different from the other Nordic countries and without the Faroe Islands. Twenty-two samples

of bottled Nordic groundwater are presented here, of which 15 are sold as natural mineral waters and 7 are sold as spring water. The localities are shown in Fig. 1.

The tradition of mineral waters in the Nordic countries is limited to a very few brands, namely Ramlösa of Sweden, dating back to 1707 and bottled from 1912 and Farris of Norway, dating back to 1875 and bottled from 1907. These were enjoyed by a limited number of people and normally on relatively rare occasions. During the last 20 years, intense marketing efforts and more travelling to Southern Europe have changed the attitude of Nordic people towards buying increasingly more of their drinking water in bottles. The number of Nordic mineral water brands has increased much the very last years, but many are still not registered in the EU Commission's list of natural mineral waters recognised by member states and of the EEA; i.e. ten brands in Sweden, eight brands in Norway, one brand in Finland and Iceland, respectively (European Commission, 2009). In addition, a limited range of bottled spring waters exists, of which some rather obscure brands are still only available in a single hotel. The oldest mineral waters were originally marketed for their mineral content and their ability to cure. The newer brands tend to contain lesser minerals and are marketed as pure.

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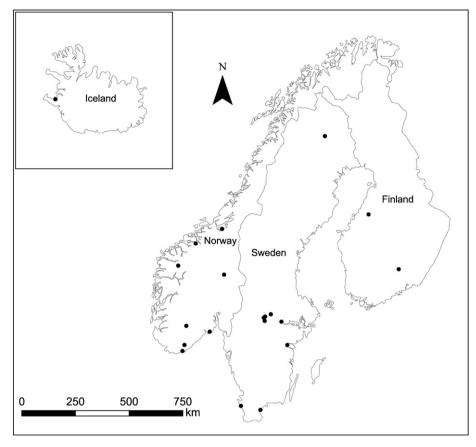


Fig. 1. Map of the Nordic countries showing where the mineral and spring waters are bottled.

In a large-scale regional geochemical mapping, coarse-meshed patterns can be revealed such as the influence of sea salts in the precipitation or which areas were glaciated during the latest Ice age (e.g. Reimann et al., 2003). This subset of European bottled groundwater with only twenty-two samples, is of course too small to be truly representative of Fennoscandian groundwater. Groundwater chemistry is encumbered with a range of factors which may disturb the overall picture, such as groundwater residence time, well depth, type of overburden and soil, CO₂ concentrations of infiltrating water, seasonal variations, marine influence or just accidental contamination. Nevertheless, is it possible to gain an impression of Nordic groundwater features based on less than two dozens of unevenly distributed mineral water samples?

2. Geologic and climatic setting

Norway, Sweden and Finland mainly comprise crystalline bedrock of Archaean to Paleozoic age. The Southernmost part of Sweden is made up of thick sequences of Paleozoic and Mesozoic sedimentary rocks, while Iceland is a geologically young part of the volcanic Mid-Atlantic ridge. Thick glaciers repeatedly covered the land areas during the Ice Age, leaving rather unweathered rocks overlain by unconsolidated Quaternary deposits. The mean annual precipitation ranges from less than 500 mm up to 3000 mm and is generally increasing from east to west. The evaporation rate is generally low, but during summer time potential evaporation may exceed precipitation in the southern part of the countries. The climate is temperate to subarctic. Iceland, the coast of Norway and the west coast of Sweden have a relatively mild and humid climate, while Finland and the inland areas experience a dryer continental climate with larger seasonal temperature differences.

Most of the nine Norwegian bottled groundwater samples are derived from unconsolidated Quaternary aquifers, of which glaciofluvial deposits are the most common (Table 1). Nor001 taps into an end moraine which has emerged from the sea during Holocene isostatic uplift. Nor006 and Nor007 are pumped from bedrock boreholes (<100 m deep) in Precambrian gneiss and Ordovician greenstone, respectively. All the water bottling plants are situated in forested remote areas, except Nor001 and Nor007 where the mineral waters are bottled in town centres.

The nine Swedish bottled water samples originate from different types of geological conditions. Swe001–004 are extracted from wells in rock aquifers, drilled down to between 55 and 200 m depth. Swe005–009 are extracted from coarse Quaternary sediments, of which all but 006 are esker deposits. Swe006 and Swe007 are situated in areas with several old mines and mineralisations in the underlying bedrock units, both iron- and sulphide ores. Generally speaking, the bedrock geology in Sweden ranges from Archaean to Proterozoic and Phanerozoic, however no bottled waters are produced in the Archaean area.

The Finnish samples comprise a mineral water (Fin001) pumped from a crystalline bedrock aquifer and a spring water from a shallow Quaternary aquifer (Fin002). The surrounding areas at both locations are dominated by forest.

The Icelandic samples are all from the same area and thus only two of the originally four bottled groundwater are presented here. The local geology at the site consists of interglacial rock units (olivine tholeiite) covered with layers of postglacial lavas. The site is a municipal conservation area with forest. The annual precipitation is approximately 1300–1400 mm/year, but most of the groundwater has its origin in the surrounding mountains where the annual precipitation increases to almost 3000 mm/year.

Table 1Geological and climatic settings for the 22 Nordic bottled groundwater samples.

Sample ID	Aquifer type	Bedrock lithology	Quaternary cover	Annual precipitation	Mean annual temperature	Land use
Nor001	Sediment	Permian monzonite	Submarine till	922	6	Urban
Nor002	Sediment	Late Precambrian dark grey sandstone	Glaciofluvial	645	2	Forested
Nor003	Sediment	Precambrian quartz-monzonitic gneiss	Thick talus and alluvial	936	4	Forested
Nor004	Sediment	Precambrian granitic and granodioritic gneiss	Glaciofluvial	-	-	Forested
Nor005	Sediment	Precambrian banded gneiss	Glaciofluvial/till	1595	5	Forested
Nor006	Rock	Ordovician greenstone	Marine sediments	868	4	Forested
Nor007	Rock	Precambrian banded gneiss	Glaciofluvial	1332	6	Urban
Nor 008	Sediment	Precambrian granitic and granodioritic gneiss	Glaciofluvial	-	-	Forested
Nor 009	Sediment	Precambrian quartz-dioritic gneiss	Alluvial	1290	6	Forested
Swe001	Rock	Jurassic sandstone	-	600	7	_
Swe002	Rock	Proterozoic, granites and metavolcanites	Esker	750	4	-
Swe003	Rock	Acid metavolcanics	Glacial/postglacial clay	650	5	-
Swe004	Rock	Quartz clauconite sandstone		600	7	-
Swe005	Sediment	Proterozoic metagreywacke	Esker	750	4	-
Swe006	Sediment	Proterozoic granite	Glaciofluvial	550	-1	Forested
Swe007	Sediment	Proterozoic acid metavolcanics	Esker	800	4	_
Swe008	Sediment	Proterozoic metagreywacke	Esker	700	3	_
Swe009	Sediment	Proterozoic metagreywacke	Esker	750	4	-
Fin001	Rock	Precambrian Gneiss	=	650	-	Forested
Fin002	Sediment	Precambrian granodiorite	Esker	550	_	Forested
Ice001 and 002	Rock	Quaternary olivin tholeiite	Holocene lavas	1400	-	Forested

3. Methods

The samples were bought from the shelves in ordinary super markets in Norway, Sweden, Finland and Iceland and were sent to the laboratory at Bundesanstalt für Geowissenschaften und Rohstoffe in Berlin where Dr. Manfred Birke and his team took care of all the analyses. One Finnish sample was kindly provided by the producer. The bottles were opened for the first time in the laboratory and pH and electrical conductivity were measured in a 100 ml beaker filled with mineral water. Thereafter the bottles were degassed before the bicarbonate content was determined by titration in 25 ml of the sample. The bottle content was further split into a 250 ml HDPE bottle and a 125 ml FLPE bottle both rinsed with the degassed mineral water and then filled. The 250 ml subsample was used for analysis of 8 anions by ion chromatography and ammonium ion by a photometric method. 1.5 ml ultrapure 69% nitric acid was added to the 125 ml subsample in a clean room and the bottle was shaken. The major cations and the trace elements were then determined by use of ICP-OES and ICP-OMS techniques. Ion balance errors were lower than 2% for all the samples which indicates a high quality of the analyses. Further details on the analytical procedures and quality assurance can be found in Reimann and Birke (2010).

To be able to compare bottled water with its most obvious alternative, samples of tap water were also collected. Eighteen 0.5 litre HDPE bottles were sent to friends and relatives spread over Norway and Sweden. They were asked to fill the bottle with domestic water from a tap in regular use, applying the following procedure:

- Let the water run for at least two minutes prior to sampling.
- Rinse the PET bottle five times with running tap water.
- Fill the bottle to the very top with the water and close it carefully, and avoid any contact with the inside of the bottle or the cap.
- Return the bottle by mail as fast as possible and try to keep it cool.

The bottles were then kept cool and sent as a whole to BGR in Berlin where they underwent the same procedures and analyses as the mineral water and spring water samples.

The data has been handled statistically and presented graphically by means of the data analysis programs DAS (Dutter et al., 1992) and AquaChem. Analytical values below the detection limit have been plotted as half the detection limit.

4. Results and discussions

Maximum, minimum and median values for all the analysed parameters are listed in Table 2. Please note that the units vary between $\mu g/l$ and m g/l. The table shows that the concentration of many elements varies over several orders of magnitude even within the same type of aquifer. The solubility of different chemical elements and compounds are in many cases dependent of the pH and the redox conditions of the water or are limited by saturation controls and sorption processes. Many reaction rates are governed by chemical kinetics and are in most cases slow compared to the residence time of water in the ground. As the present dataset in addition is derived from a broad range of settings, a typical distribution might be found, but a typical value is out of reach for most elements.

4.1. Other datasets for comparison

4.1.1. Tap water and surface water

In order to put the hydrochemistry of Nordic bottled waters into a larger picture, the results are presented and discussed together with other datasets of Nordic water. The eighteen samples of tap water were collected with an even geographical distribution in mind and they are assumed to represent the domestic water supply in Norway and Sweden fairly well. Large cities and towns often use treated surface water, smaller towns may use groundwater or artificially infiltrated water from Quaternary unconsolidated aquifers, while single houses and farms or groups thereof often use groundwater from bedrock boreholes or dug wells. Maximum, minimum and median values for all these tap waters are listed in Table 2. One would expect this dataset to also be representative of Finnish tap water due to rather similar geological conditions. It is however unknown whether Icelandic tap water is comparable to this dataset.

Many of the maximum values are derived from a bedrock borehole sample. In the subsequent graphical presentations, however, only the 12 samples taken from waterworks based on surface water are included in the dataset "Tap water" to better distinguish typical groundwater chemistry from surface water chemistry. The surface water samples are all derived from waterworks serving towns and cities from 30 000 to 60 0000 inhabitants and are likely to have undergone some water treatment, e.g. filtration, pH-adjustment and disinfection.

Table 2
An overview of minimum, median and maximum concentrations and spread (max/min) for all analysed parameters of the Nordic bottled water dataset (n = 22) and the Nordic tap water dataset (n = 18) respectively. The median concentration of European bottled waters (n = 884, including the Nordic bottled waters) (Reimann and Birke, 2010) is given for comparison.

	Nordic bottled water				Nordic tap water			European bottled water	
	Min	Median	Max	Spread	Min	Median	Max	Spread	Median
рН	3.9	5.6	7.9	2.0	6.8	8.025	8.36	1.2	6.8
EC [μS/cm]	23	197	2590	113	74	116.5	411	6	587
Ag [μg/l]	< 0.001	0.0019	4.37	8740	< 0.001	< 0.001	< 0.001	1	< 0.001
Al [μg/l]	0.401	5.83	85	212	0.544	13.4	53.3	98	1.16
As [μg/l]	0.0216	0.0616	1.72	80	0.0303	0.109	0.985	33	0.231
β [μg/l]	1.39	20.1	913	657	1.53	3.485	58.5	38	39
Ba [mg/l]	< 0.001	0.0085	0.039	78	1.36	7.585	82.2	60	0.028
Be [μg/l]	< 0.001	0.0027	0.0809	162	0.0010	0.0029	0.0188	18	0.0024
i [µg/l]	< 0.0005	< 0.0005	0.0048	19	< 0.0005	0.0004	0.0010	4	0.0007
a [mg/l]	0.93	6.36	66.2	71	7.13	20.35	56.6	8	65.9
d [μg/l]	< 0.001	0.0054	0.226	452	< 0.001	0.0071	0.0369	74	0.0032
e [μg/l]	< 0.0005	0.0077	6.16	24640	0.0007	0.0100	0.0573	78	0.0010
o [μg/l]	0.0084	0.0215	0.23 2.99	27	0.0037	0.0115	0.18 0.205	49	0.0232
r [µg/l]	0.0309	0.0992		97	0.0313	0.0653		7	0.121
s [µg/l]	0.0011 0.0803	0.0229 0.4185	0.218 12.8	198 159	<0.001 0.427	0.0074 4.79	0.303 110	606 258	0.0389 0.272
u [μg/l]	0.0003	0.4183	0.419	2085	0.427	0.0027	0.0141	36	0.0012
y [μg/l] : [μg/l]	< 0.0002	0.0045	0.23	4600	0.0004	0.0027	0.0141	66	0.0012
ι [μg/1] u [μg/l]	< 0.0001	0.0043	0.0623	1246	0.0002	0.0022	0.0078	14	0.0023
u [μg/1] e [μg/l]	<0.1	1.42	51.1	1022	0.541	2.865	196	362	0.678
- [μg/1] a [μg/l]	< 0.0005	0.0070	0.287	1148	0.0298	0.1525	1.51	51	0.0023
a [μg/1] d [μg/l]	< 0.0003	0.0107	0.677	6770	0.0009	0.0029	0.0204	22	0.0025
α [μg/I] e [μg/l]	< 0.005	0.0298	2.06	824	< 0.005	0.0023	0.149	60	0.0263
f [μg/l]	< 0.0005	0.0007	0.0036	14	0.0006	0.0034	0.0081	13	0.0005
ο [μg/l]	< 0.0003	0.0018	0.0871	1742	0.0002	0.0008	0.0032	16	0.0005
μg/l]	0.364	3.335	431	1184	0.611	1.3	19	31	4.75
[mg/l]	0.2	1	74.3	372	0.05	0.85	8	160	2.1
[µg/l]	< 0.0005	0.0414	10	40 000	0.0014	0.0126	0.207	150	0.0023
[µg/l]	<0.1	0.6435	12.8	256	<0.1	0.5365	8.43	169	9.94
[μg/l]	< 0.00005	0.0010	0.0227	908	< 0.00005	0.0006	0.0067	268	0.0002
g [mg/l]	0.307	1.018	30.7	100	0.473	1.6	15.4	33	16.4
n [mg/l]	< 0.001	0.004	0.075	150	0.05	1.17	27.3	546	0.002
lo [μg/l]	0.03	0.173	4.81	160	0.00276	0.17	3.66	1326	0.282
a [mg/l]	1.4	11.55	704	503	1	4.35	49.4	49	15.2
b [μg/l]	< 0.001	< 0.001	0.0609	122	0.00104	0.00554	0.0209	20	0.0012
d [µg/l]	< 0.0002	0.0443	5.12	51 200	0.00167	0.0107	0.141	84	0.0021
i [μg/l]	< 0.01	0.142	1.03	206	0.0452	0.369	1.59	35	0.176
[μg/l]	0.0042	0.01795	0.503	119	0.00291	0.0301	0.151	52	0.0158
[µg/l]	< 0.00005	0.01	1.54	61600	0.0004	0.0027	0.035	80	0.0004
o [μg/l]	0.09	1.51	4.65	52	0.0533	0.6745	3.23	61	2.08
ρ [μg/l]	0.0078	0.4155	0.739	94	0.0075	0.027	0.193	26	0.269
[µg/l]	0.0117	0.0455	0.078	7	0.005	0.0272	0.0736	15	0.0697
e [μg/l]	< 0.01	0.0359	2.28	456	0.0139	0.0383	0.358	26	0.0543
n [µg/l]	0.0005	0.0095	0.697	1512	0.0003	0.0025	0.022	77	0.0013
ı [µg/l]	0.0016	0.0048	0.301	189	0.0070	0.0117	0.112	16	0.0065
[mg/l]	0.002	0.04	0.335	168	0.012	0.043	0.355	30	0.32
ι [μg/l]	< 0.001	< 0.001	0.0022	4	0.0013	0.0049	0.0191	14	< 0.001
[μg/l]	< 0.00005	0.0017	0.0825	3300	0.0000	0.0004	0.0021	82	0.0002
[µg/l]	< 0.005	0.0058	0.0118	5	< 0.005	0.0079	0.02	8	0.0068
ı [µg/l]	0.0002	0.0010	0.0195	80	0.0003	0.0016	0.0103	39	0.0004
[µg/l]	< 0.01	< 0.01	0.0786	16	< 0.01	0.0575	0.295	59	0.0344
[µg/l]	< 0.0005	0.0036	0.0208	83	0.0015	0.0045	0.0179	12	0.0041
n [μg/l]	0.00006	0.0008	0.0273	433	0.0001	0.0005	0.0019	26	0.0002
[µg/l]	0.0055	0.1020	32.4	5891	0.0049	0.1071	56.2	11400	0.228
[µg/l]	0.0732	0.1875	13.7	187	0.0574	0.0879	0.634	11	0.168
[µg/l]	0.0021	0.0223	4.52	2173	0.0045	0.0117	0.0741	17	0.0222
[µg/l]	<0.0005 0.0002	0.0703	3.55	14200	0.0032	0.0236	0.0923 0.028	29 56	0.0122
) [μg/l]) [μg/l]	0.0002	0.0047 0.675	0.148 19.1	685 277	0.0005 0.682	0.0034 3.795	130	56 191	0.0010 0.89
n [μg/l] [μg/l]	< 0.009	0.0048	0.198	396	0.082	0.0083	0.249	167	0.89
- [μg/1] [mg/l]	< 0.001	0.0205	2.58	1720	< 0.0015	0.006	0.249	107	0.0072
CO ₃ [mg/l]	<0.003 4	79.3	1930	483	35.2	58.95	299	8	284
_ [mg/l]	0.39	79.3 11.3	699	483 1792	1.38	8.575	49.1	8 36	13.3
[IIIg/I] - [mg/l]	0.39	0.168	2.59	1792	0.0015	0.0335	1.35	900	0.186
	< 0.018	< 0.005	2.59	820	< 0.0015	< 0.005	0.023	900	<0.005
O^{2-} [mg/l]	< 0.005	< 0.005 < 0.005	2.05 0.152	820 61	< 0.005	< 0.005 < 0.005	0.023	10	<0.005 <0.005
0^{3} [mg/l]	0.005	< 0.005 0.22	0.152 4.43	886	0.005	< 0.005 0.92	4.32	432	<0.005 1.32
$O_4^{111g/1}$	< 0.005	0.22	1.54	154	< 0.02	< 0.02	0.02	432	0.1
O_4^2 [mg/l] O_4^{2-} [mg/l]	<0.02 0.93	0.1 5.355	1.54 32.8	35	<0.02 1.12	<0.02 7.645	40.7	36	0.1 19.9
	0.55	3,333	32.0	22	0.28	7.645 1.45	6.97	25	6.45

Hg concentrations are not given as all samples returned values below the detection limit of < 0.5 ng/l.

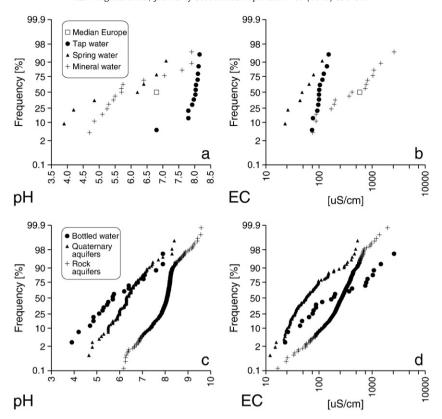


Fig. 2. cdf-diagrams showing a) pH of the subsets Spring water and Mineral water compared to 12 treated tap water samples from Norwegian and Swedish lakes and the median pH value of 884 European bottled waters (Reimann and Birke, 2010); b) electrical conductivity (EC) of the subsets Spring water and Mineral water compared to 12 treated tap water samples from Norwegian and Swedish lakes and the median EC value of 884 European bottled waters (Reimann and Birke, 2010). Note the log scale; c) pH of 22 Nordic bottled waters compared to pH of 89 groundwater samples from Fennoscandian Quaternary unconsolidated aquifers (Cramer et al., 2010) and pH of 476 Norwegian crystalline bedrock aquifers (Frengstad et al., 2000); and d) EC of 22 Nordic bottled waters compared to pH of 89 groundwater samples from Fennoscandian Quaternary unconsolidated aquifers (Cramer et al., 2010) and pH of 476 Norwegian crystalline bedrock aquifers (Frengstad et al., 2000). Note the log scale.

4.1.2. Groundwater from Quaternary aquifers

Another dataset used for comparison is called "Quaternary aquifers" (N=89). It includes 30 Norwegian, 29 Swedish and 30 Finnish groundwater monitoring reference stations comprising springs and relatively shallow wells in unconsolidated Quaternary deposits (Cramer et al., 2010). The monitoring areas are intended to represent pristine aquifers in various geological and climatic settings typical for the three countries. The samples were collected in a collaborative project by the three Fennoscandian geological surveys and the Finnish Environment Institute (SYKE). Comparable equipment and procedures were used in the three countries, and the water samples were analysed at the laboratory of the Geological Survey of Finland (GTK).

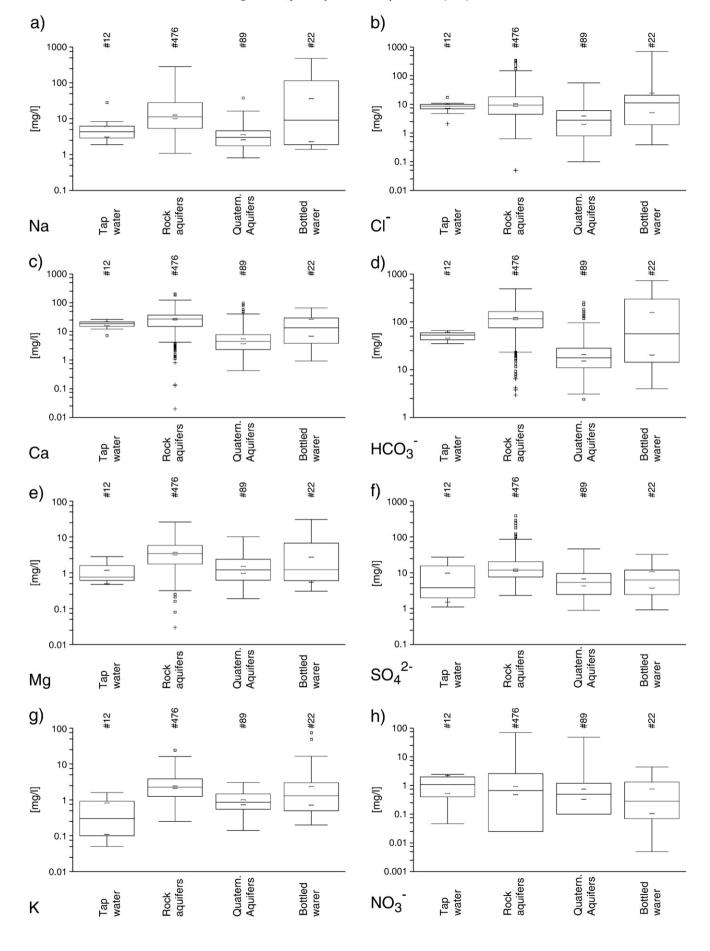
4.1.3. Groundwater from crystalline bedrock aquifers

The third dataset "Rock aquifers" comprise water samples from 476 Norwegian crystalline bedrock boreholes (Frengstad et al., 2000). This dataset is a subset of almost 2000 samples collected from private wells and smaller waterworks which were analysed for major and minor elements at the laboratory of the Norwegian geological survey (NGU) (Banks et al., 1998a,b). The subset was further analysed for trace elements by ICP-MS at BGR.

4.2. pH and electrical conductivity

Fig. 2a shows that the pH of bottled waters which are sold as mineral water is generally one pH-unit higher than those sold as spring waters, although both subsets display large variations and are overlapping. The differences in pH values between the two subsets are probably even larger prior to abstraction of the waters since mineral waters are more likely to be carbonated than spring waters. CO₂ gas is added to nine of the bottled waters, of which eight are marketed as mineral water. Carbonation of water may lower the pH by 2-3 U by the reaction $H_2O + CO_2 = H^+ + HCO_3^-$. This implies that no significant correlations between pH and any other parameter in the dataset can be seen. The median pH of the Nordic bottled waters is 5.6 and considerably lower than the median pH of 6.8 for the European bottled waters (Table 2). One possible explanation can be that European bottled waters have around ten times higher median Ca concentrations than the Nordic bottled waters (Table 2). High Ca content tends to raise pH towards 8.3 in un-carbonated waters and will also act as a buffer against lowering of pH in carbonated waters (Appelo and Postma, 1994).

Fig. 2b displays that the electrical conductivity (EC) of the so called mineral waters generally are ten times higher than for the ones labelled as spring waters, reflecting that the latter group is much less hydrochemically mature. Simple correlation analysis by use of the program code DAS (Dutter et al., 1992) revealed that EC is almost perfectly correlated with the Na concentrations (r=0.97), while concentrations of Cl $^-$ (r=0.85), Mg (r=0.82), HCO $_3^-$ (r=0.71), K (r=0.37), Ca (r=0.35), and SO $_4^2$ (r=0.31) in decreasing degree follow the EC values (figures not shown). It should be noted that the number of samples is low and that the correlation factors are only given as indications of the relative importance of the different major



ions for the total ionic strength. Compared to the median value for electrical conductivity of the entire dataset of European bottled waters, the subset of Nordic mineral waters alone has a similar median value. Fig. 2a and b also displays that pH of tap waters is well regulated around pH = 8 and that the variation of electrical conductivity of tap waters (i.e. surface waters) is small. More surprising is that the electrical conductivity of spring waters is lower than that of tap waters. This might be due to addition of ions in waterworks during pH regulation processes. Another explanation is that many of the waterworks are situated along the coast where seaspray adds marine salts to the precipitation, while spring waters are often bottled at more inland locations.

In Fig. 2c and d the Nordic mineral waters and spring waters are merged together as bottled waters and pH and EC are presented in relation to groundwater samples of "Quaternary aquifers" and "Rock aquifers". pH of bottled waters is generally lower than pH of groundwater from both unconsolidated and crystalline aquifers. This is again an effect of carbonation of the bottled waters. The variation of electrical conductivity among bottled waters fills almost the same span as EC for Rock aguifers and Quaternary aguifers together. In that respect, the dataset of Nordic bottled waters seems to be representative of Nordic groundwater. However, those bottled waters which are plotting in the upper 50percentile have up to five times higher EC values than the corresponding samples in the other datasets. Water with such a high content of solute salts is unlikely to be consumed on a daily basis. As a consequence, the "Rock aquifers" dataset which are based on drinking water wells do not comprise water samples with so high ionic strengths.

4.3. Major elements

The major elements are displayed as boxplots in Fig. 3a-h, comparing the basic statistics of "Tap water", "Rock aquifers" "Quaternary aquifers" and "Bottled waters". The following observations can be made:

- "Bottled waters" resembles "Rock aquifers" regarding Na, Cl⁻, and Ca and resembles "Quaternary aquifers" regarding Mg and SO₄.
- The variation of HCO₃⁻ concentrations within "Bottled waters" is almost as large as the variation within "Rock aquifers" and "Quaternary aquifers" together.
- "Bottled waters" seems to have the lowest median value of NO₃, although the differences are not significant.
- "Tap water" has a rather uniform content of Na, Cl, Ca, and HCO₃ compared to the other datasets.
- The median values of all major elements, except NO₃ are significantly higher in "Rock aquifers" than in "Quaternary aquifers".
- The median values of Ca, Mg and SO_4^{2-} are significantly higher in "Rock aquifers" than in the other datasets.

4.3.1. Water types

The Durov (1948) diagram in Fig. 4 displays the content of Na, Ca, Mg, Cl⁻, HCO₃⁻ and SO₄²⁻ in Nordic bottled waters according to nationality. Although many of the water samples plot somewhat in the middle of the central square in the diagram, some samples are found close to three of the corners. One sample plots in the lower right corner of the square. This means that it is of Na–Cl type, i.e. the major hydrochemistry being dominated by marine salts. This Norwegian mineral water is pumped from a moraine which has emerged from the sea in late Holocene time due to isostatic uplift. Three of the bottled waters plot in the upper right corner of the square and are of Ca–HCO₃ type, pointing towards calcite dissolution as the main geochemical process. In the upper right corner of the square Na–HCO₃ type waters plot, of which there are four in this dataset. This water type indicates a more mature hydrochemistry

and might be the result of silicate weathering and subsequent calcite precipitation (Banks and Frengstad, 2006). One of these Na–HCO₃ type waters is pumped from 132 m depth in a layer of Jurassic sandstone intercalated with rather impermeable clay- and silt-stones. The properties of the water has been attributed to ion exchange processes of Ca for Na taking place as the water passes through layers containing kaolin.

The bulk of the Nordic bottled waters basically plots as a combination of Ca-Na-HCO₃-Cl which are typical of rather immature groundwater in coarse-grained glaciofluvial and fluvial aquifers. Sea salts in the precipitation are almost as important for the ion composition as weathering of minute amounts of calcite in the ground. SO_4^{2-} counts for more than 20% of the major anions in only four samples, and not more than one sample has SO_4^{2-} as the dominating anion. It should be noted that even if several of the Swedish wells (i.e. SWE003, 005, 006, 007, and 009, see Table 1) are situated in mining areas like the Bergslagen and the Norrbotten districts, this cannot be seen in elevated content of SO_4^{2-} or metals. Neither are there noticeable SO_4^{2-} contents in the samples from Iceland where one could expect sulphur input from volcanic activity. In the Icelandic case, the area has probably been chosen for drinking water production because of the low S content. Only three samples display a magnesium content above 20% of the cations and none of the sampled Nordic bottle waters is dominated by Mg. This corresponds well with the findings of Banks et al. (1998b) for unconsolidated aquifers while Banks et al. (1998a) found that less than 1% of 1604 Norwegian bedrock boreholes yielded water of Mg-HCO₃ composition.

Potassium and NO_3^- are not included in the Durov diagram. Fig. 3h indicates that the content of NO_3^- is pleasantly low in Nordic bottled waters and would not have been dominating in company with the other major anions. For one sample (NOR003), which has a low ion content, NO_3^- makes 19% of the anions. For the other samples, the NO_3^- share is <5% and normally <0.3%. Fig. 3g shows two samples where the K content is remarkably high, almost 50 and 75 mg/l, respectively. In both cases, the elevated K values are accompanied with a substantial content of Na, Ca, and Mg, and their share of the cation composition in their respective Swedish mineral waters is still lower than 20%. In the other samples the K content make <10% and normally <2%. The origin of K might thus be enhanced weathering of silicates, including K-feldspar, probably due to long residence time and high initial CO_2 content of the water when it infiltrated the aquifer (Banks and Frengstad, 2006).

4.4. Selected trace elements

For the sake of brevity, only twelve of the analysed trace elements are presented here. They are selected because of their health implications (As, Cd, F, I, Ni, Pb, Sb, and U), their use as indicators of water-rock interaction (B, Br, and Li), their use as indicators of contamination from plumbing and bottle materials (Ni, Pb, and Sb) and their expected high levels in Nordic waters (Er, F, and U). Fig. 5a-1 shows cdf-diagrams of selected trace elements in the datasets "Tap water", "Rock aquifers" (Frengstad et al., 2000), "Quaternary aquifers" (Cramer et al., 2010) and "Bottled waters". The median value of European bottled mineral and spring waters (Reimann and Birke, 2010) is also plotted as a single point for reference. For all elements shown, except iodine, the concentrations among "Rock aquifers" are elevated compared to "Quaternary aquifers". This is in accordance with the electrical conductivity (Fig. 2d) and the major elements (Fig. 3) for the two datasets. The strongest mineral waters are thus most likely to be abstracted from rock boreholes, and if someone wants to bring mineral water to the market with pH above 9.5 these waters can be found among private bedrock boreholes in Norway (Frengstad et al., 2001) (Fig. 2c). The "Quaternary aquifers" dataset was unfortunately not analysed for Er

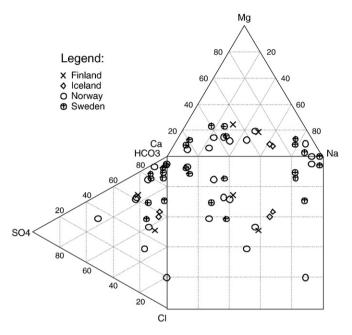


Fig. 4. Durov (1948) diagram displaying the distribution of main water types according to national subsets.

or other rare earth elements. Also the detection limits in this dataset are higher for some of the elements, and the lower part of the distribution therefore might plot as vertical sequences, representing half the detection limit.

4.4.1. Arsenic

Arsenic is carcinogenic and chronic poisoning induces keratotic lesions of palms and soles. Maximum admissible concentration (MAC) for drinking water in the EU is set to 10 µg/l (EU, 1998). The distribution of As concentrations for bottled waters and tap water are rather similar and plots between "Quaternary aquifers" and "Bedrock aguifers" (Fig. 5a). Bedrock aguifers displays a median value comparable to the median value of European bottled waters, around 4 times higher than for Nordic bottled waters (Table 2). Excessive As concentrations are known to be a problem in groundwater many places in the world (Nordstrom, 2002) with Bangladesh and West Bengal as the grimmest examples when it comes to the number of people affected. Elevated As concentrations have been found in water from bedrock aguifers in the southern Pirkanmaa region of Finland (Backman et al., 2008) and the Västerbotten region of Sweden (Jacks et al., 2009). The dataset "Bedrock aquifers" which is used for comparison shows minor breaches of the EU drinking water limit of 10 μg/l in 1% of the Norwegian bedrock boreholes (Frengstad et al., 2000). The present dataset of bottled waters was not able to point out As as a potential problem in some areas of Fennoscandia. There were no samples from potential risk areas in central Finland and northern Sweden.

4.4.2. Boron

Boron is assessed to be non-toxic for humans and EU operates a MAC in drinking water of 1.0 mg/l (EU, 1998) while a MAC for boron in bottled waters is still pending in Norway (Norwegian Ministry of Health and Care Services, 2004). The highest B concentration among Nordic bottled waters is 913 μ g/l in SWE001. B concentrations in bottled waters follow "Bedrock aquifers" except for two extreme values (Fig. 5b), while B concentrations in "Tap water" follows "Quaternary aquifers". In the present survey the best correlation involving B is that with F (r=0.78), while there is also good correlation with Na (r=0.59) and EC (r=0.55). The correlation factors are given only as illustrations, as the data are compositional

(i.e. concentrations of the separate elements are given as relative parts of the total water sample and are thus dependent on each other). Traditional correlation analysis of geochemical data may thus be ambiguous (Filzmoser et al., 2010). B has no clear solubility limits and may indicate the extent of water–rock interaction the sample has undergone. The median value of B in European bottled waters is two times the median value in Nordic bottled waters (Table 2), pointing towards generally more mature mineral waters in continental Europe than in Fennoscandia.

4.4.3. Bromide

The bromide distribution of "Bottled waters" covers more than three orders of magnitude as for "Bedrock aquifers" and their median values are close to the median value for European bottled waters. The Br/Cl ratio is rather homogeneous, ranging from $0.2*10^{-3}$ to $6.7*10^{-3}$ and many bottled waters show Br/Cl ratio close to a modern sea water ratio of $3.47*10^{-3}$ (Edmunds, 1996). This is as expected, since Fennoscandian rocks do not comprise any evaporites. "Tap waters" has a low and rather homogeneous Br distribution (Fig. 5c) which probably is due to the similar homogeneity of Cl (Fig. 3b) and minimal input from mineralogical sources.

4.4.4. Cadmium

Cadmium accumulates in the body and is known to induce bone and renal disease (Crounse et al., 1983). MAC in the EU is set to 5 µg/l. The distributions of Cadmium concentrations are overlapping between "Bottled waters", "Quaternary aquifers" and "Tap water" and their median values are slightly higher than for European bottled waters (Fig. 5d). Cadmium is associated with sulphides and is well correlated with Zn (r = 0.94) As (r = 0.89), and Se (r = 0.98) and to some degree with SO_4^{2-} (r = 0.56) in the "Bottled waters" subset. One water sample with higher Cd concentrations (0.23 µg/l, NOR006) is taken from a borehole drilled into greenstone which is known to be sulphide-bearing (Grenne and Roberts, 1980). The same water sample also shows a high value of Se (2.28 µg/l) which also probably has its origin in oxidation of pyrite or other sulphides. The Swedish samples from mining areas do not show any particularly high Cd concentrations.

4.4.5. Erbium, other lanthanoids and yttrium

The distribution of erbium in Fig. 5e represents the rare earth elements which broadly speaking behave more or less in the same way in the environment. "Bottled waters" has almost as high concentrations of Er as "Bedrock aquifers" with a median value around five times higher than the median value of European bottled waters (Table 2). The distribution of Er concentrations spans four orders of magnitude.

Five Norwegian and five Swedish bottled waters display elevated concentrations of some or most of the lanthanoids. The lanthanoids, or so called rare earth elements, often occur together and seem to be up-concentrated in late phases of magmatic crystallisation and thus enter the lattice of many rock-forming minerals in acid magmatic rocks (Reimann et al., 2003). Ottesen et al., 2000 show high concentrations of Er in the acid-soluble part of overbank sediments in south Norway, while Reimann et al., 2003 show a belt of higher La concentrations in agricultural soils through S-Norway, Mid-Sweden and S-Finland. Lanthanoid concentrations are elevated also in stream waters in Finland, Norway and Sweden in the geochemical baseline mapping of Europe (Salminen et al., 2005). The bottled water with the decidedly highest concentrations of lanthanoids (e.g. Er: 0.23 μg/l, La: 10 μg/l) (NOR005) is abstracted from an area well-known for its pegmatite dykes and rich variety of minerals. There has even been mining of Er in this area, Rare earth elements seem to be widespread in the Fennoscandian environment although the spread between the minimum and maximum values are enormous (Table 2). Fig. 5e

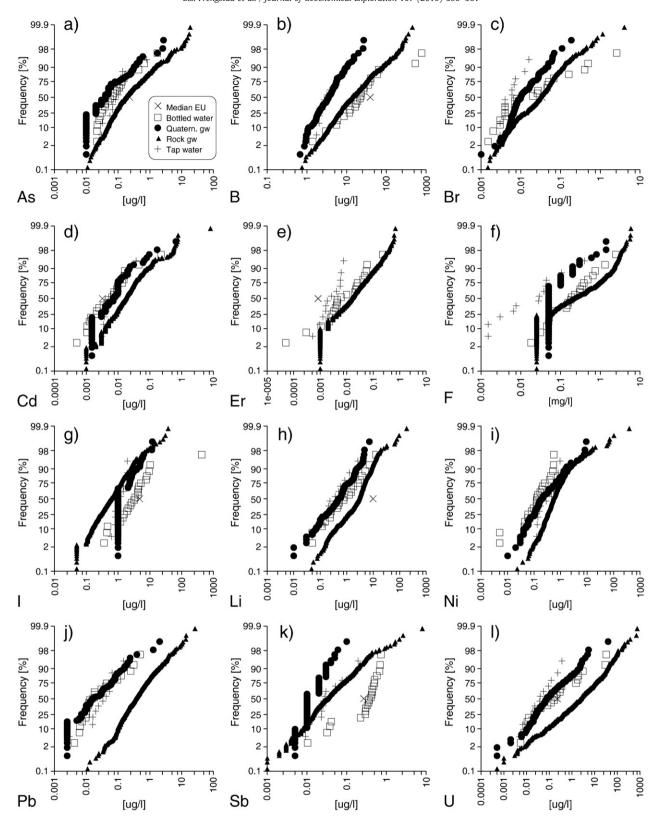


Fig. 5. cdf-diagrams of some selected trace elements comparing Nordic bottled water with tap water from lakes, groundwater from Quaternary aquifers, and groundwater from crystalline bedrock aquifers, see 4.1.1–3 for explanation: a) arsenic, b) boron, c) bromide, d) cadmium, e) erbium, f) fluoride, g) iodide, h) lithium, i) nickel, j) lead, k) antimony, and l) uranium. The median values of 891 European bottled waters (Reimann and Birke, 2010) are also inserted for each element. Note the log scale.

shows that even surface waters in Norway and Sweden have a higher median value of erbium than have the European bottled mineral and spring waters. Also elevated concentrations of Y are found in some of the Norwegian samples with elevated lanthanoid content as could be expected because of their related geochemical behaviour. Reimann et al., 2003 also show higher levels of Y in agricultural soils in Norway, Sweden and part of Finland compared to other countries around the Baltic Sea.

4.4.6. Fluoride

Fluoride concentrations in "Bottled waters" are comparable to the levels in "Rock aguifers" and are close to a log normal distribution (Fig. 5f). Fluoride is an essential element for teeth health and is thus added to the drinking water in some countries to avoid caries. In concentrations above 1.5 mg/l it may however cause damage to teeth under formation (dental fluorosis). Bedrock boreholes in Fennoscandia tend to yield water with elevated fluoride content: 25% of 9225 bedrock wells in Sweden had F concentrations >1.6 mg/l (Aastrup et al., 1995), 16% of 1604 bedrock wells in Norway had F concentrations > 1.5 mg/l (Banks et al., 1998b), while the median F concentration of 113 bedrock wells in Finland was 0.63 mg/l (Lahermo and Backman, 2000). Elevated F⁻ concentrations are more likely to occur in Na-HCO3 type groundwater (Banks et al., 1998b). This might be because a fluorite (CaF₂) saturation ceiling keep the F⁻ concentration low if Ca²⁺ concentrations are high (Handa, 1975). Both SWE001 and SWE003 with F⁻ concentrations of 2.6 mg/l and 1.3 mg/l respectively are Na-HCO₃ type water from bedrock boreholes. European bottled waters display a similar median value of 0.18 mg/l. "Tap water" and "Quaternary aquifers" have low to moderate levels.

4.4.7. Iodine

Iodine is an essential element. A daily intake of 0.15-0.25 mg/l is recommended to avoid development of goitre (WHO/ICCIDD/UNICEF, 2007). The distributions of Iodine above the different detection limits are rather similar in the datasets of "Rock aquifers", "Quaternary aquifers" and "Tap water" (Fig. 5g). Bottled groundwater displays a median value which is around three times higher and is approaching the median value for I in European waters (Table 2). It is difficult to imagine that bottled groundwater should have a I distribution at a totally different level than other Nordic groundwater and the cdfdiagram probably highlights the potential problem of comparing datasets analysed in different laboratories. The single high value of I (431 μ g/l) is found in NOR001. The water has a rather brackish character and the iodine is assumed to have a marine origin. Excessive PO₄³⁻ content was also found in the same spring and is probably derived from marine fossils deposited in the surrounding sediments. As the spring is situated in an urban area, an origin in fertilizers or sewage cannot be entirely ruled out. A comparable PO_4^{3-} content is however found in a Swedish mineral water from a bedrock borehole covered with 7 m of Holocene marine clay.

4.4.8. Lithium

Lithium is regarded as an essential element and is used in the treatment of manic-depressive disorders (Crounse et al., 1983). The distribution of Li in bottled groundwater fall between "Rock aquifers" and "Quaternary aquifers", while "Tap waters" have only slightly lower Li values (Fig. 5h). European bottled waters have a median value of Li which is around 15 times higher than the value for Nordic bottled waters. Typical host minerals for lithium are micas and amphiboles, and granites and schists are thus enriched in Li. Since the element has no clear solubility ceiling, it can possible be used as an indicator of water–rock interaction. In the present dataset Li has a modest correlation with electrical conductivity (r=0.50, figure not shown), but sodium which also has few solubility restrictions, seems to be a better indicator of hydrochemical maturity when samples with obvious marine influence are omitted (see Section 4.2).

4.4.9. Nickel, lead and gallium

Nickel (Fig. 5i) is an essential element, but high concentrations may cause damage to heart and liver and irritation of the skin (USEPA, United States Environmental Protection Agency, 2009). In the EU, the maximum admissible concentration in drinking water is set to $20\,\mu\text{g/l}$ (EU, 1998). Lead (Fig. 5j) accumulates in the body and may cause damage to the nervous system, blood and kidneys,

particularly in small children. MAC in the EU is set to 10 µg/l (EU, 1998). The highest concentrations in the "Bottled waters" subset were 1.03 µg/l for Ni and 0.50 µg/l for Pb. Nickel and lead are particularly soluble in soft, acid water and might in addition to mineralogical sources be derived from plumbing materials. Both the "Tap water" and the "Rock aquifer" datasets are sampled from taps and may thus show higher values than the natural level of these metals. On the other hand, the data reflects the water quality as it is drunk. Without any contamination, one could expect that groundwater samples displayed higher levels of dissolved metals than surface water samples as the contact with minerals is closer and the residence time is likely to be longer. pH of the tap water samples is also higher than for the bottled waters (Fig. 2), implying that metals are less soluble in the tap waters. The ratios between median values of "Tap water" and "Bottle water" for different metals, may thus give an indication of the minimum influence of plumbing materials: Copper $10\times$, Nickel $2.6\times$, Lead $2\times$, Titanium $10\times$ and Zn $5.6\times$. Interestingly, the median gallium value of tap waters is found to be 10 times higher than of bottled waters. This was at first thought to reflect that Ga often is associated with Zn in nature and thus is also following Zn in plumbing material. However, Ga and Zn show no significant correlation in the present dataset.

4.4.10. Antimony

In all cases but one, the mineral water bottles intended for the Nordic market are made of clear or bluish PET material with PE caps. This container material is known to occasionally leach detectable amounts of Sb due to use of antimony trioxide as a catalyst during the manufacturing process of PET (Shotyk et al., 2006). Fig. 5k shows elevated and rather uniform Sb concentrations for most of the bottled waters indicating contamination. Sb is toxic and suspected to be carcinogenic in the long term (USEPA, United States Environmental Protection Agency, 2009). The European Union (EU, 1998) has thus set a MAC of 5 μ g/l. All the Sb analyses of Nordic bottled waters are below 1 μ g/l, but the level in plastic bottles is 10 to 100 times higher than in glass bottles.

Glass bottles are not necessarily a better choice. Misund et al. (1999) found significantly higher levels of Pb, Li, U, and Zr, as well as Na and K, in mineral waters sold in glass bottles.

4.4.11. Uranium, radon and thorium

Fig. 51 shows a rather large spread of U concentrations for all datasets, with "Rock aquifers" covering almost six orders of magnitude. The distribution of the "Bottled waters" dataset seems to reflect the difference between groundwater from bedrock and from unconsolidated aquifers. Several previous surveys have shown that U in elevated concentrations is fairly common in Fennoscandian groundwater, mainly from crystalline bedrock aquifers (e.g. Reimann et al., 1996; Frengstad et al., 2000; Vesterbacka et al., 2005; Ek et al., 2007). Even concentrations of more than 14 mg/l U has been measured in a drinking water borehole in granitic bedrock outside Helsinki (Asikainen and Kahlos, 1979). EU has not set any MAC for U in drinking water, while WHO (2006) suggests a guideline value of 15 µg/l. One of the Finnish bottled water samples shows an elevated U concentration of 29 µg/l, which is typical for bedrock well water in Svecofennian orogenic crust surrounding younger rapakivi type granites in South-Finland (Karro and Lahermo, 1999). The analyses also return a U value of $32.4\,\mu\text{g/l}$ for a Swedish bottle water.

Among the tap water samples collected for this study (of which only the surface waters has been displayed in the figures) the single water sample from a private Swedish bedrock well interestingly enough contains an elevated uranium concentration ($56.2 \,\mu g/l$). Even a tap water sample from a waterworks based on artificial infiltration in an esker returns an elevated U value ($27.3 \,\mu g/l$). Approximately 20% of all drilled wells in Sweden exceed the current Swedish guideline value of $15 \,\mu g/l$ (Ek et al., 2007). In Norway, 18% of 476 bedrock

boreholes yielded water above the previous US maximum admissible concentration of 20 μ g/l (Frengstad et al., 2000). No limits are yet set for uranium in EU drinking water regulations.

Radon is a product of the uranium decay series and the gas is a widespread problem in Fennoscandian groundwater, especially in granitic rocks (Banks et al., 1995; Asikainen, 1982; Åkerblom, 1994). This is however not likely to be a problem in bottled waters due to radon's short half life of 3.8 days. The concentrations of Rn will be minimised before the bottles reach the shop shelves and not constitute a problem in the final product. SWE006 contains measurable concentrations of Rn at the source, which is removed prior to bottling.

Thorium has, in opposition to uranium, a very low solubility in natural waters, but somewhat elevated concentrations $(0.01-0.02 \,\mu\text{g/l})$ are unexpectedly found in two of the most diluted bottled waters from Norway (NOR003 and NOR008). Both of these are abstracted from unconsolidated aquifers derived from light gneissic rocks.

5. Conclusions

The studied small dataset of Nordic bottled groundwater samples is able to pick up which elements generally tend to cause health-related drinking water quality problems in Nordic groundwater, namely uranium and fluoride in bedrock boreholes. Nitrate and arsenic usually display moderate levels, but some known regions and/or settings with high levels of As were not identified in this dataset.

The very coarse-scale geological differences between the Nordic countries and continental Europe shine through in the major ion hydrochemistry. Median values of calcium and magnesium are 10 and 16 times higher in European bottled waters, respectively, showing that carbonates are much more widespread in continental Europe.

Bottled waters labelled as mineral waters generally have a content of solved minerals which is an order of magnitude higher than in those sold as spring water. It is more common to carbonate mineral waters than spring waters and the comparison of pH between different bottled waters has limited relevance.

The majority of Nordic bottled waters are water types dominated by a combination of Ca–Na–HCO₃–Cl which are typical of shallow groundwater in coarse-grained unconsolidated aquifers. More mature mineral waters of Na–HCO₃ type are found in rock aquifers. A Na–Cl water type of marine influence is also represented.

Both groundwater and surface water in Fennoscandia seem to be enriched in rare earth elements compared to continental Europe.

Bottled waters have significantly lower levels of metals associated with plumbing than tap waters, though both water types are well within given drinking water limits for Cu, Ni, Pb and Zn.

Water sold in plastic bottles is contaminated with antimony, an element suspected to be carcinogenic, but the levels are well below the drinking water limit.

For people connected to public waterworks in the Nordic countries this paper reveals no obvious health-related reasons to choose mineral water as the main source for drinking water. There is however an indirect positive health effect as far as mineral water displaces sugar-containing soft beverages and alcohol drinks. For those who have a private water supply from bedrock boreholes with elevated contents of fluoride, uranium, and/or arsenic some bottled groundwater, but not all, may offer a better drinking water quality if well water treatment is not practicable.

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