



## Uranium and lanthanum in Norwegian drinking water – Is there cause for concern?



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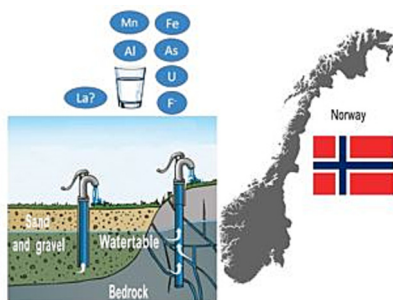
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### HIGHLIGHTS

- Exceedances of drinking water regulations' parametric values were noticed for Mn, Fe, As, Al, U, and F<sup>-</sup>.
- Sedimentary well water exceeded the Australian health-based guideline value for La.
- Bedrock well water showed significant positive correlation between precipitation and U concentration.

### GRAPHICAL ABSTRACT



Do the Drinking Water Regulation's limit values provide a sufficient basis for ensuring safe drinking water for the Norwegian population?

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### ABSTRACT

Due to natural conditions such as geology, topography, and climate, and historical features such as resource utilization, land use, and settlement patterns, the drinking water supply in Norway is separated into many public and private water supply systems. This survey sheds light on whether the Drinking Water Regulation's limit values provide a sufficient basis for ensuring safe drinking water for the Norwegian population. Participating waterworks, both private and public, were spread throughout the country, in 21 municipalities with different geological conditions. The median value for the number of persons supplied by the participating waterworks was 155. The two largest waterworks, both of which supply >10,000 people, have water sources from unconsolidated surficial sediments of latest Quaternary age. Fourteen waterworks have water sources from bedrock aquifers. Raw and treated water were analysed for 64 elements and selected anions. The concentration of manganese, iron, arsenic, aluminium, uranium, and fluoride exceeded the respective drinking water regulations' parametric value given in Directive (EU) 2020/2184. Regarding the rare earth elements, neither WHO, EU, USA nor Canada have established any limit values. However, concentration of lanthanum in groundwater from a sedimentary well exceeded the health-based guideline value that applies in Australia. Results from this study raise the question of whether increased precipitation can have an impact on the mobility and concentration of uranium in groundwater from bedrock aquifers. Furthermore, findings of high levels of lanthanum in groundwater create uncertainty as to whether the current quality control of Norwegian drinking water is sufficient.

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## 1. Introduction

Sufficient and clean water is a prerequisite for good health. Since groundwater in general is well protected against disease-causing microorganisms, groundwater is considered a safer source of clean water compared to surface water. Still, the groundwater can be affected by natural processes and man-made activities. Groundwater may contain elements from the bedrock and minerals in concentrations that can have negative health effects. Groundwater exposed to granites and granitic gneisses may display an elevated content of fluorine, radon, and uranium (Reimann et al., 1996; Morland et al., 1997; Frengstad et al., 2000). Other elements that can have negative health effects, even in trace amounts, are arsenic, antimony, cadmium, and lead, however, such elements are rarely found in significant concentrations in Norwegian groundwater.

About 4.7 million of the Norway's inhabitants, i.e., close to 90 % of the country's population, are supplied with drinking water from a water supply system that supplies >50 permanent residents (Norwegian Food Safety Authority, 2019). The remaining part of the population receives drinking water from smaller waterworks and single household water supply systems. The quality of drinking water is regulated in the Norwegian drinking water regulations (HOD, 2016). The single household water supplies are also covered by the drinking water regulations, but they are not subject to the registration obligation, and the exact number, and how many are supplied, is therefore unknown. Approximately 10 % of the Norwegian population is supplied by smaller waterworks and private wells that are not registered in the Waterworks Register (VREG)/Norwegian Food Safety Authority's form services (MATS), and consequently have an unregulated drinking water quality.

The first Norwegian survey of the chemistry of groundwater was initiated in 1971 and included 507 groundwater samples from 98 bedrock boreholes in three areas located in south-eastern Norway (Englund and Myhrstad, 1980). Variations in specific electrical conductance largely reflected the different reservoir rock types. The survey also revealed that groundwater samples from below the late post-glacial marine limit to a variable extent were influenced by ancient sea salts. The next account of groundwater chemistry of bedrock wells in Norway was conducted in the mid-1990s by the Geological Survey of Norway (NGU) (Banks et al., 1995; Reimann et al., 1996; Morland et al., 1997; Midtgård et al., 2007). However, most of these studies were limited to either certain areas of Norway, or to specific chemical elements. A comprehensive study conducted by Banks et al. (1998a) included analysis of 1604 samples from water wells drilled in bedrock throughout Norway for determination of radon, major and minor elements, pH, and alkalinity. Regarding pH, sodium, radon, and fluoride together, 30 % of all wells were non-compliant with respect to one or more of these parameters (Banks et al., 1998a). This in contrast with groundwaters from Quaternary deposits where no exceedance of the maximum recommended concentrations for radon, fluoride, and sodium were noticed, while 10 % fell outside the required pH range (Banks et al., 1998b).

The European Parliament and the Council of the European Union (2020) require that by 12 January 2026, Member States shall take the measures necessary to ensure that water intended for human consumption complies with the parametric value of 30 µg/L stated for uranium in the Directive (EU) 2020/2184. According to data on groundwater quality published by the Geological Survey of Norway (NGU, 2020), water from one in eight privately owned bedrock wells in Norway exceed uranium levels that are considered safe. The rare earth elements (lanthanoids, scandium and yttrium) are not covered by the drinking water regulations; nevertheless, more knowledge about their occurrence and levels in Norwegian groundwater is needed.

Rainwater is the source of most groundwater, however, the evolution in water chemistry from rain hitting ground at the site of infiltration to the sampling point in the aquifer depend on residence time and is determined by hydrogeochemical processes like dissolution and precipitation reactions, reduction and oxidation, hydrolysis, ion exchange and complexation occurring along the groundwater flow path (Appelo and Postma, 2005).

Consequently, the groundwater's chemical composition will reflect precipitation, processes in the soil, pollution, marine salts, and reactions with minerals in the bedrock. As pointed out by Barbieri et al. (2023), literature data concerning the ways climate change affects groundwater quality are scarce. Increased rainfall and more intense rainfall events give cause for concern.

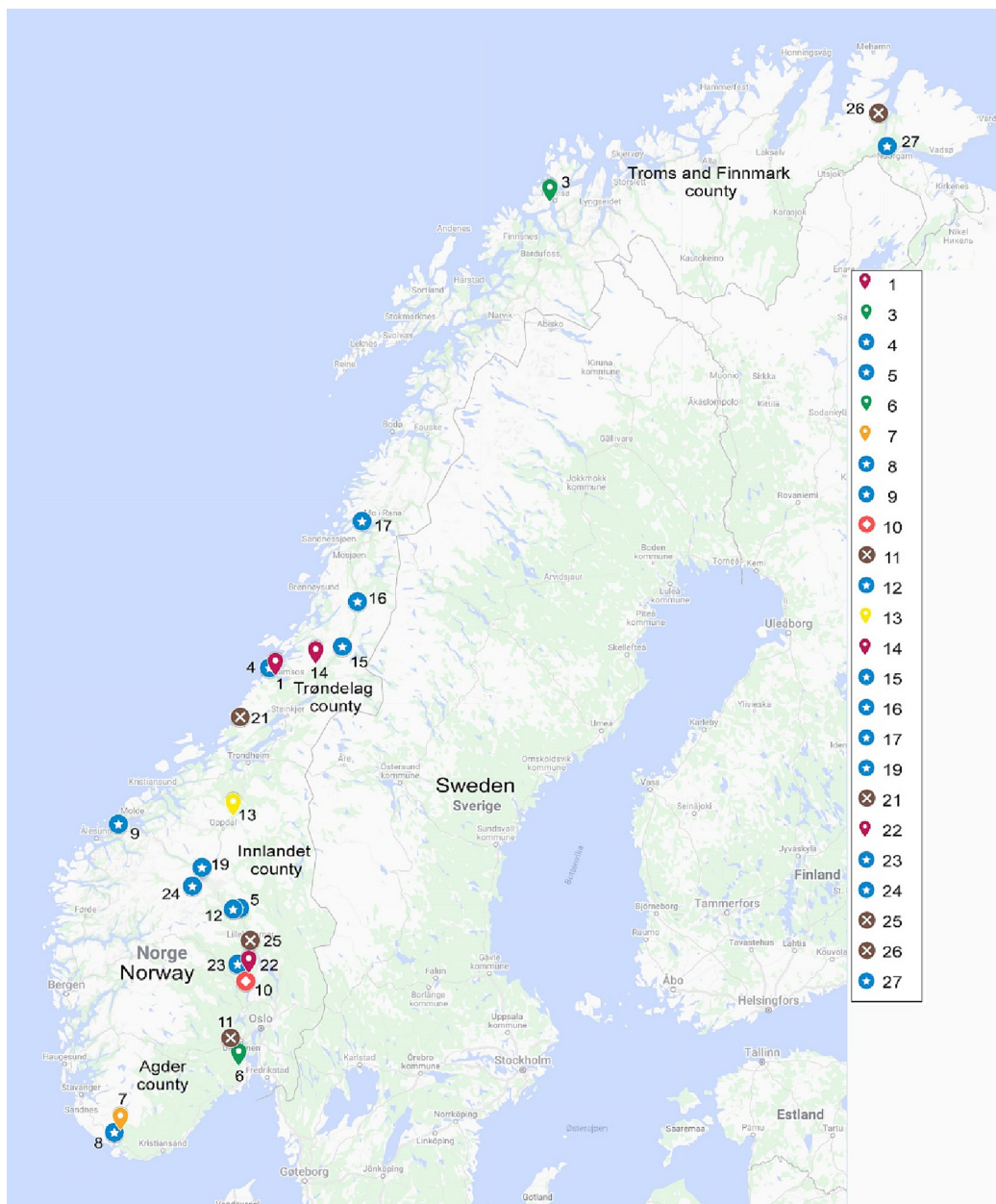
A nationwide survey of drinking water quality carried out in Norway in the period April–June 2016 (Abiyos, 2017), which included 201 groundwater sources spread over 111 municipalities, revealed samples with elevated concentrations of iron, manganese, arsenic, aluminium, and fluoride compared to the values in the drinking water regulation. In addition, some waterworks showed exceedances of guideline values or provisional guideline values for uranium set by the World Health Organization (WHO), USA, and Australia. Participating waterworks in a follow-up survey performed in 2019 (Joudi, 2020) were selected based on exceeded guideline values noticed in 2016. In addition, new waterworks with wells with similar geology were invited to participate. Many trace elements occur in very low concentrations in groundwater. By using Inductively Coupled Plasma Mass Spectrometry (ICP-MS), we could determine a wide range of elements down to ppt level (ng/L). In this work, 64 elements were determined, including lanthanoids, scandium and yttrium. Previous investigations on the chemical composition of Norwegian groundwater lack information on temporal variation, hence we wanted to follow up the results from the nationwide survey in 2016 with samples taken over an entire year. In the present work we aim to explore how factors such as geology, marine salt deposits, and changes in the amount of precipitation throughout the year affect the concentration of elements and anions in the groundwater.

## 2. Material and methods

### 2.1. Sampling of water

A total of 24 waterworks accepted the invitation to participate in the project and provided samples at five different times throughout the year, from February to November 2019. Sampling procedure, sampling tubes, and a sampling form were sent in advance. During predetermined periods of sampling, an employee at each waterwork collected two 15-mL samples of water from the source (raw water) and, if any kind of water treatment took place, two 15-mL samples of treated water from the distribution network. The samples were obtained in 15-mL polypropylene (PP) tubes. In addition, a parallel 200-mL Low Density Polyethylene (LDPE) bottle was filled with clean water from the distribution network. The water samples were immediately shipped by mail to the Norwegian University of Life Sciences, Aas, Norway. The 200-mL samples were temporary kept in refrigerated storage (1–4 °C) for 3–5 days until the determination of anions and physicochemical parameters took place. To prevent oxidation of iodine ( $I^-$ ) to iodate ( $I_2$ ), which is volatile, an alkaline sample preparation is necessary when determining iodine in water. The 15-mL samples, to which 0.6 mL of an alkaline solution was added, were analysed for chlorine, bromine, and iodine. The alkaline solution was prepared by mixing 25 mL 25 % (weight (w)/w) ammonium hydroxide ( $NH_4OH$ ) and 0.0625 g ethylenediaminetetraacetic acid (EDTA) and diluted to 50 mL with deionized water. The other elements were determined in the parallel 15-mL samples preserved with 0.75 mL sub boiled ultrapure 65 % (w/w) nitric acid ( $HNO_3$ ). Blank samples (five samples for each sample series) received the same pre-treatment as the other samples. Deionized water (>18 MΩ) and reagents of analytical grade or better were used throughout.

Waterworks in 21 municipalities, located throughout the country, from Troms and Finnmark in the north to Agder in the southernmost part of Norway, participated in the survey (Fig. 1). An overview of the various waterworks, type of water source, water treatment, number of people supplied, and bedrock geology is shown in Table 1. Information on bedrock geology for each sampling point was obtained from NGU's digital bedrock map, as background map to GRANADA, which is Norway's national groundwater database (NGU, 2018). In some cases, the bedrock is dominated by two or more main rocks. Hydrological data from nearby precipitation



**Fig. 1.** Participating waterworks are identified with numbers (see Table 1). The colour of the tags indicates exceedance of the drinking water regulations' parametric values given in Directive (EU) 2020/2184; brown: manganese and/or iron, yellow: arsenic, pink: aluminium, red: uranium, green: fluoride; blue: no exceedance. Orange tag show waterworks with high level of lanthanum (see Table 3).

stations were obtained from the Norwegian Centre for Climate Services (2022).

**2.2. Chemical analysis**

The water samples were analysed by means of an Agilent 8900 ICP-QQQ (Triple Quadrupole Inductively Coupled Plasma Mass Spectrometer; Agilent Technologies, Hachioji, Japan) for quantitative determination of 64 elements. The physico-chemical parameters pH, colour, turbidity, conductivity, and alkalinity were determined using Norwegian Standards available from Standard Online AS (<https://www.standard.no/en/>). The anions fluoride, chloride, sulphate, and nitrate were determined using ion chromatography (IC5000, Lachat Instruments, Burlington, Canada). Simultaneous analysis of reference materials from the National Institute of Standards and Technology (Gaithersburg, MD, USA) (NIST SRM 1640a Trace elements in natural water) and Environment and Climate Change

Canada (Environmental Science and Technology Laboratories Division, Ottawa, ON, Canada) (ION-96.4 lot 0618 A river water sample), ensured traceability and satisfactory accuracy in the methods. In addition, an in-house standard 1643h traceable to NIST SRM 1643e Trace elements in water was analysed. The in-house standard was also supplemented with several elements that were not included in the certificate for 1643e using certified standard solutions manufactured by Spectrascan (Teknolab as, Ski, Norway). The certified calibration solutions used were manufactured by Inorganic Ventures (Christiansburg, VA, USA).

**2.3. Statistics and data managements**

The limits of detection (LOD) and the limits of quantification (LOQ) for elements and anions were determined as three and ten times the standard deviation of the method blank, respectively. In the statistical analysis, values under LOD and LOQ were replaced with LOD/2 and

**Table 1**

Overview of participating waterworks, type of water source, water treatment, number of people supplied and main rock. See Fig. 1 for geographical location of the participating waterworks. Some samples from some waterworks are missing, either because the sample was not collected at the right time or because samples were lost in the mail. Considering Waterworks no. 2 and 18, we received no samples in time and this waterworks are omitted in Fig. 1.

Waterworks no.	Water source	Water treatment	People supplied	Main rock
1	Bedrock	UV <sup>a</sup>	160	Dioritic to granitic gneiss, migmatite
3	Bedrock	No treatment	62	Mica gneiss and mica schist, metasandstone, amphibolite
4	Bedrock	Sand filter	100	Dioritic to granitic gneiss, migmatite; mica gneiss and mica schist, metasandstone, amphibolite
5	Sedimentary	UV	50	Sandstone
6	Bedrock	Chlorine, UV	340	Rhomb porphyry; rhyolite, rhyodacite, dacite, ceratophyre
7	Sedimentary	No treatment	200	Amphibolite, hornblende gneiss, mica gneiss, locally migmatite
8	Sedimentary	UV, marble filter	600	Amphibolite, hornblende-gneiss, mica gneiss, locally migmatite
9	Sedimentary	No treatment	140	Amphibolite, hornblende gneiss, mica gneiss, locally migmatite
10	Bedrock	No treatment	110	Augen gneiss, granite; gabbro, amphibolite
11	Sedimentary	Sodium silicate, chlorine	11,775	Quartzite
12	Sedimentary	UV	386	Sandstone, shale
13	Bedrock	No treatment	107	Granite and granodiorite
14	Bedrock	No treatment	774	Dioritic to granitic gneiss, migmatite; augen gneiss, granite; mica gneiss and mica schist, metasandstone, amphibolite
15	Bedrock	No treatment	120	Mica gneiss and mica schist, metasandstone, amphibolite
16	Bedrock	UV	62	Mica gneiss and mica schist, metasandstone, amphibolite
17	Bedrock	Descaling	500	Calcareous mica schist, calc silicate gneiss
19	Bedrock	UV, sand filter	150	Quartz diorite, tonalite, trondhjemite
21	Sedimentary	UV, sand filter	59	Mica gneiss and mica schist, metasandstone, amphibolite
22	Bedrock	Sand filter	300	Amphibolite, hornblende gneiss, mica gneiss, locally migmatite
23	Bedrock	No treatment	66	Amphibolite, hornblende gneiss, mica schist, locally migmatite
24	Sedimentary	No treatment	230	Dioritic to granitic gneiss, migmatite
25	Sedimentary	UV, aeration	24,100	Sandstone
26	Bedrock	No treatment	25	Sandstone, shale
27	Sedimentary	No treatment	250	Sandstone, shale

<sup>a</sup> UV: treatment with ultraviolet light.

LOQ/2. Elements with >20 % concentration values below LOQ were discarded in the statistical analyses (Helsel, 2006). All statistical calculations were performed in Microsoft Excel (Microsoft 365 Apps for Enterprise, version 2210). To examine the variation in the data set, and possibly look for trends and common features in the various water samples, principal component analysis (PCA) was performed using the XLSTAT, a statistical add-on for Microsoft Excel (XLSTAT by Addinsoft, version 2019.3.2). Since the data were not normally distributed, Spearman's correlation analysis was performed in XLSTAT. For all statistical analysis, a confidence interval of 95 % was used, which corresponds to a significance level ( $\alpha$ ) equal to 0.05. For hypothesis testing, a linear regression model in the statistical program R studio (RStudio Team, 2016) was used.

### 3. Results

#### 3.1. Data basis for statistical analysis

An overview of LOQ values and the percentage of analysis results less than LOQ are shown in Table 2. With respect to beryllium (Be), chromium (Cr), selenium (Se), zirconium (Zr), niobium (Nb), silver (Ag), indium (In), tin (Sn), tungsten (W), tellurium (Te), and bismuth (Bi), all elements showed high percentages (i.e., >20 %) below LOQ in all measurement series and are therefore not included in any statistical analysis. The elements phosphorus (P), titanium (Ti), gallium (Ga), germanium (Ge), cadmium (Cd), antimony (Sb), terbium (Tb), lutetium (Lu), thallium (Tl), and thorium (Th) have occasionally high percentages below LOQ, and the respective elements were therefore in time series where relevant omitted from the statistical analysis. In particular, the elements aluminium (Al), P, Ti, iron (Fe), copper (Cu), Ga, Ge, Cd, Sb, Tb, Lu, Tl, Th, iodine (I) and the anions fluoride ( $F^-$ ) and nitrate ( $NO_3^-$ ) all showed great temporal variation in terms of percentage below LOQ. The results from the periods April–May and August–September 2019 were used for Spearman's correlation analysis as the percentage of analysis results below the LOQ were roughly the same and therefore most of the elements were involved in the correlation analysis.

Some samples from some waterworks are missing, either because the sample was not collected at the right time or because samples were lost in the mail. In the period February–March 2019, however, water samples were received from all waterworks and the results from this period were further selected in the PCA analysis where all waterworks (observations) were included in the statistical analysis.

#### 3.2. Chemical composition of groundwater from wells in bedrock and in unconsolidated Quaternary sediments

The data set from the period February–March 2019 had no missing samples and the results from this period were chosen as the data basis in the principal component analysis where all waterworks (observations) are included in the statistical analysis. Principal components 1 and 2 (PC1 and PC2) explained 56 % of the variations in the data set (Fig. 2). The loading plot in Fig. 2 shows a distribution of the parameters in water samples where the elements aluminium, cobalt, and rare earth elements (REEs) are correlated with PC1, while the elements lithium, boron, sodium, magnesium, potassium, molybdenum, caesium, and uranium, as well as the parameters alkalinity, conductivity, and pH are strongly correlated with PC2. The trends present a clear difference in concentrations of these elements between the different water samples. The score plot shows the results among waterworks two-dimensionally, where most waterworks with bedrock wells are located towards the top, while sedimentary waterworks are located towards the bottom. That is, PC1 and PC2 were effective in separating the two types of drinking water sources. Waterworks located at the top present groundwater with higher concentrations of lithium, boron, sodium, magnesium, potassium, calcium, vanadium, rubidium, strontium, caesium, and uranium, as well as higher conductivity, alkalinity, and pH.

#### 3.3. Exceedances of limit values

The results from the physico-chemical analysis showed that all waterworks, except one, meet the drinking water regulations (HOD, 2016). Waterworks no. 7 in Agder county has the lowest median value for pH (5.4), which is outside the drinking water regulations' limit of 6.5–9.5.

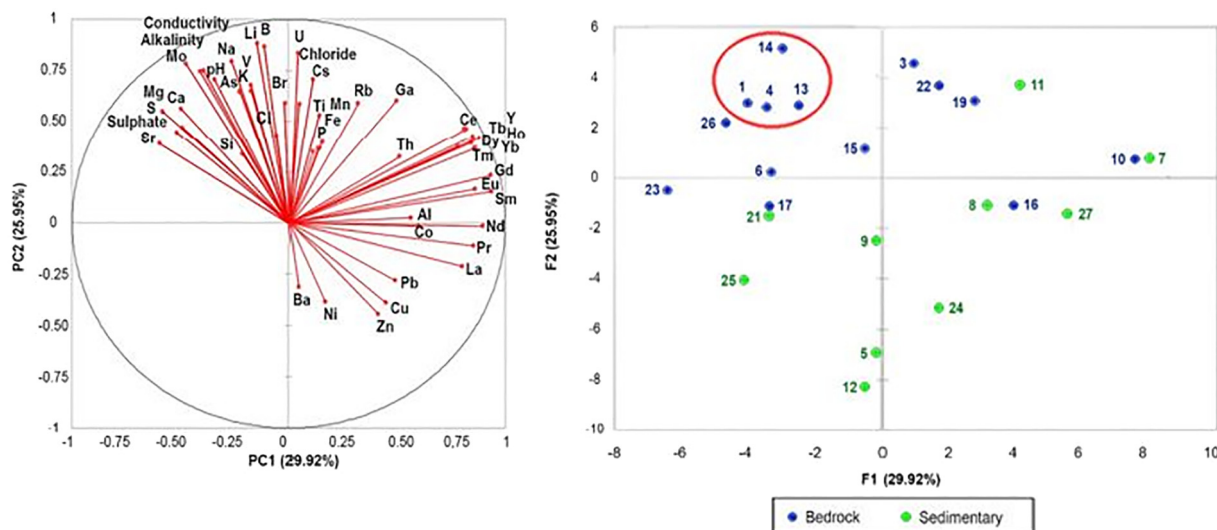
**Table 2**

Limits of quantification limit (LOQ) and percentage below the limits of quantification (% <LOQ) for the elements and anions determined in groundwater samples collected at five different time periods throughout the year, February–March, April–May, June, August–September, and October–November. The first three sample series were analysed separately, while the two last sample series were analysed simultaneously, thus, the two columns at the right have equal LOQ.

Element and anion	Feb–March		April–May		June		Aug–Nov	Aug–Sep	Oct–Nov
	LOQ (µg/L)	%<LOQ	LOQ (µg/L)	%<LOQ	LOQ (µg/L)	%<LOQ	LOQ (µg/L)	%<LOQ	%<LOQ
Li	0.01	0	0.01	4	0.015	9	0.01	5	0
Be	0.004	57	0.014	52	0.081	45	0.005	40	44
B	1.9	7	1.9	17	1.8	9	3.2	5	22
Na	3.7	0	4	0	7.4	0	6	0	0
Mg	0.9	0	1	0	0.69	0	0.8	0	0
Al	1.3	16	1.1	0	0.51	0	2.2	10	28
Si	3	0	4	0	1.4	0	4	0	0
P	3.6	0	0.5	48	1.2	81	3.5	80	83
S	1.2	0	2	0	1.1	0	1.7	0	0
K	3.3	0	7	0	12	0	8.3	0	0
Ca	4	0	10	0	1.4	0	6	0	0
Sc	0.001	0	0.001	0	0.003	0	0.001	0	0
Ti	0.05	16	0.06	43	0.02	13	0.07	45	50
V	0.0013	0	0.005	4	0.002	0	0.003	0	5
Cr	0.03	30	0.03	26	0.03	22	0.04	35	44
Mn	0.012	4	0.03	0	0.03	9	0.04	5	5
Fe	0.49	11	0.3	4	0.2	0	0.64	10	30
Co	0.003	11	0.004	0	0.0005	0	0.001	0	0
Ni	0.088	11	0.022	0	0.05	9	0.93	10	17
Cu	0.04	0	1.1	26	0.06	0	0.45	15	17
Zn	0.058	0	0.09	0	0.17	9	0.18	10	11
Ga	0.0012	0	0.006	56	0.003	45	0.003	20	28
Ge	0.0078	50	0.004	0	0.01	27	0.01	55	56
As	0.005	0	0.01	8	0.005	0	0.004	0	0
Se	0.047	42	0.02	22	0.04	31	0.1	65	61
Rb	0.039	0	0.02	0	0.02	0	0.02	0	0
Sr	0.007	0	0.02	0	0.01	0	0.01	0	0
Y	0.0002	0	0.0005	0	0.0002	0	0.0005	0	0
Zr	0.002	23	0.001	26	0.002	20	0.005	30	44
Nb	0.001	77	0.001	56	0.001	77	0.001	75	77
Mo	0.012	4	0.04	13	0.017	9	0.02	5	5
Ag	0.0023	73	0.005	87	0.0016	57	0.002	55	100
Cd	0.0036	50	0.002	13	0.0006	9	0.002	25	17
In	0.002	100	0.0004	83	0.0007	54	0.007	100	100
Sn	0.01	88	0.014	83	0.006	90	0.02	95	95
Sb	0.007	35	0.007	22	0.001	4.5	0.005	15	17
Te	0.0072	100	0.005	100	0.004	86	0.004	100	100
Cs	0.0006	19	0.001	17	0.0006	9	0.0005	5	17
Ba	0.029	0	0.07	0	0.037	0	0.019	0	0
La	0.0003	0	0.0002	0	0.0002	0	0.0007	0	0
Ce	0.0002	0	0.0006	4	0.0002	0	0.0004	0	0
Pr	0.00008	0	0.0002	0	0.0001	0	0.0004	0	0
Nd	0.00084	0	0.001	0	0.0002	0	0.002	0	0
Eu	0.001	4	0.001	0	0.0016	9	0.0007	0	0
Gd	0.0003	0	0.0006	0	0.0002	0	0.0004	0	0
Tb	0.0003	4	0.0002	17	0.0003	4.5	0.0004	45	40
Dy	0.0003	0	0.0004	0	0.0007	0	0.0017	0	0
Ho	0.0004	11	0.0003	17	0.0002	0	0.0005	15	11
Er	0.0005	0	0.0005	0	0.0002	0	0.0004	0	0
Tm	0.0034	11	0.0003	0	0.0001	0	0.0002	15	11
Yb	0.0026	16	0.0012	0	0.0002	0	0.0002	0	0
Lu	0.0006	23	0.0004	26	0.0004	0	0.0003	5	5
W	0.004	30	0.006	23	0.0035	40	0.004	20	44
Tl	0.0012	0	0.0014	43	0.0004	9	0.001	25	16
Pb	0.005	0	0.009	4	0.0013	4.5	0.007	15	11
Bi	0.0021	96	0.0013	82	0.0007	86	0.005	100	100
Th	0.0003	7	0.0007	39	0.0049	45	0.001	50	60
U	0.0004	0	0.0003	0	0.0008	0	0.0008	0	0
Cl	0.1	0	0.1	0	0.1	0	0.1	0	0
Br	0.31	0	0.68	0	0.53	0	0.58	0	0
I	0.83	38	0.27	4	0.37	9	0.56	15	16
F <sup>-</sup>	0.04	24	0.04	0	0.04	28	0.04	11	11
Cl <sup>-</sup>	0.4	0	0.4	0	0.4	0	0.4	0	0
SO <sub>4</sub> <sup>2-</sup>	0.6	0	0.6	0	0.6	0	0.6	0	0
NO <sub>3</sub> <sup>-</sup>	0.1	20	0.1	0	0.1	42	0.1	42	21

Table 3 shows an overview of measured values (median value, minimum value, and maximum value) for elements that exceed limit values in accordance with the drinking water regulations and guidelines from other

countries. This applies to the elements aluminium, manganese, iron, arsenic, lanthanum and uranium, and the anion fluoride measured in raw water and after water treatment. For a statistical analysis to be able to



**Fig. 2.** Results from PCA shown as loading plot (left) and score plot (right) for elements, anions, and the parameters conductivity, alkalinity, and pH in groundwater collected in the period February–March 2019. The red circle in the score plot shows waterworks located in Trøndelag county (see text for further explanations).

give a significant median value, at least five measurements are required in the data set. For waterworks that do not meet this requirement, the data are presented as minimum value and maximum value.

**3.4. Seasonal variation in element concentrations**

The results from the regression analysis for concentrations of aluminium, manganese, arsenic, lanthanum, uranium, and fluoride are summarized in Table 4. Six waterworks exceeded the drinking water regulations' limit value for manganese, but only waterworks no. 21 in Trøndelag county showed significant seasonal variation ( $p = 0.001$ ) in the concentration of

this element. Otherwise, no significant seasonal variation was found at the waterworks with exceedances of any elements this year.

**3.5. Relationship between precipitation and element concentrations**

With one exception, the regression analysis of the relationship between precipitation and element concentrations shows no significant correlations (Table 5). For uranium, a significant correlation ( $p = 0.03$ ) was found at waterworks no. 14A in Trøndelag county. The amount of precipitation for each individual waterworks for the periods January–February, April–May, June, August–September, and October–November in the year 2019

**Table 3**

Elements and anion with exceedance in accordance with the drinking water regulations and other national values. Limit values are taken from Norway (HOD, 2016; with reference to the EEA Agreement – Annex XX Environment and Directive (EU) 2015/1787), the World Health Organization (WHO, 2017), the United States of America (USEPA, 2018), and Australia (NHMRC and NRMCMC, 2011).

Waterworks no.	Element and ion	n	Raw water median (min–max)	After treatment median (min–max)	Limit value			
					Norway	WHO	USA	AUS
7	Al (µg/L)	5	310 (300–330)	NT	200 <sup>a</sup>	900 <sup>b</sup>	200 <sup>c</sup>	200 <sup>c</sup>
10	Al (µg/L)	2	(160–270)	NT	200 <sup>a</sup>	900 <sup>b</sup>	200 <sup>c</sup>	200 <sup>c</sup>
11	Mn (µg/L)	5	100 (63–110)	100 (58–110)	50 <sup>a</sup>	400 <sup>b</sup>	50 <sup>c</sup>	50
13	Mn (µg/L)	5	79 (47–100)	NT	50 <sup>a</sup>	400 <sup>b</sup>	50 <sup>c</sup>	50
21	Mn (µg/L)	5	300 (240–410)	21 (12–72)	50 <sup>a</sup>	400 <sup>b</sup>	50 <sup>c</sup>	50
22	Mn (µg/L)	2	(210–240)	(1.9–2.6)	50 <sup>a</sup>	400 <sup>b</sup>	50 <sup>c</sup>	50
25	Mn (µg/L)	5	120 (77–170)	80 (16–110)	50 <sup>a</sup>	400 <sup>b</sup>	50 <sup>c</sup>	50
26	Mn (µg/L)	2	(120–130)	NT	50 <sup>a</sup>	400 <sup>b</sup>	50 <sup>c</sup>	50
22	Fe (µg/L)	2	(910–1500)	(36–38)	200 <sup>a</sup>	2000 <sup>b</sup>	300 <sup>c</sup>	300 <sup>c</sup>
26	Fe (µg/L)	2	(760–790)	NT	200 <sup>a</sup>	2000 <sup>b</sup>	300 <sup>c</sup>	300 <sup>c</sup>
13	As (µg/L)	5	14 (13–14)	NT	10	10	10	10
7	La (µg/L)	5	150 (140–150)	NT	–	–	–	2
1	U (µg/L)	5	28 (26–29)	28 (26–29)	30 <sup>a,d</sup>	30	30	17
14A	U (µg/L)	5	84 (80–86)	NT	30 <sup>a,d</sup>	30	30	17
14B	U (µg/L)	5	63 (59–66)	NT	30 <sup>a,d</sup>	30	30	17
22	U (µg/L)	2	(35–38)	(32–34)	30 <sup>a,d</sup>	30	30	17
1	F <sup>-</sup> (mg/L)	5	1.6 (1.5–1.7)	1.6 (1.5–1.7)	1.5	1.5	4.0	1.5
3	F <sup>-</sup> (mg/L)	3	5.0 (1.1–5.3)	NT	1.5	1.5	4.0	1.5
6	F <sup>-</sup> (mg/L)	5	1.8 (1.6–1.9)	1.8 (1.6–1.9)	1.5	1.5	4.0	1.5

n: number of water samples; NT: No treatment.

<sup>a</sup> Action limit.

<sup>b</sup> Health-based guideline value.

<sup>c</sup> Aesthetic guideline value.

<sup>d</sup> Member States shall take the measures necessary to ensure that water intended for human consumption complies with the parametric value by 12 January 2026 (Directive (EU) 2020/2184).

**Table 4**

Results from regression analysis of concentrations of the elements aluminium (Al), manganese (Mn), arsenic (As), lanthanum (La), uranium (U), and the anion fluoride (F<sup>-</sup>) measured over time (seasonal variation).

Waterworks no.	Element and ion	n	$\beta_{Trend}$	SE	R <sup>2</sup>	P
7	Al	5	2.0	4.0	0.07	0.6
13	Mn	5	-1	7.4	0.006	0.90
11	Mn	5	3.7	7.2	0.08	0.6
21	Mn	5	42.0	3.8	0.97	0.001*
25	Mn	5	6.8	13.5	0.07	0.6
13	As	5	-0.2	0.1	0.5	0.18
7	La	5	$7.5 \times 10^{-15}$	2	0	1.0
1	U	5	-0.30	0.4	0.17	0.46
14A	U	5	0.20	0.87	0.01	0.83
14B	U	5	-0.6	1.1	0.09	0.6
3	F <sup>-</sup>	3	-0.1	2.3	0.004	0.9
1	F <sup>-</sup>	5	$7.8 \times 10^{-15}$	$3.1 \times 10^{-2}$	0	1.0
6	F <sup>-</sup>	5	-0.06	0.02	0.75	0.06

$\beta_{Trend}$ : regression coefficient. SE: standard error of the mean. R<sup>2</sup>: the square of the correlation coefficient.

\* Significant (p < 0.05).

was estimated based on precipitation data from nearby measuring stations (the Norwegian Centre for Climate Services, 2022).

### 3.6. Other factors that may be relevant to the chemical composition of groundwater

Spearman's correlation analysis was performed on the total depth of the bedrock wells and the parameters in groundwater measured in the periods April–May and August–September 2019, time periods where the percentage of analysis results under LOQ was approximately equal and consequently most elements were included in the correlation analysis. Results from the analysis show that the total depth of the well, i.e., the sum of the thickness of soil over bedrock and the depth of the borehole in the bedrock, was significantly positively correlated with the concentration of calcium (p = 0.02), manganese (p = 0.04), rubidium (p = 0.003), and caesium (p < 0.0001), and significantly negatively correlated with the elements chlorine (p = 0.008) and bromine (p = 0.02), and the anion chloride (p = 0.004).

The highest median concentration for fluoride (5 mg/L) was measured at waterworks no. 3 in Troms and Finnmark county. Spearman correlation analysis showed a significant relationship (p = 0.04) between sodium and fluoride in the groundwater.

## 4. Discussion

The present study includes both municipal and private waterworks, and with one exception all the waterworks are registered in the VREG/MATS. The two largest waterworks, both of which supply >10,000 residents, have water sources from unconsolidated Quaternary sediments. A total of

14 of the waterworks have water sources from bedrock aquifers. The median value for number of persons supplied by the participating waterworks is 155 (Table 1).

### 4.1. The chemical composition of the drinking water and exceedances of limit values

New formation or supply of groundwater takes place through inflows from rivers and lakes and through infiltration of precipitation and meltwater. In unconsolidated Quaternary sediments, the water flows between the soil particles, while in rocks, the water moves through heterogeneously distributed open fractures and cavities that reach the surface of the bedrock. Some rocks can contribute to increased concentrations of certain elements in the groundwater, but minerals precipitated along fractures and in cavities may be more significant for the chemical composition of the water than the rock minerals. The results from the PCA analysis show clear differences in the chemical composition of the groundwater collected from the bedrock and the sedimentary wells (Fig. 2). Compared to the sedimentary wells, the groundwater in bedrock wells has higher concentrations of lithium, boron, sodium, magnesium, potassium, calcium, vanadium, rubidium, strontium, caesium, and uranium, in addition to higher conductivity, alkalinity, and pH. This observation is consistent with a previous study on inorganic components in groundwater in Norway (Seither et al., 2012).

The chemical composition of groundwater is determined by hydrogeochemical processes, i.e., by reactions such as dissolution and precipitation, reduction and oxidation, hydrolysis, ion exchange and complexation (Appelo and Postma, 2005). Rainwater is naturally acidic and oxidizing. As water moves through the pedosphere, natural processes like root

**Table 5**

Results from regression analysis of the ratio between the amount of precipitation (millimetre) and concentrations of the elements aluminium (Al), manganese (Mn), arsenic (As), lanthanum (La), uranium (U), and the anion fluoride (F<sup>-</sup>).

Waterworks no.	Element and ion	n	$\beta_{Trend}$	SE	R <sup>2</sup>	P
7	Al	5	-0.02	0.03	0.2	0.4
13	Mn	5	-0.03	0.09	0.04	0.7
11	Mn	5	0.2	0.1	0.6	0.1
21	Mn	5	0.2	0.2	0.2	0.4
25	Mn	5	0.2	0.2	0.2	0.3
13	As	5	0.001	0.002	0.08	0.6
7	La	5	-0.01	0.01	0.1	0.5
1	U	5	0.005	0.003	0.4	0.2
14A	U	5	0.02	0.005	0.8	0.03*
14B	U	5	0.003	0.01	0.02	0.8
3	F <sup>-</sup>	3	0.02	0.02	0.6	0.4
1	F <sup>-</sup>	5	-0.0004	0.0003	0.6	0.1
6	F <sup>-</sup>	5	-0.0006	0.0003	0.5	0.1

$\beta_{Trend}$ : regression coefficient. SE: standard error of the mean. R<sup>2</sup>: the square of the correlation coefficient.

\* Significant (p < 0.05).

exudation can lower the pH further. Most rocks consist of minerals that are alkaline and reducing and newly formed groundwater can therefore react actively with the bedrock, but since many reactions are very slow, the water's residence time will basically be of great importance for the content of solutes. Groundwater in bedrock is usually less acidic than groundwater in unconsolidated Quaternary sediments. Waterworks no. 5, 7, 9, 12, and 24, all sedimentary wells, and waterworks no. 10 with groundwater from bedrock, showed median pH < 7.0 (data not shown).

Several elements occur in very low concentrations and some elements and anion show great variation over time regarding the percentage below LOQ (Table 2). On the other hand, the elements aluminium, manganese, iron, arsenic, lanthanum and uranium, and the anion fluoride, however, exceeded the drinking water regulations' limit values or guideline values from other countries (Table 3).

A survey conducted by Seither et al. (2012) indicate that 10 % of Norwegian groundwater wells in both bedrock and unconsolidated Quaternary sediments have iron content above the limit value of the drinking water regulations, while for manganese approximately 28 % of the bedrock wells and 21 % of the sedimentary wells show such exceedance. In this study, two bedrock wells showed exceedance of the drinking water regulations' limit value for iron, while three bedrock wells and three sedimentary wells showed exceedance of the limit value for manganese (Table 1 and Table 3). Solubility of iron and manganese in groundwater depends on pH and redox conditions of the groundwater. Manganese is more soluble than iron. With access to air, iron and manganese can precipitate and the precipitation rate will increase with higher pH in the groundwater, especially for manganese, when the pH is above 8. Wells with high values of manganese showed pH in the distributed water in the range 7.2–8.1. Waterworks no. 21 in Trøndelag county showed significant seasonal variation ( $p = 0.001$ ) in the concentration of manganese in groundwater from sedimentary wells (Table 4), probably explained by changes in redox conditions and pH in the groundwater. Regarding seasonal variation in element concentration, Frengstad (2002) noticed that the chemical composition of crystalline bedrock groundwaters is relatively stable, unless there is a direct or indirect intrusion of water of short residence time and "surficial nature" via poor well-head sealing or shallow transmissive fractures. In such cases the chemistry can vary dramatically with rain or snowmelt events.

Heavy metals, i.e., elements with a specific weight greater than or equal to  $5 \text{ g/cm}^3$ , are rarely a problem in groundwater, probably due to low solubility at neutral to moderate alkaline pH. Considering arsenic, one waterworks (no. 13), located in Trøndelag county, exceeded the drinking water regulations limit value of  $10 \text{ }\mu\text{g/L}$  with a median (minimum-maximum) equal to 14 (13–14)  $\mu\text{g/L}$ . The median pH of the groundwater was 8.0. Waterworks no. 13 has its water source from bedrock. The waterworks has no treatment of the raw water (Tables 1 and 3). The main rocks in the area are granite and granodiorite. Elevated concentration of arsenic in drinking water from crystalline bedrock groundwater in Norway is, however, rare. In a study conducted by Frengstad et al. (2000), a maximum concentration of  $19 \text{ }\mu\text{g/L}$  was recorded and 7 of the 476 samples (1.5 %) exceeded the limit value. Arsenic was controlled by pH, but as opposed to cadmium, lead, nickel, and zinc, an increase in concentration of arsenic with increasing pH was noticed (Frengstad et al., 2000).

Two waterworks, both without treatment of the raw water, showed exceedances for aluminium (Table 3). Waterworks no. 10 in Innlandet county has its water source from bedrock with main rock augen gneiss, granite, gabbro, and amphibolite, while waterworks no. 7 in Agder county has its water source from unconsolidated Quaternary sediments and the main rocks in the area are amphibolite, hornblende gneiss and mica gneiss, locally migmatite (Table 1). The median value for pH (5.4) in groundwater from the sedimentary well (no. 7) is below the lower limit of the Drinking Water Regulations of 6.5. The groundwater from this sedimentary well also shows high levels of lanthanum. In acidic water and until pH 8, without any other oxyanions present lanthanum is mainly present as a trivalent positive ion, while at pH < 5 dissolved aluminium is mainly found as  $\text{Al}(\text{H}_2\text{O})_6^{3+}$  (D'Haese et al., 2019). With rising pH, the insoluble  $\text{Al}(\text{OH})_3$  forms, which has an amphoteric character and a tendency to form complex ions

and to polymerise. As pointed out by D'Haese and co-authors, the speciation of aluminium is remarkably like that of lanthanum, albeit with most similar Al-OH species offset by 4–5 pH units lower. The solubility of sources of aluminium and lanthanum is highly dependent on pH and temperature, in addition to ligands present and amount of these. Dissolved aluminium is found in every natural water body due to chemical erosion of minerals of the feldspar group. The lanthanoids are found everywhere in the earth's crust, including in massive rocks such as basalts, granites, and gneiss, but only in very small concentrations (Henderson, 1984; Yaroshevsky, 2006).

Seither et al. (2012) noticed elevated REE concentrations in groundwater from Quaternary aquifers in the southernmost Norway. Considering lanthanum, bedrock groundwaters in general provided lower concentrations than groundwaters from unconsolidated sediments. All groundwater samples with concentrations of lanthanum above  $10 \text{ }\mu\text{g/L}$  were from sediment wells located in the western part of Agder County and a distribution of lanthanum that very similar to that of cerium (Ce) and holmium (Ho) was noticed (Seither et al., 2012). In the present work, the presence of lanthanum in groundwater was more dependent on the parameters pH, alkalinity, and water hardness, than the geological conditions. The well with the highest measured median concentration for lanthanum ( $150 \text{ }\mu\text{g/L}$ , in waterworks no. 7; Table 3), also located in Agder county, showed the highest median concentrations of Ce ( $130 \text{ }\mu\text{g/L}$ ), neodymium (Nd;  $95 \text{ }\mu\text{g/L}$ ), and praseodymium (Pr;  $26 \text{ }\mu\text{g/L}$ ).

The toxicity of lanthanum primarily depends on the inorganic salt with which it occurs. Lanthanum forms soluble chlorides and nitrates, while the phosphates and carbonates are generally insoluble, as lanthanum has a strong preference for oxygen-containing anions. In the acid environment of the upper gastrointestinal tract, soluble lanthanum salts and the less soluble carbonates, dissociate into their ionic forms. Within the lumen of the gut, lanthanum binds to form insoluble lanthanum phosphate, which is eliminated through fecal excretion (D'Haese et al., 2019). Because of its low solubility and accordingly low potential for systemic absorption, lanthanum carbonate was preferred to lanthanum chloride in investigations on its therapeutic use as an intestinal phosphate binder in dialysis patients. According to Hutchison et al. (2016), significant toxic effects were absent when lanthanum carbonate was used therapeutically. There is, however, an ongoing discussion in the literature as to whether ingestion of lanthanum chloride might be more toxic than lanthanum carbonate (Behets et al., 2020). Negative effects on the animals' liver, kidney, and nervous system are reported in experimental studies in which lanthanum chloride was administered orally (Briner et al., 2000; Feng et al., 2006; He et al., 2008; Huang et al., 2011; Zheng et al., 2013; Cheng et al., 2014; Jin et al., 2017). For preventive health reasons, in 2014 the Australian authorities enforced a guideline value for lanthanum ( $2 \text{ }\mu\text{g/L}$ ; Table 3) (NICNAC, 2014). Regarding potential gastrointestinal absorption of lanthanum from drinking water or leisure activities, the extent, and possible harmful consequences of lanthanum accumulation in humans are still uncertain. Neither WHO, EU, USA nor Canada has set limit values for any of the elements in the REE group.

Uranium is one of 15 metallic elements in the actinoid series in the periodic system. Of the actinoids, primordial thorium and uranium occur naturally in substantial quantities. According to Frengstad (2020), the concentration of uranium in Norwegian groundwater wells from unconsolidated sediments are very low, while for groundwater in bedrock, one of eight wells shows exceedance of the preliminary limit of  $30 \text{ }\mu\text{g/L}$  set by WHO (2017). Previous studies of groundwater wells in bedrock in Norway show that the concentration of uranium varies from  $<0.001 \text{ }\mu\text{g/L}$  up to  $750 \text{ }\mu\text{g/L}$  (Frengstad et al., 2000). In this study, waterworks no. 22 located in Innlandet county and waterworks no. 14 A and B located in Trøndelag county showed exceedance of the newly established parametric value of  $30 \text{ }\mu\text{g/L}$  for uranium in the Directive (EU) 2020/2184 (Table 3). The first waterworks showed concentrations of uranium in well water slightly above the limit, while the last waterworks showed uranium concentration ranging from 59 to  $86 \text{ }\mu\text{g/L}$ . Referring to the national groundwater database, GRANADA (NGU, 2018), the drilling log from one well at waterworks no. 14 mentions dark grey drill tailings indicating the possible



occurrence of Cambrian black shale, locally found at this bedrock level, a rock known to contain high concentrations of U. A global overview of national regulations and standards for drinking water quality states that uranium is one of the least specified parameters (WHO, 2018); with only 17 out of 104 countries and territories setting a regulatory or guideline value. According to epidemiological studies on oral exposure to natural uranium referred to by Health Canada (2019), only kidney effects have been consistently found in humans, although recent studies have also shown effects in bone. For total natural uranium in drinking water, Canada maintains a maximum acceptable concentration of 20 µg/L, a limit developed based on studies of kidney effects in rats. USA has set an upper limit of 30 µg/L (USEPA, 2018), while Australia has set the upper limit at 17 µg/L (NHMRC and NRMCMC, 2011).

Fluoride in the right amounts has a positive effect on dental health, but systemic exposure to fluoride through drinking water is associated with an increased risk of dental and bone fluorosis in a dose-response manner without a detectable threshold (SHER, 2010). The drinking water regulations' limit for fluoride is 1.5 mg/L. Fluorine is released to groundwater by weathering or anion exchange at high pH (Frengstad et al., 2001). Granites and granitic gneisses are risk rocks for fluoride, and up to 8.26 mg/L have previously been measured in groundwater samples from a crystalline bedrock aquifer in southwestern Norway (Banks et al., 1998a). A sporadic occurrence of dental fluorosis in children and skeletal fluorosis in cattle using bedrock groundwater was noticed in south and western Norway (Midtgård et al., 1997). Groundwater with a low calcium content and a high pH (pH > 8) often contains increased levels of fluoride (Banks et al., 1998a). In this study, the highest median concentration of fluoride (5.0 mg/L) was measured at waterworks no. 3 in Troms and Finnmark county. The waterworks is located near the coast in an area with bedrock of mica-gneiss and -schist, metasandstone, and amphibolite.

#### 4.2. What factors affect the water quality of groundwater?

Type of rock, depth of well in bedrock, fracturing, and soil coverage over bedrock can affect the quality of groundwater. Other factors that may be important for water quality of groundwater are precipitation and marine salts (Domenico and Schwartz, 1997; Barbieri et al., 2019; Ricolfi et al., 2020).

In groundwater collected in the periods April–May and August–September 2019, a significantly positive correlation was found between total depth of well in bedrock and concentration of calcium ( $p = 0.02$ ), manganese ( $p = 0.04$ ), rubidium ( $p = 0.003$ ), and caesium ( $p < 0.0001$ ), while depth of well was significantly negatively correlated with the concentration of chlorine ( $p = 0.008$ ), bromine ( $p = 0.02$ ), and the anion chloride ( $p = 0.004$ ). The reasons for these results may be low oxygen content, longer residence time, and less leakage of marine salts into deeper bedrock wells. Total depth of well is the sum of depth to bedrock, i.e., thickness of overburden of unconsolidated sediments, and borehole length in bedrock. The information about the bedrock wells was obtained from GRANADA (NGU, 2018).

The results from this work indicate that the presence of fluoride in groundwater is more affected by hydrological and local conditions than geological conditions. The highest fluoride concentration (5.3 mg/L) was measured at waterworks no. 3 in Troms and Finnmark county. A significant positive correlation between fluoride and sodium ( $p = 0.04$ ) in groundwater from bedrock wells suggest that the concentration of fluoride in groundwater in near-coastal areas is probably more affected by sea salts in precipitation rather than by bedrock geology.

In most parts of Europe, heavy precipitation events are likely to become more frequent, and according to the European Environment Agency (2021), the projected changes are strongest in Scandinavia and northern Europe in winter. Precipitation contains natural gases such as oxygen and carbon dioxide. Microorganisms in the soil use oxygen in respiration and produce carbon in the form of carbon dioxide, which dissolves in the pore water and gives an acidic solution. Newly formed groundwater can react actively with the bedrock and release elements from minerals into the

groundwater, especially elements that are easily soluble under oxidizing conditions (Frengstad, 2013; Frengstad and Banks, 2014). Uranium is an element that shows high solubility under oxidizing conditions, especially at low pH. Under oxidizing conditions oxidation stage + VI is most stable and uranium will then usually occur as the uranyl ion  $UO_2^{2+}$ , while under reducing conditions, uranium is most stable at oxidation stage + IV as  $UO_2(s)$  (Vitorge et al., 2007). In this study, a significant correlation was found ( $p = 0.03$ ) between the concentration of uranium and the amount of precipitation at waterworks no. 14A. Transport of uranium usually takes place in oxidized water, such as uranyl ion or uranyl fluorides, phosphate, or carbonate complexes (Langmuir, 1978). Increased precipitation can therefore have an impact on the mobility and concentration of uranium in the groundwater. Despite more precipitation, higher temperatures and increased evaporation can pose a risk of drought in the summer, which in turn results in lower groundwater levels (NCCS, 2015). In regions with cold climates, in which snow accumulation plays an important role in the seasonal hydrological cycle, anomalies in air temperature also influence hydrological drought development and recovery (Van Loon et al., 2015). Thus, both precipitation and temperature may affect the drinking water quality. This could affect the drinking water quality by chemical reactions due to increased redox fluctuations. Therefore, it is important to perform a competent assessment of the development of element concentration in well water over a longer period. The amounts of precipitation that is used as a basis in the regression analysis in this work are based on estimates. In a future study, it is important to plan for access to measured meteorological data from the precipitation fields to assess climate change as another driver that would change the water quality.

#### 5. Conclusions

Several chemical elements investigated in this work occur in very low concentrations. However, the concentration of aluminium, manganese, iron, arsenic, lanthanum and uranium, and the anion fluoride exceeded the limit values of the Norwegian drinking water regulations', or limit values or guideline values from other countries. A single sample provides a snapshot of the water quality at the time of sampling. Analysis of groundwater taken at five different times throughout the year, from February to November 2019, showed, except for manganese in a sedimentary well, no significant seasonal variation in the concentration of the elements with exceedances. This indicates that annual control of water quality may be sufficient. However, the duration of one year for this study is short, and conditions may change over a longer time period.

A significant positive correlation was found between precipitation and uranium concentrations in groundwater from a bedrock aquifer. This means that increased precipitation could contribute to an increased concentration of uranium in the drinking water. However, to verify how groundwater quality can change on climate change it is important to perform a more extensive assessment. It is imperative to have access to measured meteorological data from the precipitation fields as well as a perception of the groundwater flow path and the travel time from the site of infiltration to the sampling point in the aquifer. The level of groundwater and the development of uranium concentrations in well water should be monitored regularly to achieve reliable data that include both within and between year variations.

The results from this work showed that marine salts and local conditions can be decisive for fluoride concentration in groundwater from bedrock aquifers, where the highest concentration of fluoride (5.3 mg/L) was measured in coastal areas. A significant positive correlation between fluoride and sodium ( $p = 0.04$ ) in groundwater from bedrock indicates that the concentration of fluoride in groundwater in near-coastal areas is more affected by sea salts in precipitation than by bedrock geology.

Lanthanum is characterized as a toxic substance with possible negative health effects (NICNAC, 2014). Neither WHO, EU, USA nor Canada has set limit values for any of the REEs. However, Australia uses a health-based guideline value for lanthanum of 2 µg/L (NICNAC, 2014). In the present work, the findings of high levels of lanthanum in groundwater from

unconsolidated sediments raise the question of whether the current requirements for quality control of REE in Norwegian drinking water are sufficient. Data are needed on lanthanum concentrations in more of the Norwegian waterworks. To establish a health-based guidance value for lanthanum, more data on health effects of this element should be obtained.

### CRedit authorship contribution statement

Conceptualization, E.L.F.G. and V.L.; Methodology, E.L.F.G., and V.L.; Chemical analysis, E.L.F.G., technical staff, and B.J.; Statistical analysis, B.J.; Writing – Original Draft Preparation, E.L.F.G.; Writing – Review and Editing, E.L.F.G., B.J., I.-L.S., M.H., and V.L.; Project administration: E.L.F.G. and V.L.; Funding Acquisition: E.L.F.G. and V.L.

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### Data availability

Data will be made available on request.

### Declaration of competing interest

The authors declare no conflicts of interest.

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