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Lithium, boron, cesium and other potentially toxic metals in Swedish well water

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IMM RAPPORT:

LITHIUM, BORON, CESIUM AND OTHER POTENTIALLY TOXIC METALS IN SWEDISH WELL WATER

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PREFACE

There are no reported data on concentrations of lithium and stable cesium (not radioactive) in Swedish well water, and limited data on boron concentrations. The main aim of this study was to fill those gaps in knowledge. To that end we measured those elements in about 600 samples of well water, most of which were collected 10 years ago by the Swedish Geological Survey (SGU) for their publication: Natural radioactivity, uranium and other metals in drinking water (Naturlig radioaktivitet, uran och andra metaller i dricksvatten; Ek et al. 2007). The data have also been published as SSI rapport 2008:15 (Ek et al. 2008).

We measured the element concentrations by modern inductively coupled plasma mass spectrometry (ICP-MS), by which it is possible to obtain sensitive measures of a large number of elements simultaneously in the same sample. Thus, we also measured a number of elements not previously measured. For comparison with previously reported concentrations, we measured a number of other elements in the water samples which would provide a useful inter-laboratory comparison of elements in drinking water.

CONTENTS

1 SVENSK SAMMANFATTNING (SWEDISH SUMMARY)

Det saknas data på förekomst av litium och cesium (stabil icke-radioaktiv isotop) i dricksvatten från svenska brunnar, och mycket knapphändig information om förekomsten av bor. Vi har därför mätt koncentrationerna av dessa metaller i 610 vattenprover från brunnar i stor del av landet. Flertalet vattenprov insamlades för 10 år sedan av Sveriges Geologiska Undersökning (SGU) för publikationen ”Naturlig radioaktivitet, uran och andra metaller i dricksvatten” (Ek et al. 2007, Ek et al. 2008). Vi analyserade ytterligare ett 30-tal prover från fr a Utö (i Stockholms södra skärgård), Öland och Kalmar. Förutom litium, bor och cesium analyserades ett antal metaller och andra ämnen som tidigare inte analyserats i proverna: beryllium, selen, bromid, rubidium, antimon and vismut. För att kontrollera provernas hållbarhet under 10 års lagring jämförde vi koncentrationerna av ett antal element som analyserats tidigare (magnesium, kalcium, mangan, arsenik, kadmium, bly och uran), vilket visade mycket god överensstämmelse. Vi använde modern, mycket känslig analysmetodik (ICP-MS).

Resultaten visar generellt låga koncentrationer av de flesta toxiska ämnena, även om flertalet förekom i relativt höga halter i vissa områden. Ur hälsosynpunkt viktigaste fynden är att koncentrationen av arsenik överskred gränsvärdet på 10 µg/L i 6.1% av proverna. Förhöjda halter av litium förekom framför allt på Gotland (medelhalt 33 µg/L) med ett högt värde (175 µg/L) i Dalarna. Även bor förekom framför allt på Gotland; högsta värdet var 3,500 µg/L. Hälsoriskerna bör utredas vidare. Halterna av cesium var generellt låga (median 0.08 µg/L). Högsta koncentrationerna förekom i Bohuslän och Dalarna, med endast 2-4 µg/L.

Även förhöjda halter av mangan, uran, kadmium och bly kan utgöra hälsorisker, framför allt för små barn. I 11% av proverna var manganhalten över 200 µg/L och i 2,5% över 400 µg/L. Flera av brunnarna i Dalarna hade förhöjda halter av uran, bly och kadmium. Flertalet brunnar hade uranhalter över riktvärdet på 30 µg/L; 12 brunnar hade mer än 300 µg/L. Medelvärdet för bly var 3 µg/L och flera brunnar med kadmium över 0.5 µg/L. Eventuella hälsorisker borde utredas.

Sammantaget tyder resultaten på behov av utvidgad analys av brunnsvatten. Pågående sammanställning vid SGU av analysdata från brunnsarkivet kan ligga till grund för bedömning av behovet av ytterligare systematiska undersökningar.

2 INTRODUCTION

Lithium (Li) is an alkali metal present in rocks and soil at varying concentrations. In Sweden, lithium-rich pegmatite rocks are found in e.g. Utö, Orrvik, Långsele and Varuträsk (SGU 2009). There appear to be no published data on lithium concentrations in drinking water in these areas, or elsewhere in Sweden, and no drinking water standard. The few data available on water lithium concentrations world-wide indicate large variations. In Europe, the lithium concentrations in stream water vary from <0.005 to 350 µg/L (Salminen R. 2005), while two studies evaluating trace elements in bottled water from different countries reported lithium concentrations from 0.1 to 10,000 µg/L (Krachler and Shotyk 2009; Reimann 2010). Recently, we discovered highly elevated lithium concentrations in drinking water in the Andean part of Northern Argentina (8-1,000 µg/L). The water in this area also contained high concentrations of boron (up to 6,000 µg/L), cesium (Cs; stable isotope, up to 322 µg/L) and arsenic (As; up to 200 µg/L) (Concha et al. 2010).

Lithium is easily absorbed from the intestinal tract and most of the absorbed dose is excreted in urine with a half-life of 1-4 days (Davanzo et al. 2011). Available data on lithium toxicity comes from lithium treatment of mental disorders especially bipolar disease (daily dose usually 450-1,300 mg). Side effects of such medication include mainly thyroid and kidney disorders (Grandjean and Aubry 2009; Olsson et al. 2010; McKnight et al. 2012). Little is known about potential long-term toxicity of lithium exposure through drinking water, but our recent studies in Northern Argentina show that lithium exposure may impair thyroid function (Broberg et al. 2011; Harari et al. 2015a). Also, lithium crosses easily the placenta and lithium exposure through drinking water during pregnancy was found to be associated with shorter birth length (Harari et al. 2012; Harari et al. 2015b). Also, several ecological studies have reported lower suicide rates in areas with only slightly elevated concentrations of lithium in drinking water (for review, see Vita et al. 2015). However, these studies have been criticized for lack of consideration of other potential influencing factors and the biological relevance.

Boron (B) occurs naturally in varying concentrations in the bedrock, soil and groundwater. Concentrations in European stream water have been found to vary between 0.1 and 3,000 µg/L (Salminen R. 2005), while concentrations in well water and bottled water ranged from <2 to 120,000 µg/L (Reimann 2010). We recently reported elevated drinking water concentrations of boron, range 377-11,000 µg/L, in the Andean part of northern Argentina (Igra et al. 2016). In Sweden, a previous study by the Swedish Geological Survey (SGU) reported boron concentrations in well water up to 3,000 µg/L in Gotland and Scania (Skåne), and concentrations between 100 and 500 µg/L in Östergötland and Västergötland (Ek et al. 2008). The Swedish drinking water standard (SLVFS 2001:30) for boron in public water is 1,000 µg/L, while the World Health Organization (WHO) has established a provisional guideline value of 2,400 µg/L (WHO 2009).

Experimental toxicity data for boron suggest that high doses may affect fertility, particularly in males, although human data are contradictory (WHO 2009). Exposure during pregnancy has been associated with intrauterine growth retardation, and fetal cardiovascular and rib malformations (rat, mouse and rabbit). A no observed adverse effect level

(NOAEL) of 9.6 mg B/kg bw/day was calculated. The European Food Safety Authority (EFSA) applied an uncertainty factor of 60 to the NOAEL value and established a tolerable upper intake level (UL) of 10 mg B/person/day for adults and 4 mg/day for children 4-6 years of age (EFSA 2006; EFSA 2013). A study on pregnant women in France (197 women in urban area with unknown boron exposure) showed that boron concentrations in placenta, collected at delivery, were associated with lower ALA-D (delta-aminolevulinic acid dehydratase) activity in cord blood (Huel et al. 2004). In our study in the Argentinean Andes we found that elevated boron exposure through drinking water was associated with lower size at birth (Igra et al. 2016).

Cesium (Cs) has four isotopes, of which ^{133}Cs is stable and the only naturally-occurring isotope. It occurs primarily in the mineral pollucite, the most important source of mining for production of cesium compounds used in the electronic industry and for energy production. Most cesium compounds are highly soluble in water. The chemical properties are similar to those of potassium and rubidium (Williams et al. 2004).

While there is plenty of data on the presence of radioactive cesium, especially in food, data on exposure to stable cesium is essentially lacking. Cesium is not included in standard measurement of drinking water and there are no standards. We recently discovered elevated cesium concentrations in drinking water (mainly groundwater) in the range 0.03-322 μg/L in the Andean part of Argentina samples (Concha et al. 2010). Cesium follows potassium in the food chain and behaves in a similar manner as potassium also in the human body (Williams et al. 2004). The average daily intake is estimated to 7-10 μg/L (Iyengar et al. 2004). We recently found that elevated cesium blood concentrations during pregnancy (range 2.5-711 μg/L) was associated with impaired thyroid function (Harari et al. 2015a).

In the present study, we measured the concentrations of lithium, boron and cesium in drinking water samples stored after the recent country-wide screening of concentrations of radon and arsenic in well water in Sweden, performed by the Swedish Geological Survey (Ek et al. 2008). Additionally, we measured the concentrations of a number of other potentially toxic elements that were not measured previously, namely beryllium (Be), selenium (Se), bromide (Br), rubidium (Rb), antimony (Sb) and bismuth (Bi). For the purpose of inter-laboratory comparison and evaluation of any effect of storage on the water element concentrations, we additionally measured some of the elements that had previously been measured: magnesium (Mg), calcium (Ca), manganese (Mn), arsenic (As), cadmium (Cd), lead (Pb) and uranium (U).

3 MATERIALS AND METHODS

3.1 SELECTION OF STUDY SITES AND WELLS

A large number of samples were originally collected from selected private wells in Sweden by the Swedish Geological Survey in cooperation with the Swedish Radiation Safety Authority with the aim to establish the concentrations of metals and radioactive elements (Ek et al. 2008). Before the sampling, drilled wells in areas with a potential for increased levels of these elements were picked out from the National Wells Archive at SGU. However, the sampling strategy also included wells from areas without any known metal or radioactive occurrence. Also, a few springs and dug wells were included. From the original more than 1100 water samples, 610 remaining water samples have been reanalyzed in the present project. The samples were taken between 2003 and 2006. There were more samples collected from wells drilled in the bedrock (550 samples), compared to samples from wells in unconsolidated material (56 samples) and natural springs (4 samples). Some of the water samples (n=96) had previously been analyzed for boron (Ek et al. 2008). The majority of the water samples represent untreated water (525 samples). If water treatment equipment was installed sampling was attempted both before and after the treatment. Thus, a minor part represents water that has passed treatment equipment (85 samples).

Geographical coordinates using the Swedish national grid (RT90) were recorded at each sampling place. The program Kartex v.5.41 (Normann and Bomark, 2010) was used to plot the geographical points in a map of Sweden. ArcGIS was used to plot the elemental distributions.

In addition, and due to the lack of samples from certain areas, we collected additional water samples from Utö (outside Stockholm; N=3), Öland (N=3) and Kalmar (N=25).

3.2 SAMPLE COLLECTION AND PREPARATION

The well-owners were contacted and SGU personnel made a visit for the sampling. The water was mainly collected from the tap after the water had been allowed to flow for more than 15 minutes, depending on the estimated water volume in the system. A check was made to ensure that a stable water temperature had been achieved in the water. At the laboratory the samples were acidified using concentrated nitric acid to a 1% concentration of HNO_3 and kept refrigerated (Ek et al. 2008). Some of the samples initially collected were later stored at room temperature and those were, therefore, excluded from the present study. In 2011, the samples were transported to the Institute of Environmental Medicine, Karolinska Institutet (KI), for analyses.

3.3 ANALYSES

Water samples were diluted 1:10 with 1% nitric acid (Scharlau, ppb-trace analyses grade, Scharlab S.L., Spain) and then analyzed for lithium, beryllium, boron, selenium, bromide, rubidium, antimony, cesium, bismuth, magnesium, calcium, manganese, arsenic, cadmium, lead and uranium, using inductively coupled plasma mass spectrometry (ICPMS; Agilent 7700x, Agilent Technologies, Tokyo, Japan) with collision/reaction cell in helium, hydrogen or no gas mode depending on the element (Table 1). The limit of detection (LOD) for all the analyzed elements and the percentage of samples below the LOD values are presented in Table 1.

For analytical quality control, reference materials were analyzed after about every 15 samples. We used the Standard Reference Material 1643e Trace Elements in Water (National Institute of Standards and Technology (NIST), Gaithersburg, USA). The obtained results agreed well with the reference values (Table 1).

As an additional quality control, we compared the analytical data on elements measured at both the Swedish Geological Survey and Karolinska Institutet. When comparing the obtained concentration of boron, magnesium, calcium, manganese, arsenic, cadmium, lead and uranium at KI with the previously obtained concentrations at SGU (n=610), we found in general excellent agreement between laboratories (Table 2). The Spearman correlation coefficients varied between 0.73 (cadmium) and 0.98 (uranium). The lower correlation (0.73) for the cadmium concentrations can mainly be explained by the lower LOD with the modern ICP-MS method used at KI. The discrepancy in the highest manganese concentration was probably due to precipitation.

3.4 STATISTICAL ANALYSIS

All the statistical analyses were performed using IBM ® SPSS ® Statistics 18.0 (SPSS, Chicago, IL, USA). Median, arithmetic mean, standard deviation (SD) and range were calculated for all the element concentrations. The concentrations are presented as box plots by county, ranked from highest to lowest concentrations by mean values. The median values are indicated inside the boxes, the inter-quartile range (IQR; 25th and 75th percentiles) as the bottom and top of the boxes, and the whiskers indicating 1.5 times the IQR.

4 RESULTS

The areas of water sampling and the number of samples at each sample place (in total 641 samples) are shown in Figure 1. The nationwide concentrations of all analyzed elements are presented in Table 3. In Figures 2-17, the concentrations are presented by county, ranked from highest to lowest mean concentration. However, it should be noted that the data are not representative for the whole county. Maps of the concentrations in drilled wells for the elements not previously published in Ek et al. 2008 are presented in Figures 18-27, showing regional differences for most elements. A comparison of concentrations in drilled wells and wells constructed in the soil layer is given in Table 4.

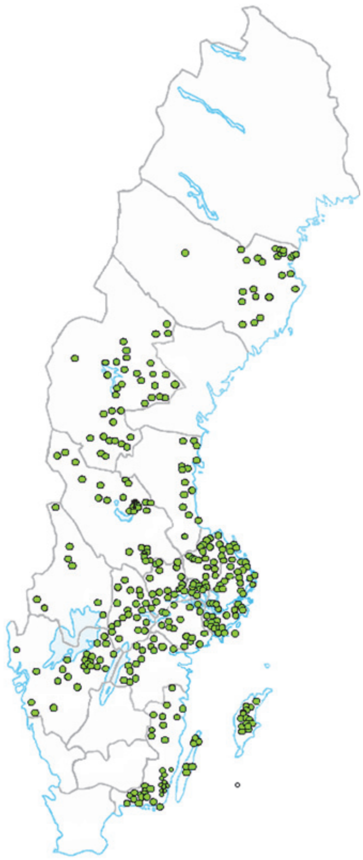

Figure 1. Sampling places and amount of samples used in this study at each Swedish county.

4.1 LITHIUM

Lithium concentrations in the water samples ranged 0.03-177 µg/L, with an overall average of 9.7 µg/L (median 6.7 µg/L) (Table 3). The highest lithium concentrations were found in the county of Gotland. There were also a few elevated values in Värmland and Dalarna (Figure 2 and 18).

4.2 BORON

Only 311 of the water samples had previously been analyzed for boron (Ek et al. 2008). Among the presently analyzed 641 samples, the boron concentrations ranged <LOD-3,522 µg/L. The highest concentrations were found in samples collected on the island of Gotland (Figure 4 and 20). Of all analyzed samples, 1.3% (N=8) had higher boron levels than the EU guideline value for boron in water of 1,000 µg/L (Table 3). Three samples (0.5%) exceeded the higher guideline value from WHO of 2,400 µg/L. All of these wells, but one (in Bohuslän 1096 µg/L) were on the island of Gotland .

4.3 CESIUM

The concentrations of cesium were generally very low with an overall median concentration of 0.08 µg/L. There were slightly higher concentrations in wells in Bohuslän (median 0.3 µg/L), and there was a single elevated value of almost 4 µg/L in Dalarna (Figure 14 and 26). In total only 3 wells exceeded 2 µg/L.

4.4 OTHER ELEMENTS

The concentrations of other elements such as beryllium, selenium, bromide, rubidium, antimony and bismuth, which had not been measured previously, are presented in Table 3. Of these elements, only selenium and antimony have drinking water guideline values or standards. Only one water sample, from Västerbotten, had an antimony concentration exceeding the EU guideline value of 5 µg/L (Figure 13 and 25). None of the samples had selenium concentrations exceeding the WHO/EU guideline values (Table 3, Figure 9 and 21).

The concentrations of beryllium were generally very low, with the majority of wells containing well below 0.5 µg/L, the median value bing 0.015 µg/L. The highest concentrations were found in Dalarna (Figure 3 and 19), the top value being 6.4 µg/L. In total of 9 wells (6 in Dalarna) exceeded 1 µg/L. Although beryllium has been classified as carcinogenic to humans (IARC 2012), there is no guideline value established for beryllium in drinking water (WHO 2001).

The highest concentrations of bromide were found in Gotland, Stockholm and Uppsala, with some wells exceeding 1 mg/L (Figure 10 and 22). There was also one single high value (6 mg/L) in Sörmland.

The concentrations of rubidium and bismuth did not vary much between areas, except for a few values; i.e. of rubidium in Uppsala and Stockholm (Figure 11 and 24) and of bismuth in Jämtland and Stockholm (Figure 16 and 27).

5 DISCUSSION

This study indicates generally low concentrations of most potentially toxic elements in well water in Sweden, although most elements occurred at clearly elevated concentrations in certain areas. We can confirm the previously reported elevated concentrations of arsenic and uranium in some areas (Ek et al. 2008). The arsenic concentrations exceeded the standard of 10 µg/L in 6.1% of the samples. We would also like to point out the elevated concentrations of boron and antimony in some water samples, some even exceeding the WHO and EU guideline values. This is the first report on well water concentrations of lithium, beryllium, antimony, cesium and bismuth in Sweden. Besides antimony, there are currently no guideline values for these elements. Still, most of them seemed to occur in low concentrations compared to the few other areas in the world studied (Concha et al. 2010; Shiraishi et al. 2004; Vaessen and Szteke 2000). To note, the present study did not cover all areas of Sweden (see Figure 1) and the original sampling of well water was focused on areas with expected elevated concentrations of radioactive substances and arsenic.

The median water lithium concentration in the present study was 6.7 µg/L. The highest concentrations were found on the island of Gotland with an average of 33 µg/L. One well in Dalarna had a lithium concentration of 175 µg/L, the highest in this study. Similar concentrations as those on Gotland have been reported for water in Santiago, Chile (range: 16-34 µg/L) (Harari et al. 2012), and about 100 µg/L in areas of Texas and Japan (Ohgami et al. 2009; Schrauzer and Shrestha 1990). Much higher lithium concentrations in drinking water (about 1,000 µg/L) have been observed in some villages in northern Argentinean Andes (Concha et al. 2010). In that area, the lithium exposure through drinking water has been associated with impaired thyroid and calcium homeostasis during pregnancy (Harari et al. 2015a, Harari et al. 2016) and lower size at birth (Harari et al. 2015b).

The obtained results for boron showed similar concentrations and geographic distribution as those 96 water samples previously reported in Ek et al. 2008. The highest concentrations of boron in Sweden, up to 3,500 µg/L, were found on Gotland. The high boron concentrations in the groundwater on Gotland have later been confirmed in a survey of 200 private wells in Gotland (Region Gotland, 2015). In total 72 wells (36 %) had a boron concentration above 1000 µg/L. Boron has been found to be associated with the clay fraction in clayey carbonate rocks and marlstones in similar sedimentary rocks in the adjacent area of western Estonia (Uppin and Karro 2013). An association of boron to marlstones has also been proposed in a German study of elevated boron (and fluoride) concentrations in the groundwater in the Muenster area (Queste et al. 2001). A common finding in these studies is elevated boron concentration in alkaline water with high sodium but low calcium concentrations, pointing to an influence of ion exchange processes in the clayey materials. Apart from Gotland, high boron concentrations have in Sweden mainly been found in the sedimentary bedrock in Skåne (Ek et al. 2008). The boron concentrations in the few water samples collected on the island of Öland were much lower than the concentrations found in Gotland. It can be noted that the geochemical atlas of Sweden shows similar boron contents in the till soil on Öland as on Gotland. (SGU 2014), indicating that boron could still be present in the groundwater. Thus, further studies in Öland are warranted.

Similar and even higher water boron concentrations as on Gotland have been reported in northern Argentina and Chile (Concha et al. 2010; Cortes et al. 2011; Harari et al. 2012). In the Argentinean Andes, we recently found that elevated boron exposure through drinking water during pregnancy, i.e. above 80 µg/L in blood serum, was associated with lower size of the babies at birth (Igra et al. 2016). An increase in serum boron of 100 µg/L in the third trimester corresponded to 0.9 cm shorter and 120 g lighter newborns ($p = 0.001$ and 0.021, respectively). The total range in the serum boron concentrations was 0.73-605 µg/L (median 133 µg/L), while the range in drinking water was 377-10,900 µg/L (median 6,000 µg/L). Below 80 µg/L in serum boron, the associations with birth weight and length were positive, indicating non-linear associations. It should be noted that the mode of action of boron at different exposure levels is not known. In one of the few previous human health studies, boron was found to be inversely associated with the ALA-D activity (a key enzyme for the production of hemoglobin) in umbilical cord blood (Huel et al. 2004). Otherwise, the limited toxicity data for boron are from experimental animal studies. Oral exposure to boric acid indicates that the male reproductive tract might be the target of toxicity (WHO 2008). Thus, more human studies are warranted in order to assess the dose-response for the adverse health effects and potential susceptibility factors.

The median well water concentration of cesium in the present study was 0.08 µg/L. The highest concentrations were found in Bohuslän and Dalarna, although still rather low (3.8 µg/L), and only 3 samples exceeded 2 µg/L. Similar concentrations as in the present study have been reported for water in Arica and Santiago, Chile (range: 0.5-3.8 µg/L) (Harari et al. 2012), while much higher cesium concentrations in drinking water (about 320 µg/L) have been observed in the Andean region of northern Argentina (Concha et al. 2010), in the same area where elevated lithium and boron are also prevalent. We have recently reported that elevated cesium concentrations in blood during pregnancy in that area were associated with impaired thyroid function (Harari et al. 2015a). To our knowledge, there are no other data on low-level cesium exposure and related health effects.

From a health point of view, the elevated arsenic concentrations are of greatest concern. Arsenic is a potent carcinogen, increasing the risk of cancer in skin, lungs and urinary bladder (IARC 2012), and there is increasing evidence of impaired child health and development (Ahmed et al. 2012; Gardner et al. 2013; Hamadani et al. 2011). Also, the cancer risk appears to increase following exposure very early in life (Steinmaus et al. 2014). Because of these health risks, water with arsenic concentrations above the drinking water standard of 10 µg/L should not be regularly used as drinking water.

Also the elevated manganese concentrations may be of concern, particularly for small children. About 11% and 2.5% of the samples had manganese concentrations above 200 and 400 µg/L, respectively. There is no health-based drinking water standard for manganese, the one included in table 3 of 300 µg/L is a technical guideline value set due to the risk of precipitation in water tubes and staining of clothes upon release of the precipitate. Also, U.S. EPA considers the risk of neurological effects low at concentrations below 300 µg/L (www.wpa.org). There is, however, increasing evidence for neurotoxicity of elevated manganese exposure at young ages (Roels et al. 2012). Because infant formula and first food based on rice and soy often contain elevated manganese levels (Ljung et al. 2011), it is recommended to prepare them using water with low manganese concentrations. The otherwise fairly strict regulation of manganese absorption is not fully developed until several months after birth, why elevated concentrations in drinking water and food may reach the brain at the critical period of brain development.

Other elements showing concentrations of potential health concern were lead, cadmium and uranium. Dalarna had the highest concentrations of all these metals. There was a mean concentration of almost 3 µg/L for lead and 120 µg/L for uranium (Figures 15 and 17). There were several well with cadmium concentrations exceeding 0.5 µg/L (Figure 12). Although all were below the current standard of 5 µg/L, the measured concentrations will contribute markedly to the total daily intake. Also, the Stockholm area showed a fairly high prevalence of elevated lead concentrations, although few were in this study above the drinking water standard of 10 µg/L. Still, they will contribute significantly to the total daily intake. Data from 15,000 wells, mostly in southern or middle Sweden, collected at the Geological Survey of Sweden show that approximately 4% of the analyzed drinking water samples from private wells contained concentrations of lead above 10 µg/L with a mean value of 3.2 µg/L (unpublished data). The European Food Safety Authority (EFSA) recently concluded that there is no dose level of lead under which there is no increased risk of effects on child development, and that a daily intake of 0.5 µg Pb/kg b.w. may be considered a low risk level (EFSA 2010). This level would correspond to about 2.5 µg/day for a child of 5 kg body weight; i.e. less than that provided by one liter of average well water from Dalarna.

As to the uranium concentrations, the overall mean concentration of all water samples exceeded the guideline value of 15 µg/L (www.slv.se), which is based on indicated kidney impairment (Svensson et al. 2009). Most of the water samples collected in Dalarna exceeded the guideline value; twelve wells had more than 300 µg/L. Also, the wells in Värmland, Uppsala and Västmanland had very high concentrations in relation to the guideline value. A recent meta-analysis of studies on cancer and kidney effects in uranium workers showed unexpectedly that the tumor risk was significantly lower for uranium workers than for control groups and no increased risk of nephrotoxicity (Stammler et al. 2016). There was no increased incidence or mortality due to renal cell carcinoma or due to acute kidney injury or chronic kidney disease. Another recent review of the health effects of uranium concluded, however, that there is strong toxicological evidence for renal and reproductive effects as well as DNA damage, but that there is limited epidemiologic evidence for these effects in people exposed to uranium in drinking water (Corlin et al. 2016). Two small epidemiological studies, one in Finland (Kurttio et al. 2006) and one in Årjäng in northern Sweden (Selden et al 2009) found no clear indications of adverse kidney effects. The Finnish study involved 193 individuals using drinking water with 0.03 - 1,500 µg/L. In the Swedish study, involving 301 individuals with <0.20-470 µg/L in drinking water, there was an indicated association between uranium in urine, but not water, and elevated urinary markers of kidney impairment; which, however, might be due to co-excretion. Thus, larger prospective studies are needed.

For evaluation of the observed concentrations in a geological context we refer to the previously publication (Ek et al. 2008). Beryllium, selenium, rubidium, antimony, cesium and bismuth, which had not been measured previously, occur in rather limited amounts in bedrock and soil and they do seldom form minerals but occur as substitutes to other elements or adsorbed to clay minerals and iron precipitates (SGU 2014). Bismuth and antimony may be connected to different kinds of mineralization.

Bromide originates mainly from sea water. Precipitations contain sea salt, but the bromide/chloride ratio (Figure 23) is often slightly higher than in sea water. Bromide can be further enriched relative to the concentration of chloride in clays deposited in sea water (Lundström and Olin 1986). Rubidium is another element often found in clay areas that earlier was covered by the sea (SGU 2014).

There was a good correlation between the element concentrations measured in the present study and those measured 10 years ago. There were some discrepancies in the concentrations of manganese between the two measurements, but only for a few samples from Stockholm (lower at SGU) and Bohuslän (higher at SGU), which may be due to precipitation in the sample tubes. The consistently lower concentrations of cadmium in the present study are probably related to the lower LOD of the ICP-MS instrumentation used.

6 CONCLUSIONS

Lithium and cesium concentrations in Swedish well water samples in the present study seem to be low. Boron and antimony occurred in higher concentrations than the guideline values in small percentages of the samples, while arsenic exceeded the standard in 6% of the water samples. For several other potentially toxic elements, we found elevated concentrations in certain areas. Because the presently studied water samples did not cover all areas of Sweden (see Figure 1) further screening is warranted. Also the areas with observed elevated concentrations of toxic metals should be followed-up with more structured sampling of wells.

There was a good correlation between measurements obtained at both KI and SGU, indicating that water samples can be stored for several years without change in metal concentrations.

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Table 1. Limits of detection and results (reference values and obtained values) of analyses of the Standard Reference Material 1643e Trace Elements in Water (NIST), used for quality control.

Elements	Analytical Mode	LOD KI	% samples <LOD	Reference value Mean±SD	Obtained value Mean±SD
Lithium (µg/L)	No gas	0.008	0.0	17.4 ± 1.7	18 ± 0.85
Beryllium (µg/L)	No gas	0.002	22	13.9 ± 0.17	14 ± 0.81
Boron (µg/L)	No gas	0.80	3.0	157.9 ± 3.9	161 ± 8.9
Magnesium (mg/L)	He	0.0003	0.0	8.0 ± 0.098	7.8 ± 0.17
Calcium (mg/L)	H2	0.004	0.0	32 ± 1.1	32 ± 0.75
Manganese (µg/L)	He	0.012	0.0	38.8 ± 0.45	38 ± 1.0
Arsenic (µg/L)	He	0.004	6.4	60.5 ± 0.72	61 ± 1.3
Selenium (µg/L)	H2	0.002	0.2	11.9 ± 0.14	12 ± 0.25
Bromide (µg/L)	He	0.90	0.0	N/A	9.0 ± 5.0
Rubidium (µg/L)	He	0.009	0.0	14.1 ± 0.18	14 ± 0.44
Cadmium (µg/L)	He	0.0009	21	6.6 ± 0.073	6.7 ± 0.16
Antimony (µg/L)	He	0.001	1.8	N/A	0.69 ± 0.71
Cesium (µg/L)	He	0.0006	4.6	N/A	0,0036 ± 0.0031
Lead (µg/L)	He	0.005	1.5	19.6 ± 0.21	19 ± 0.49
Bismuth (µg/L)	He	0.0007	13.1	14.1 ± 0.15	13 ± 0.31
Uranium (µg/L)	He	0.0001	0.7	N/A	0,0076 ± 0.0087

N/A: Not available.

Table 2. Inter-laboratory comparison of element concentrations in well water measured at Karolinska Institutet (KI) and the Swedish Geological Survey (SGU) (n=610). Presented data include limits of detection (LOD), Spearman correlation coefficients (r_s), median and range of boron (B), magnesium (Mg), calcium (Ca), manganese (Mn), arsenic (As), cadmium (Cd),lead (Pb) and uranium (U).

Elements	Institution	LOD	% <LOD	Median	Range	r_s
Boron (µg/L)	KI	0.80	3.0	22	0.87 - 3,521	0.97**
	SGU	0.50	3.0	28	1.6 - 3,060	
Magnesium (mg/L)	KI	0.0003	0.0	4.8	0.007 - 35	0.87**
	SGU	0.1	1.3	6.0	<LOD - 41	
Calcium (mg/L)	KI	0.004	0.0	39	0.014 - 250	0.89**
	SGU	0.1	0.3	46	<LOD - 282	
Manganese (µg/L)	KI	0.01	0.0	26	0.077 - 44,923	0.96**
	SGU	0.05	0.8	29	<LOD - 11,430	
Arsenic (µg/L)	KI	0.004	5.4	0.29	<LOD - 310	0.83**
	SGU	0.2	4.1	0.39	<LOD - 297	
Cadmium (µg/L)	KI	0.0009	21	0.016	0.0009 - 2.1	0.73**
	SGU	0.1	92	<LOD	<LOD - 6.5	
Lead (µg/L)	KI	0.005	1.5	0.41	0.046 - 62	0.94**
	SGU	0.1	18	0.39	<LOD - 42	
Uranium (µg/L)	KI	0.0001	0.7	4.9	0.0026 - 1,134	0.98**
	SGU	0.1	5.6	7.2	<LOD - 1,328	

**p<0.001

Table 3. Concentrations of multiple elements in collected well water samples (N=641) and the percentage of samples above existing guideline values for drinking water.

Elements in water	Median	Range	Standard Values WHO[a]/ EU (SLVFS)	% above WHO/EU standards
Lithium (µg/L)	6.7	0.03-177	-- / --	N/A
Beryllium (µg/L)	0.015	<LOD-6.4	-- / --	N/A
Boron (µg/L)	36	<LOD-3,522	2,400 / 1,000	0.5 / 0.8
Magnesium (mg/L)	4.7	<LOD-46	-- / --	N/A
Calcium (mg/L)	38	<LOD-250	-- / --	N/A
Manganese (µg/L)	24	0.07-44,922	-- / 50	38.5
Arsenic (µg/L)	0.28	<LOD-310	10 / 10	6.1
Selenium (µg/L)	0.075	0.014-2.6	40 / 10	0.0 / 0.0
Bromide (µg/L)	42	1.1-6,093	-- / --	N/A
Rubidium (µg/L)	2.1	0.11-27	-- / --	N/A
Cadmium (µg/L)	0.011	<LOD-2.1	3 / 5	0.0 / 0.0
Antimony (µg/L)	0.037	<LOD-8.6	20 / 5	0.0 / 0.3
Cesium (µg/L)	0.083	<LOD-3.8	-- / --	N/A
Lead (µg/L)	0.35	<LOD-62	10 / 10	1.5
Bismuth (µg/L)	0.0060	<LOD-0.35	-- / --	N/A
Uranium (µg/L)	4.7	0.0026-1,134	30 / 30	7.6 / 7.6

[a]WHO 2011. Guidelines for Drinking-water Quality. Fourth Edition

Table 4. Concentrations of multiple elements in water from shallow wells in soil (N=24, median and range are given) and for wells drilled in bedrock (N=360, median, range and percentiles 5^{th}, 25^{th}, 75^{th} and 95^{th}, are given). Results from water sampled before treatment facilities have been used if available. Mean values were calculated for each well if the same well was sampled several times.

	Shallow wells (N=24)		**Drilled wells (N=360)**			
Element	**Median**	**Range**	**Median**	**5^{th}-95^{th}**	**25^{th}-75^{th}**	**Range**
Lithium (µg/L)	2.8	0.30-18	6.8	0.60-3.1	3.1-13	<LOD-177
Beryllium (µg/L)	0.010	<LOD-0.43	0.014	<LOD-0.0040	0.0040-0.039	<LOD-5.1
Boron (µg/L)	14	<LOD-70	37	2.0-13	13-87	<LOD-3,522
Magnesium (mg/L)	4.5	0.26-17	4.5	0.86-2.8	2.8-7.7	0.014-35
Calcium (mg/L)	71	0.90-148	34	3.9-20	20-55	0.37-245
Manganese (µg/L)	6.9	0.10-449	26	0.5-5.0	5.0-102	0.10-44,923
Arsenic (µg/L)	0.23	<LOD-19	0.24	<LOD-0.080	0.080-0.81	<LOD-272
Selenium (µg/L)	0.16	0.010-0.62	0.060	0.010-0.030	0.030-0.14	<LOD-2.6
Bromide (µg/L)	34	6.0-368	40	8.0-20	20-90	1.0-6,093
Rubidium (µg/L)	1.3	0.20-15	2.2	0.40-1.3	1.3-3.7	0.10-25
Cadmium (µg/L)	0.023	0.0020-0.22	0.011	0.0020-0.0050	0.0050-0.027	0.0010-2.1
Antimony (µg/L)	0.084	0.013-0.22	0.032	0.0070-0.019	0.019-0.070	0.0020-8.6
Cesium (µg/L)	0.040	<LOD-0.97	0.090	0.010-0.030	0.030-0.23	<LOD-3.8
Lead (µg/L)	0.55	<LOD-2.0	0.33	<LOD-0.12	0.12-0.86	<LOD-62
Bismuth (µg/L)	0.0070	0.0010-0.039	0.0060	<LOD-0.0020	0.0020-0.011	<LOD-0.35
Uranium (µg/L)	4.8	0.20-32	3.5	0.10-0.90	0.90-9.6	<LOD-1,076

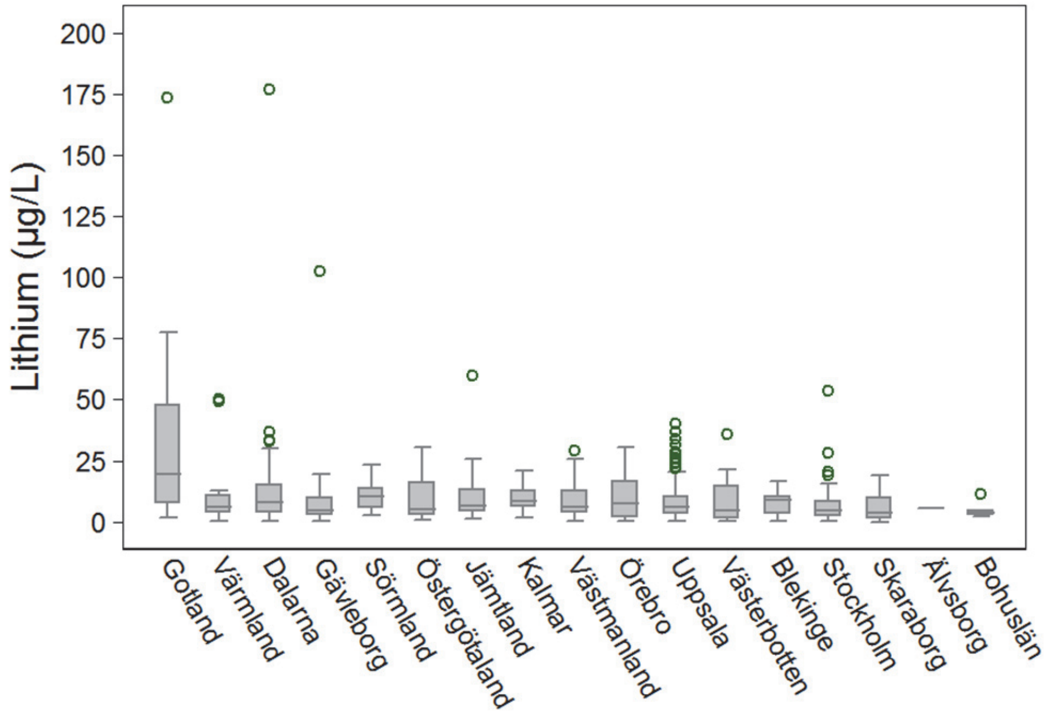

Figure 2. Lithium concentrations in Swedish well water samples, by county. Boxes represent the first and third quartiles (25th and 75th percentiles) and the band inside represents the median value. The whiskers show the lowest and highest values within 1.5 times the IQR. Dots represent outliers.

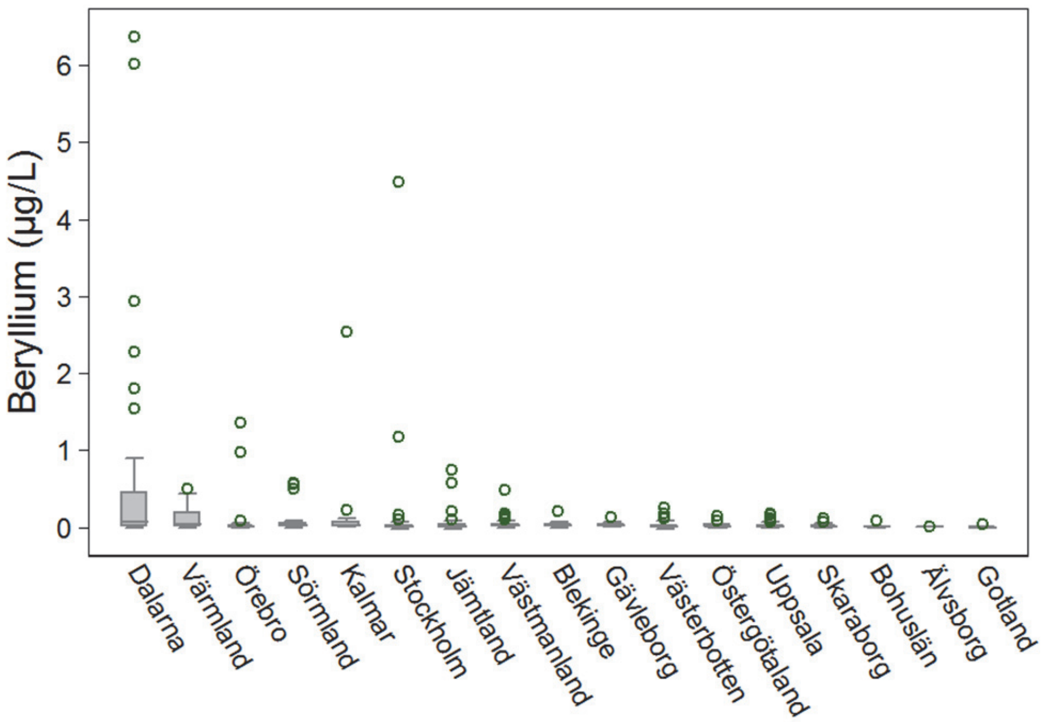

Figure 3. Beryllium concentrations in Swedish well water samples, by county.

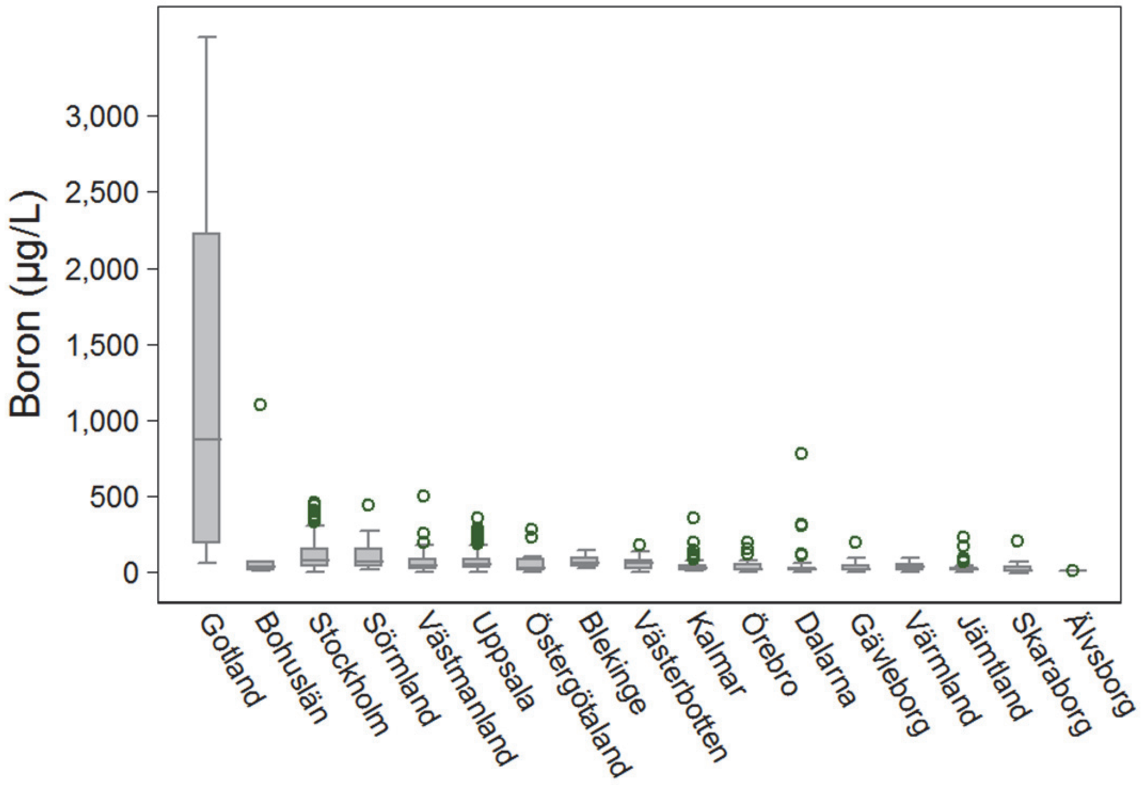

Figure 4. Boron concentrations in Swedish well water samples, by county.

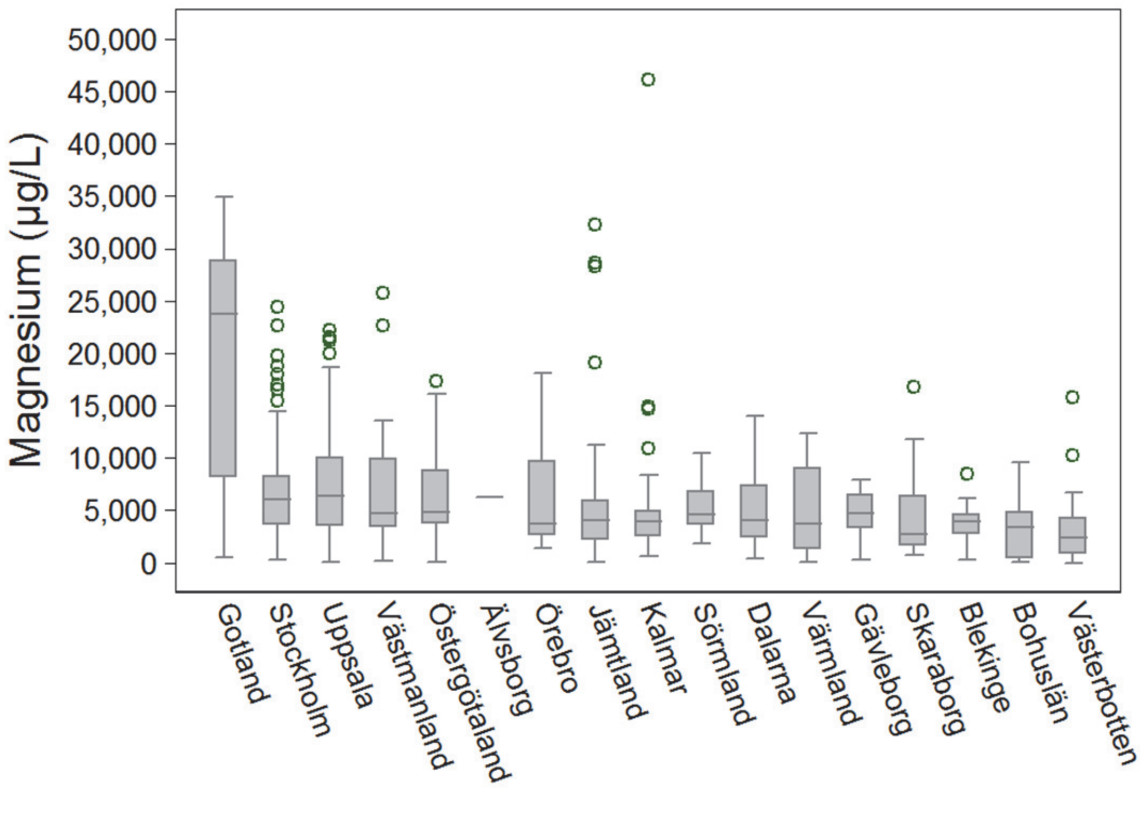

Figure 5. Magnesium concentrations in Swedish well water samples, by county.

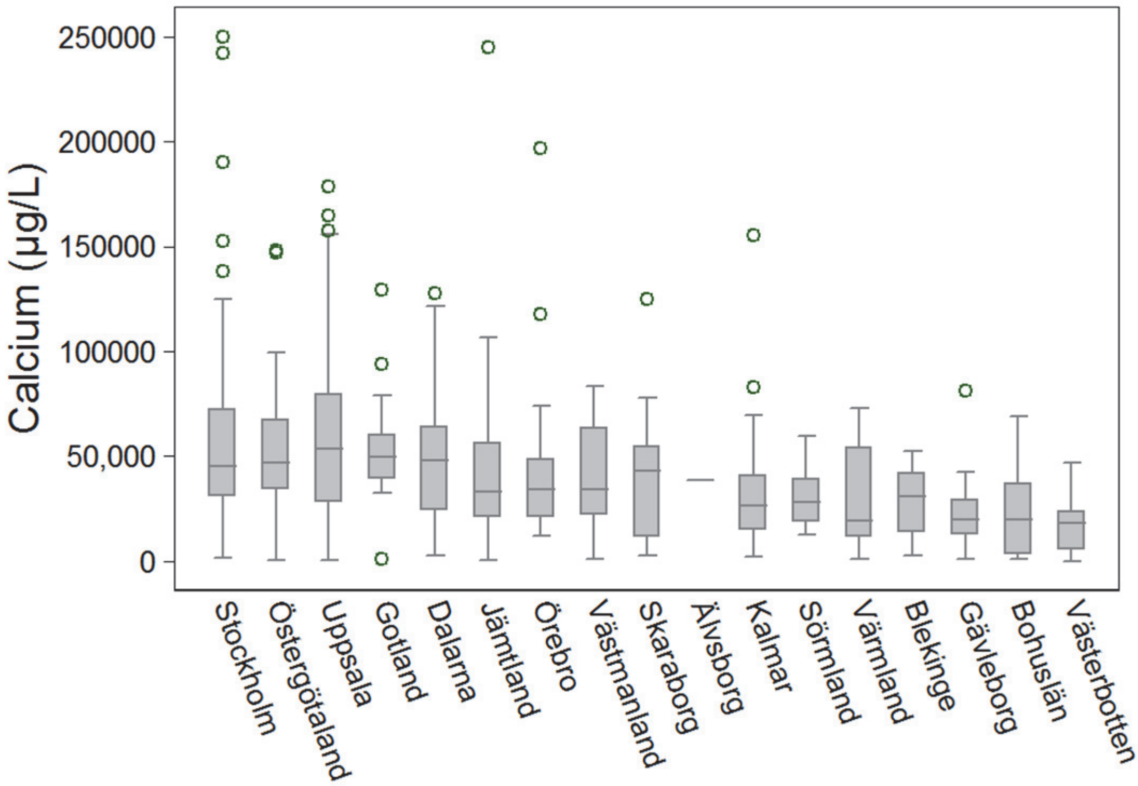

Figure 6. Calcium concentrations in Swedish well water samples, by county.

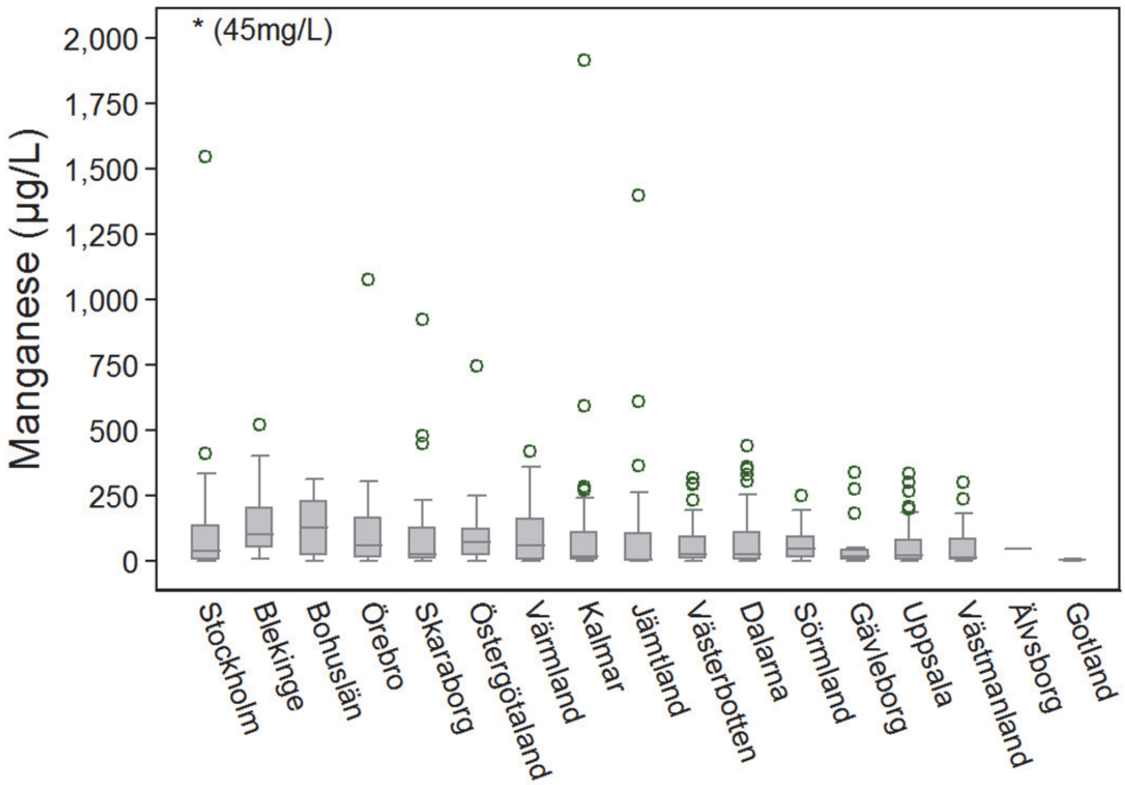

Figure 7. Manganese concentrations in Swedish well water samples, by county.

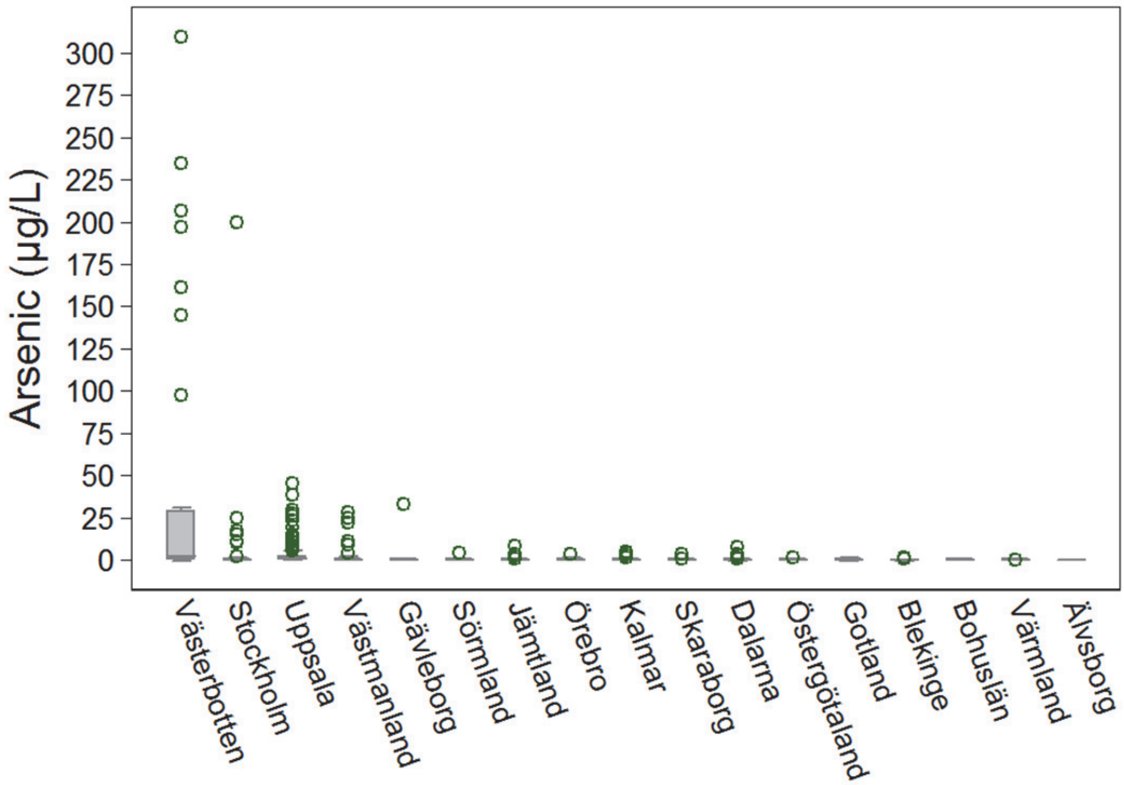

Figure 8. Arsenic concentrations in Swedish well water samples, by county.

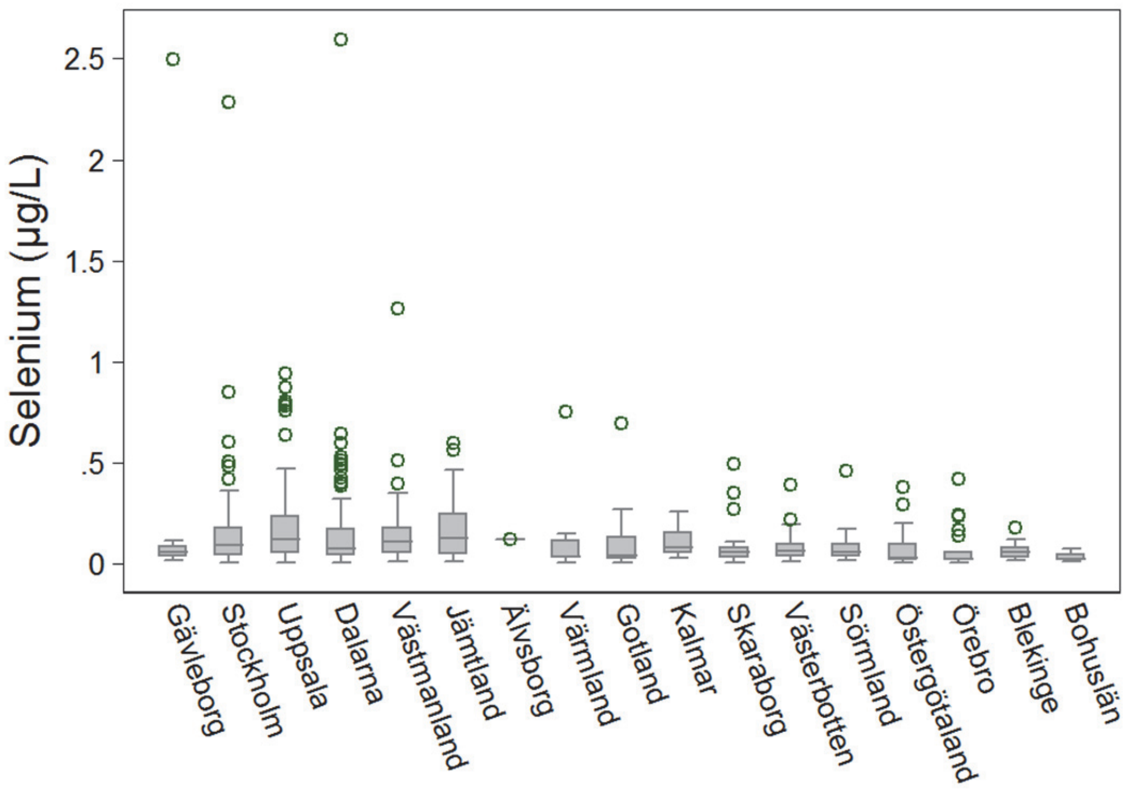

Figure 9. Selenium concentrations in Swedish well water samples, by county.

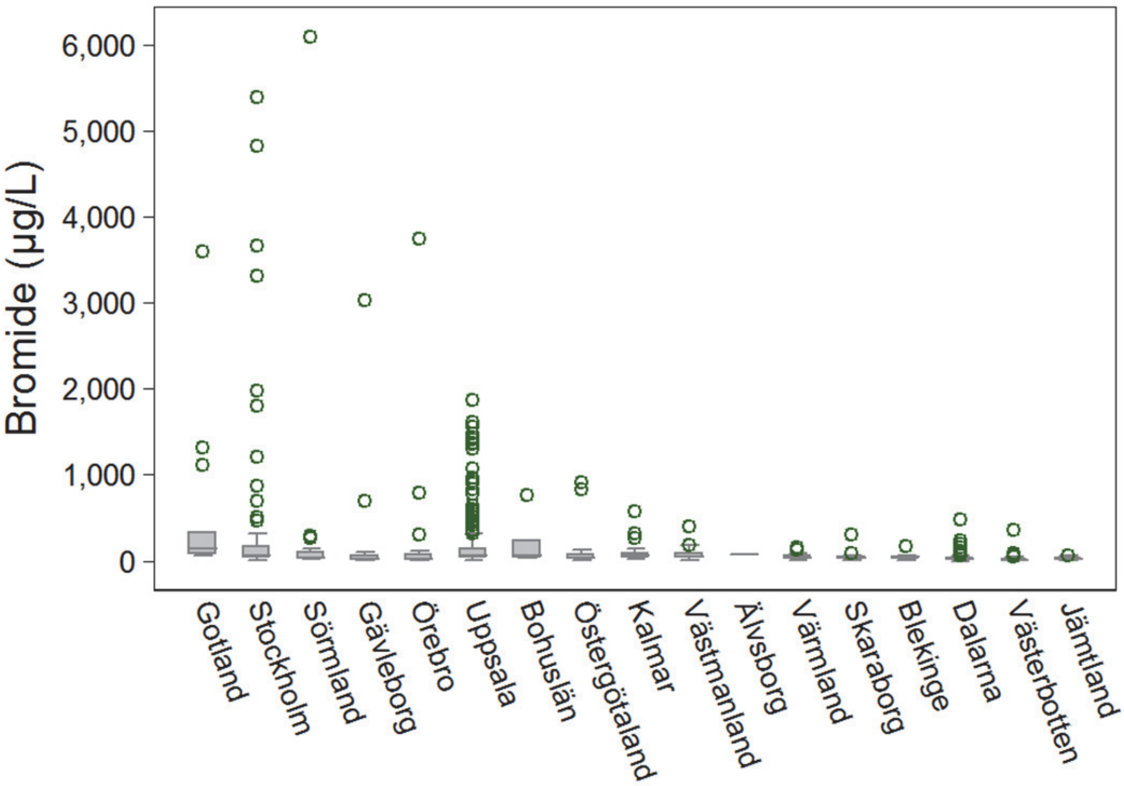

Figure 10. Bromide concentrations in Swedish well water samples, by county.

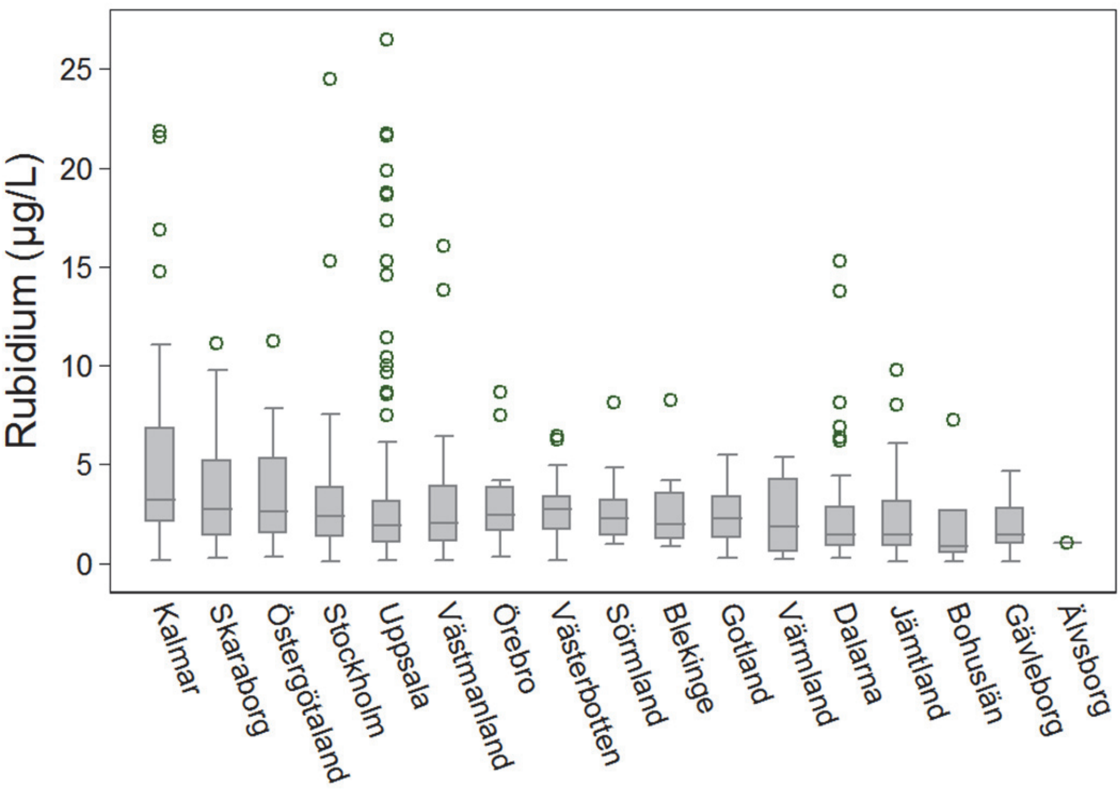

Figure 11. Rubidium concentrations in Swedish well water samples, by county.

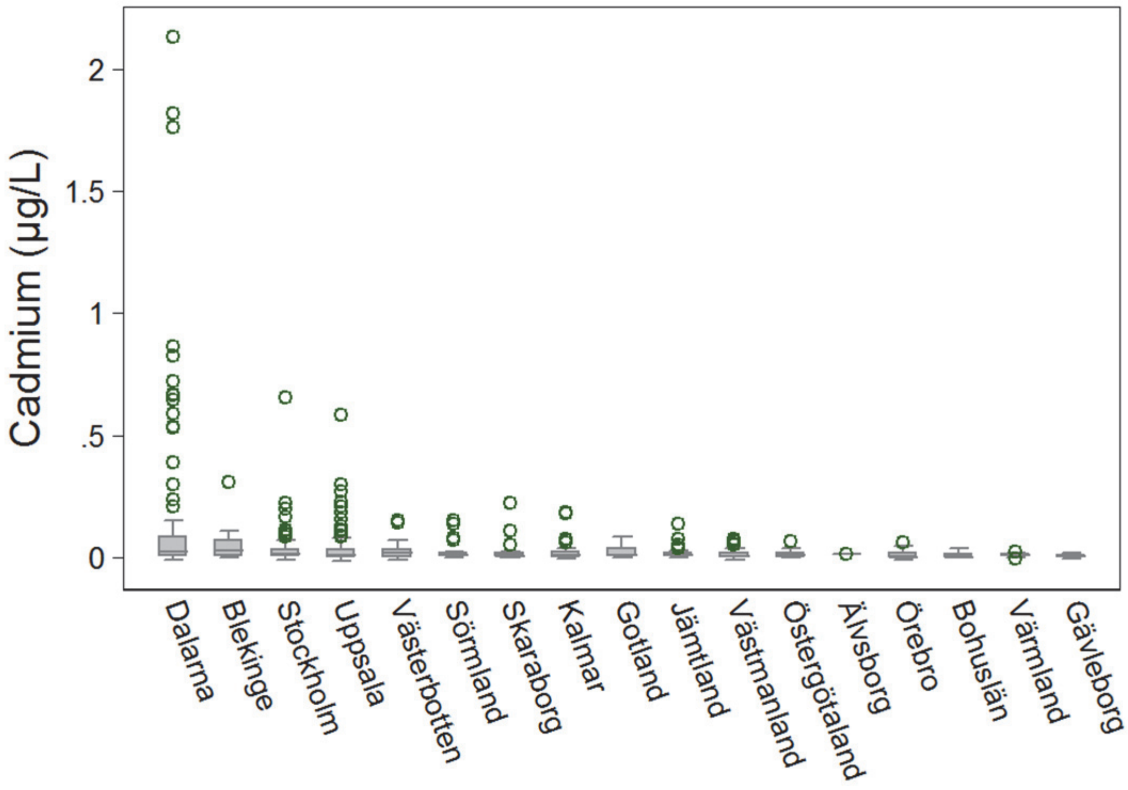

Figure 12. Cadmium concentrations in Swedish well water samples, by county.

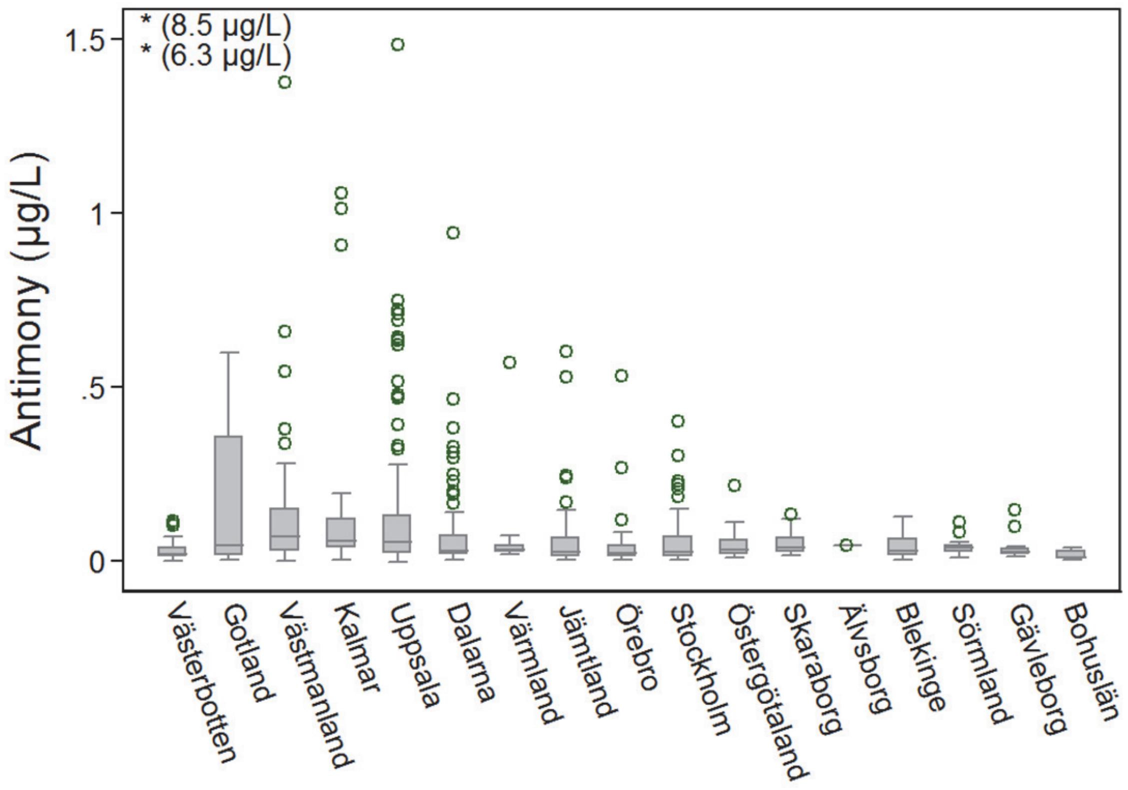

Figure 13. Antimony concentrations in Swedish well water samples, by county.

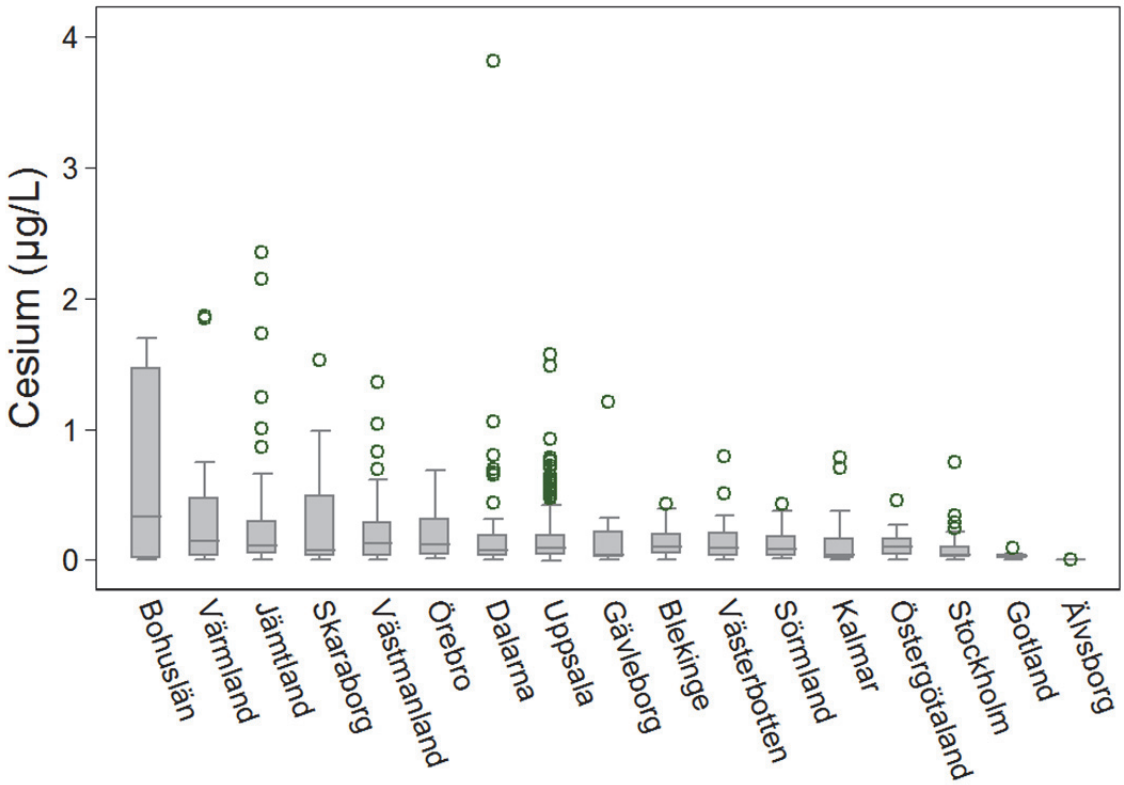

Figure 14. Cesium concentrations in Swedish well water samples, by county.

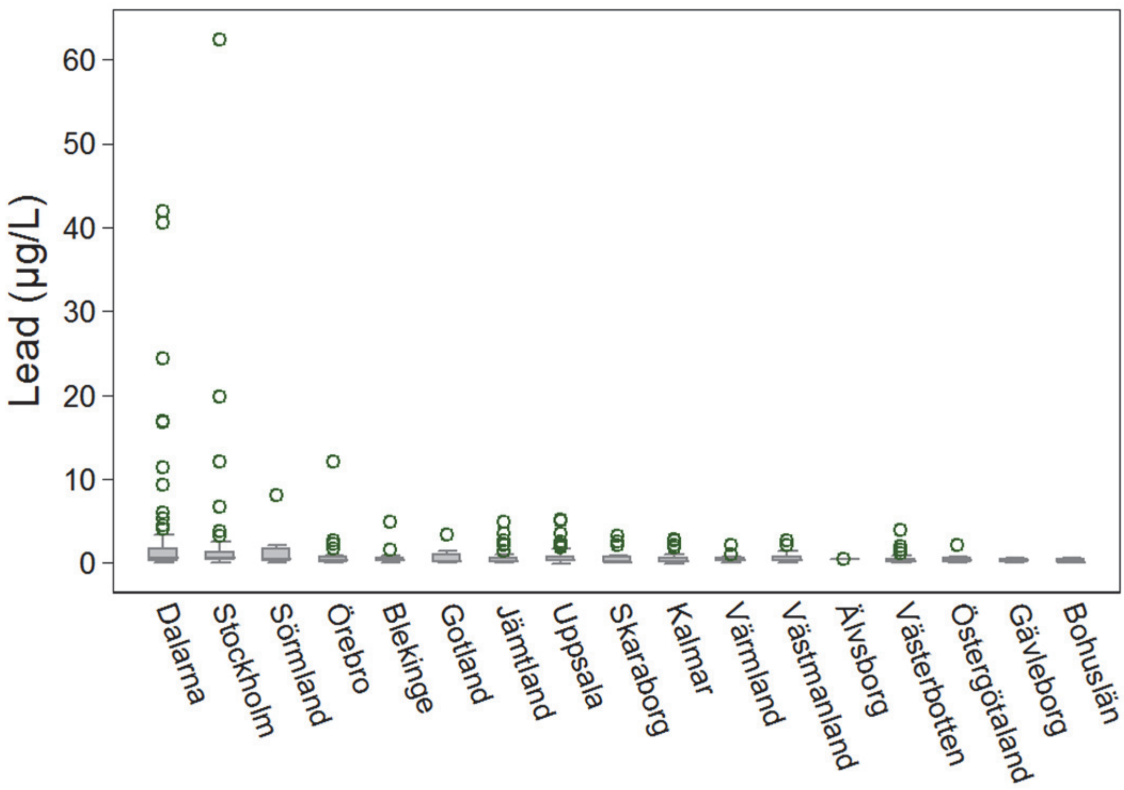

Figure 15. Lead concentrations in Swedish well water samples, by county.

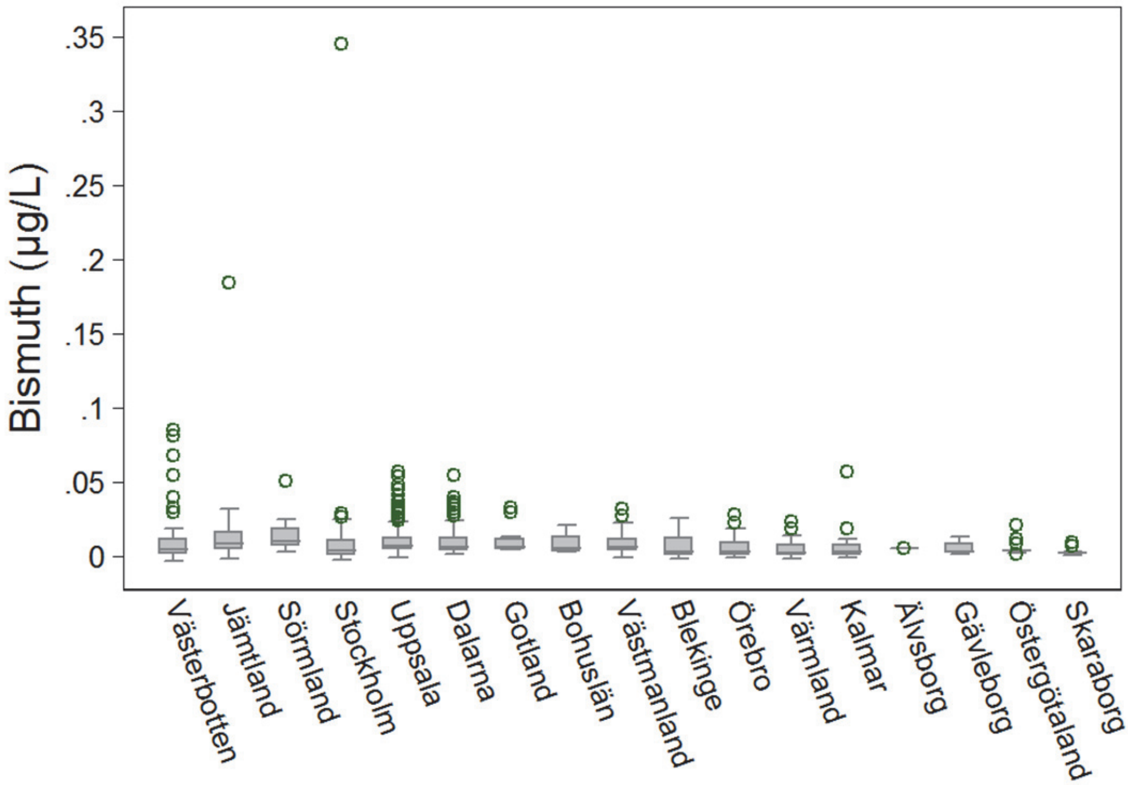

Figure 16. Bismuth concentrations in Swedish well water samples, by county.

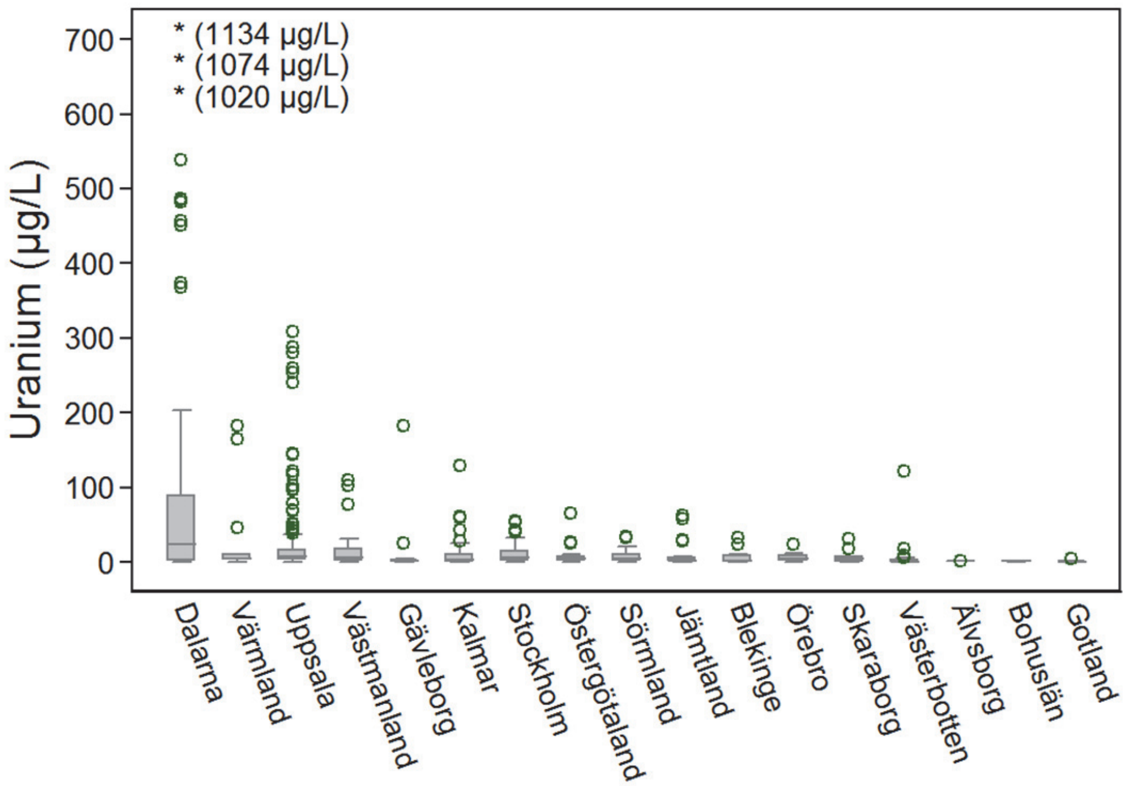

Figure 17. Uranium concentrations in Swedish well water samples, by county.

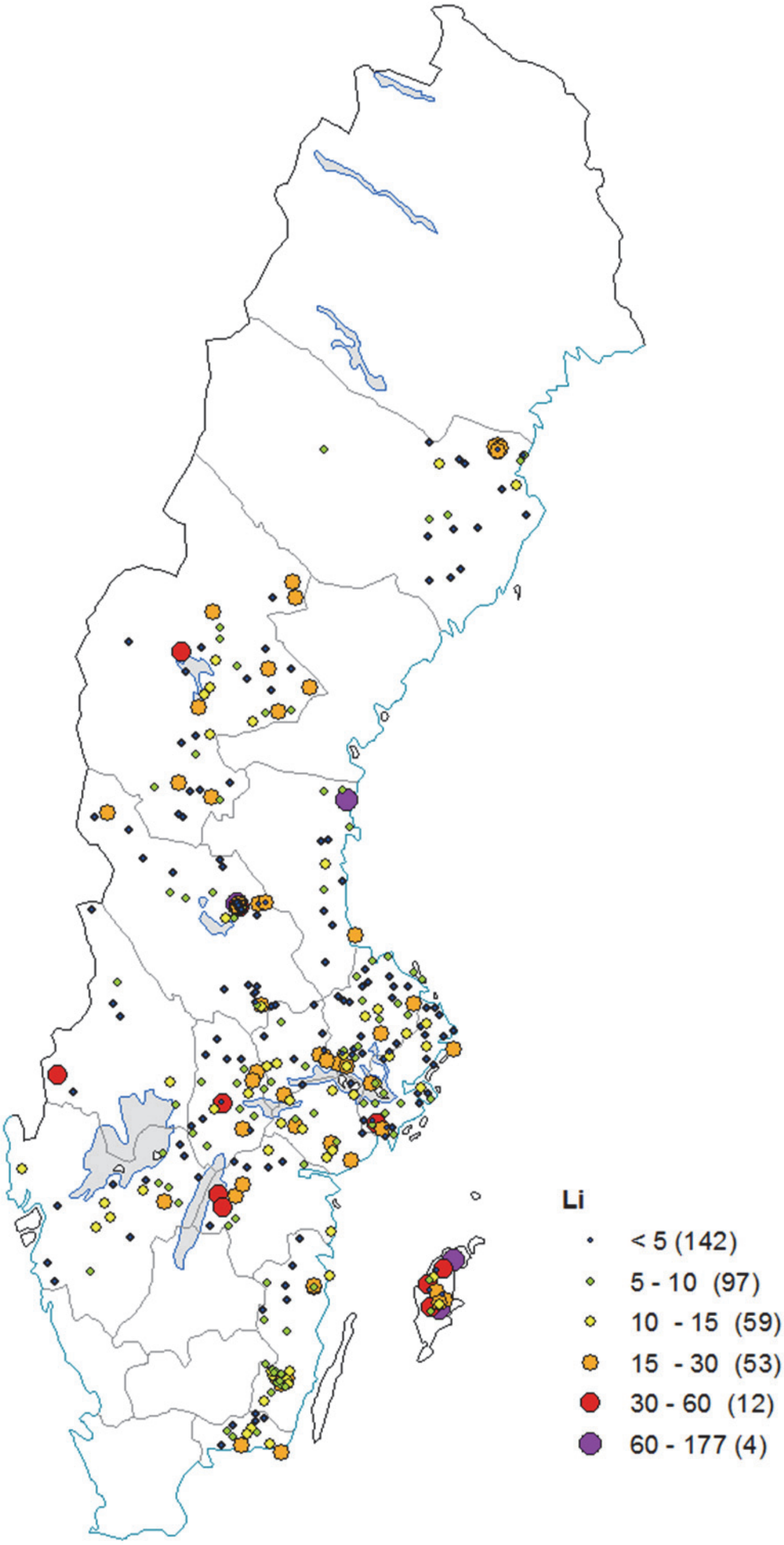

Figure 18. Lithium concentrations in Swedish drilled well water samples (μg/L).

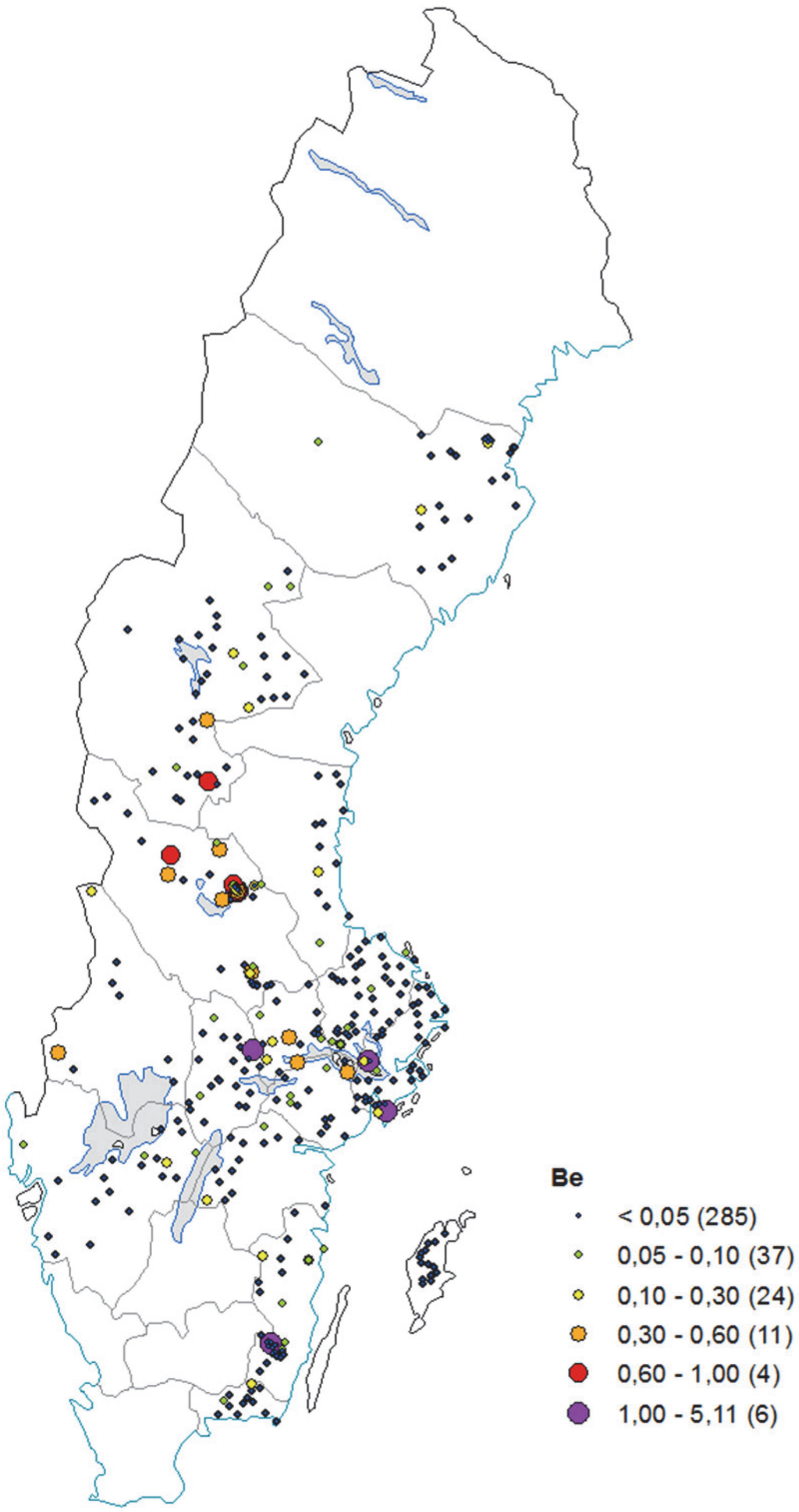

Figure 19. Beryllium concentrations in Swedish drilled well water samples (µg/L).

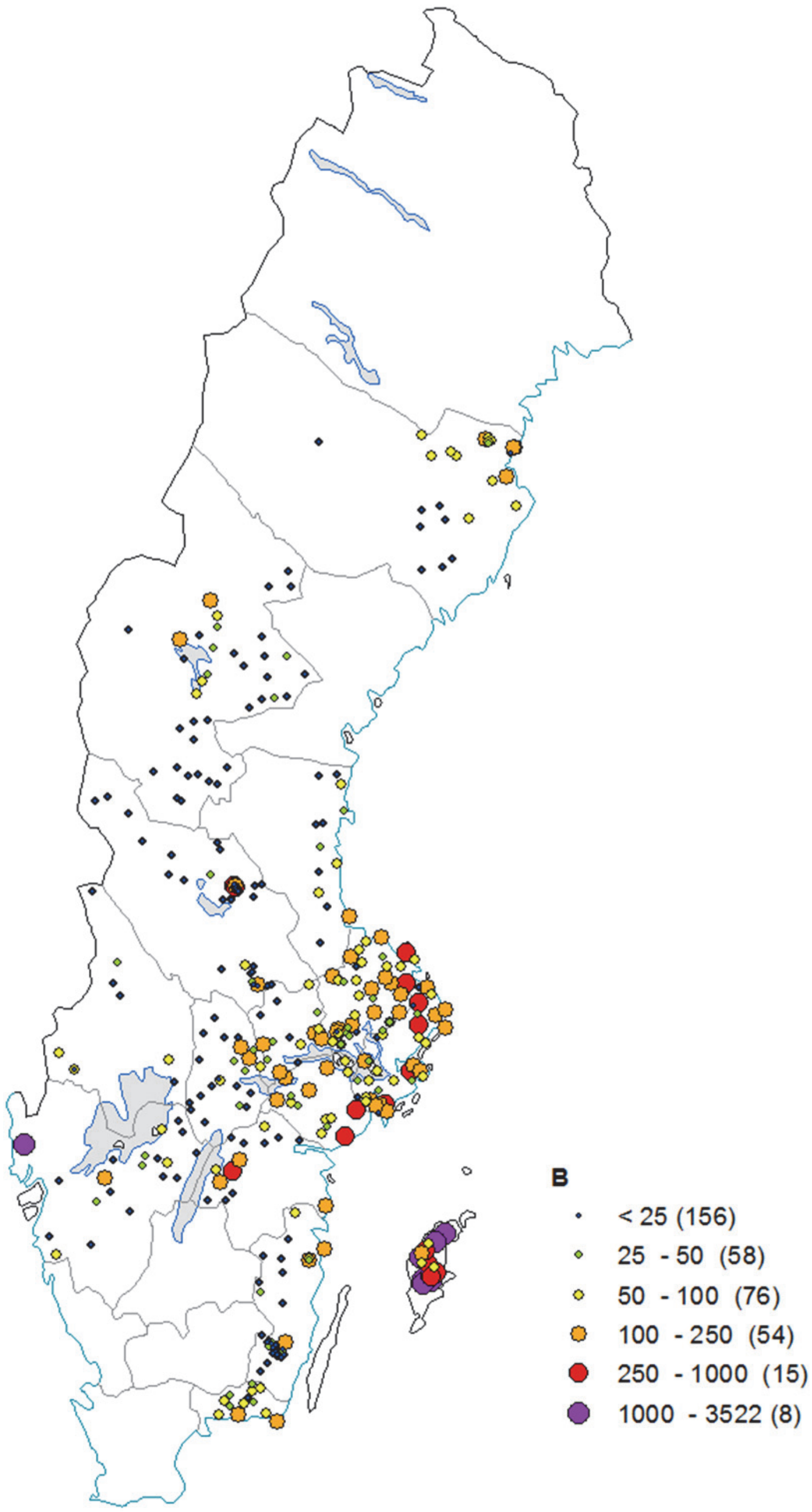

Figure 20. Boron concentrations in Swedish drilled well water samples (µg/L).

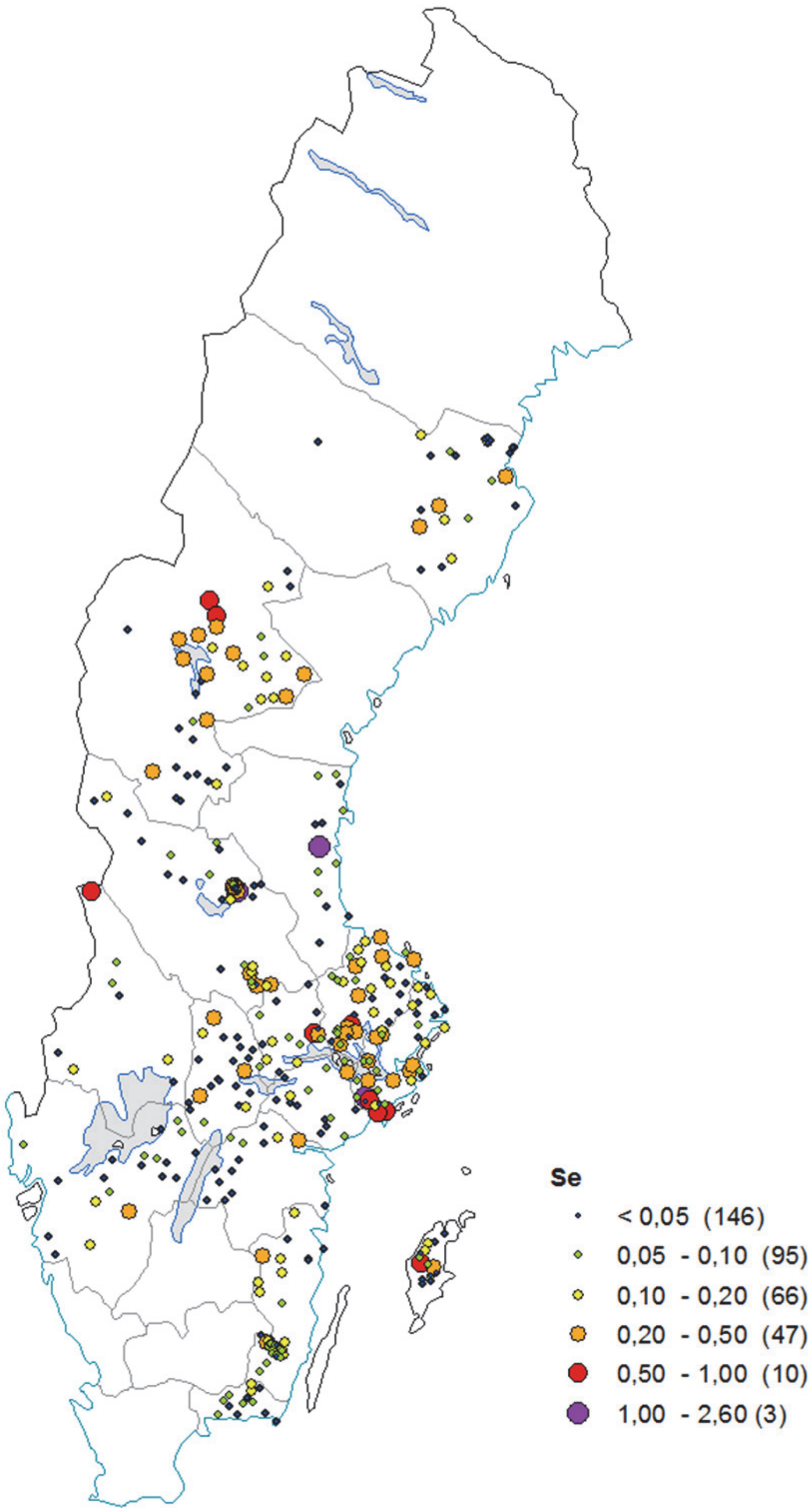

Figure 21. Selenium concentrations in Swedish drilled well water samples (µg/L).

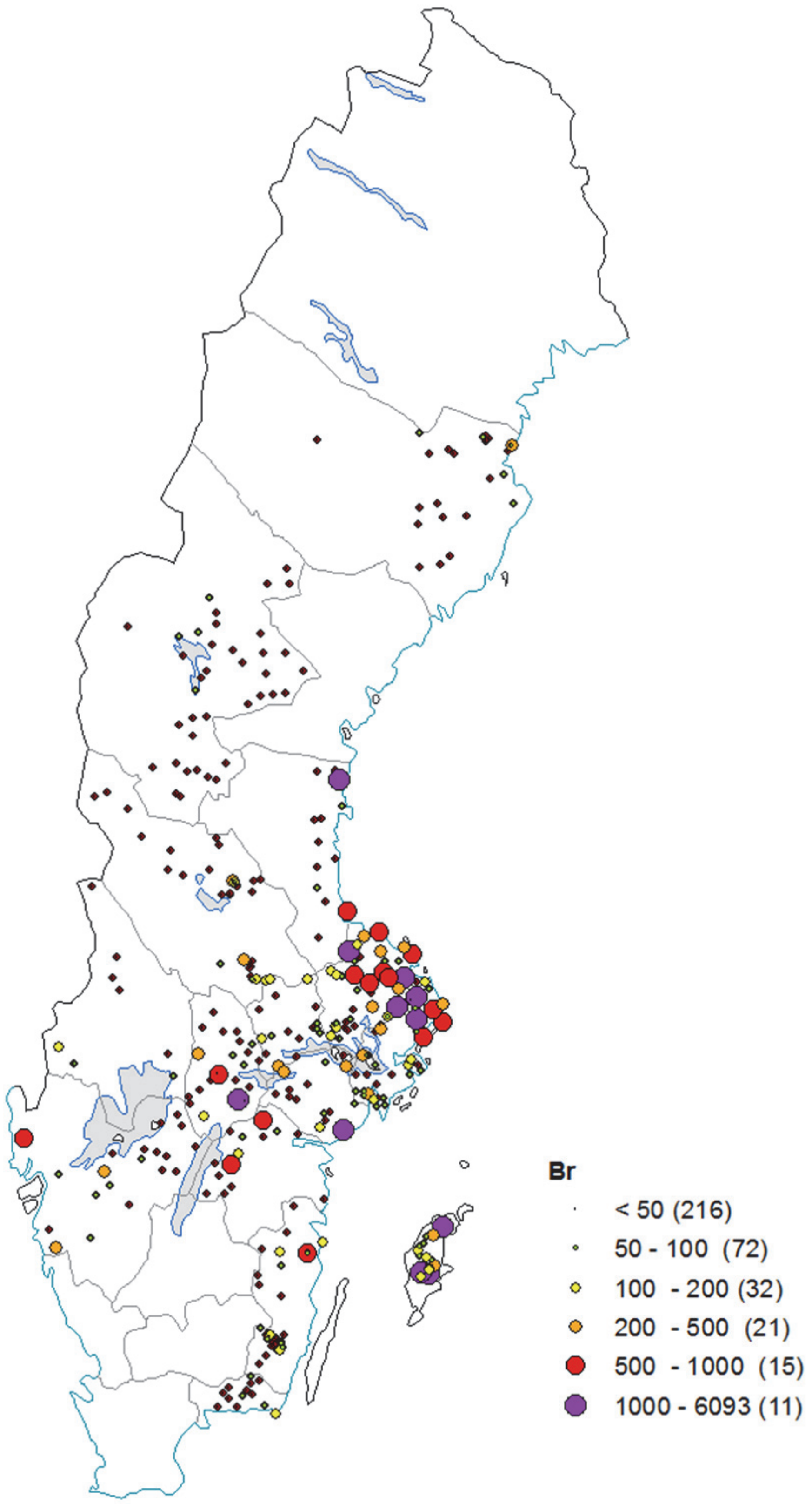

Figure 22. Bromide concentrations in Swedish drilled well water samples (µg/L).

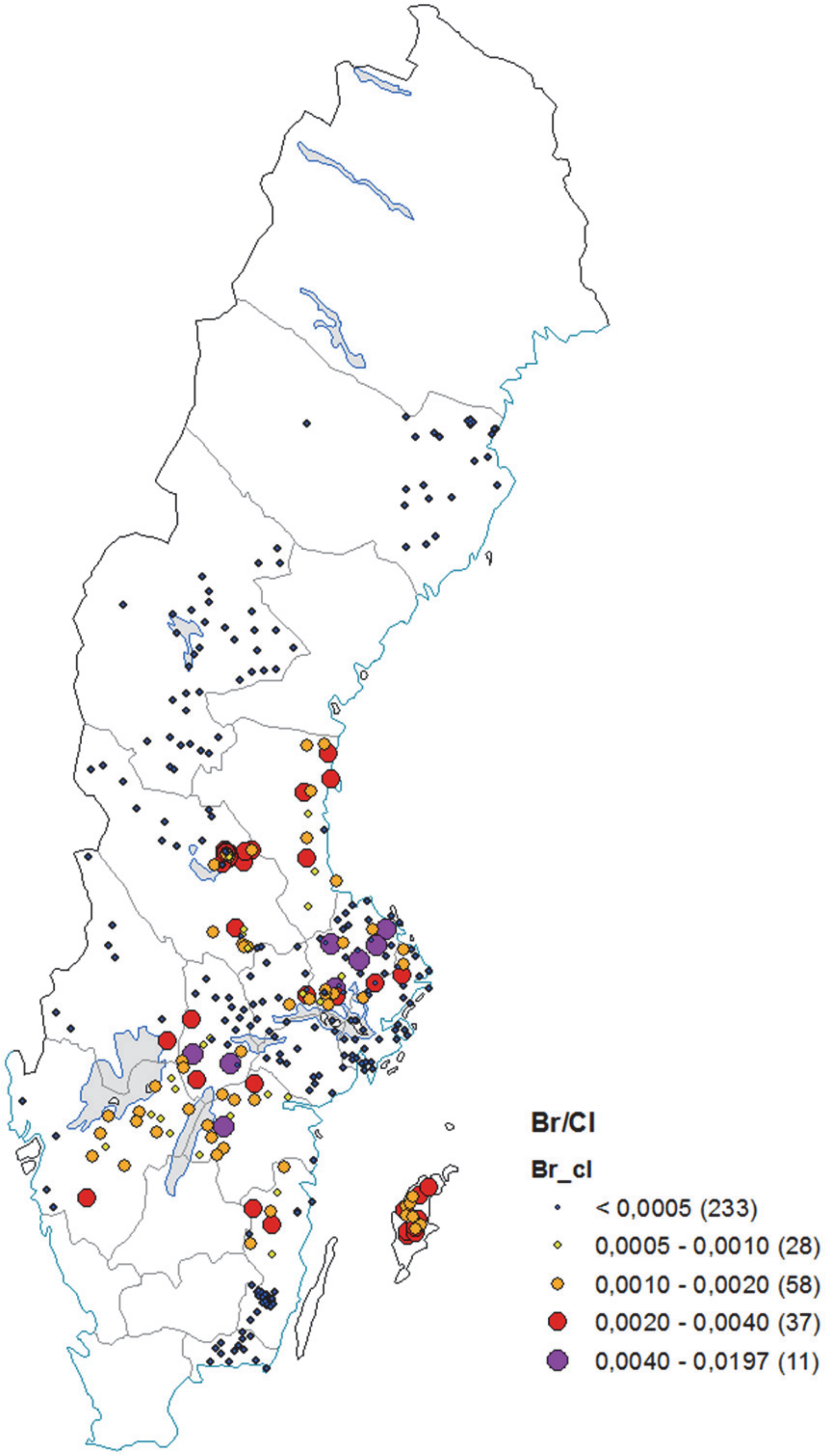

Figure 23. Bromide/Chloride ratio in Swedish drilled well water samples (calculated from mM/L).

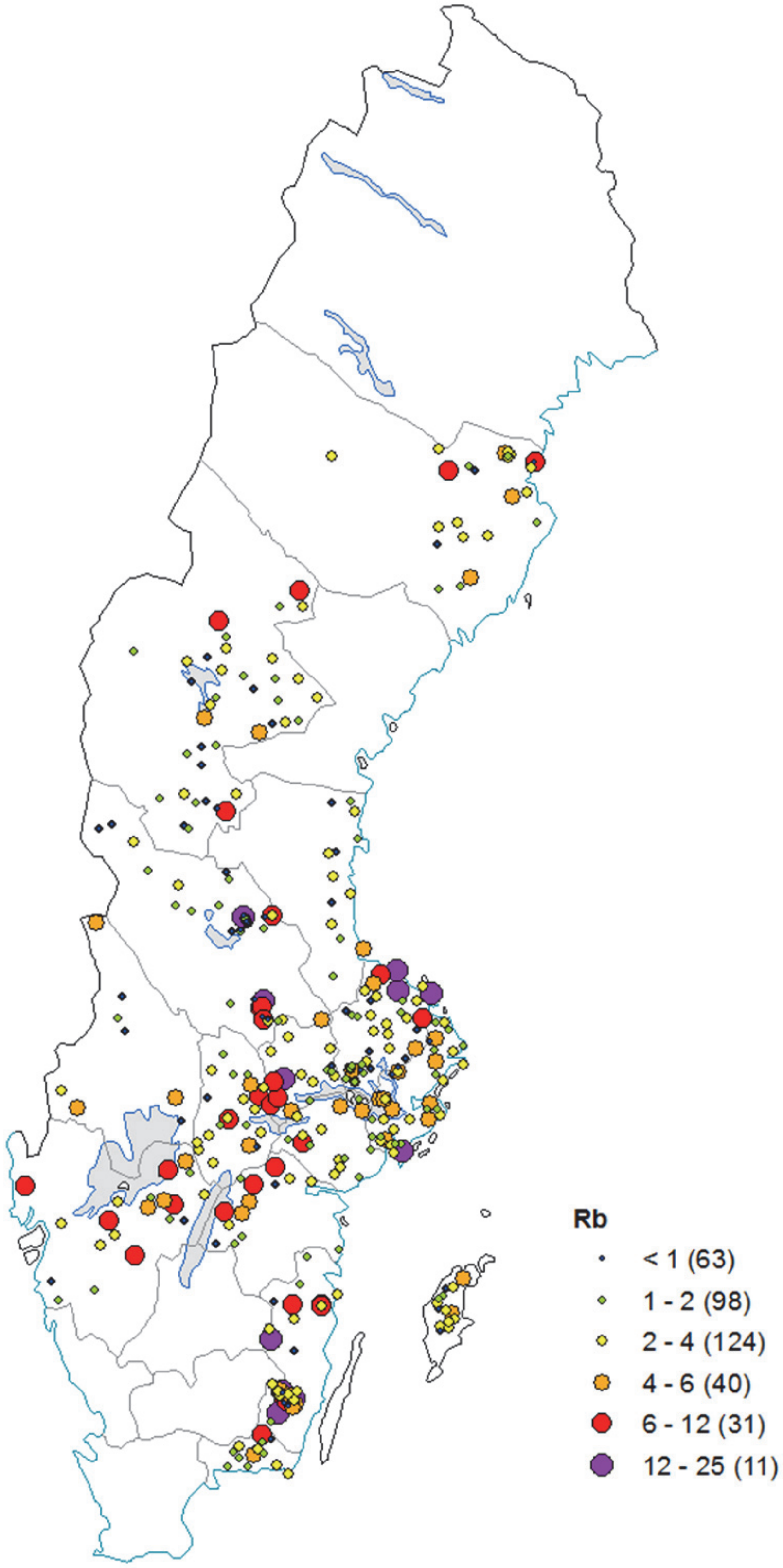

Figure 24. Rubidium concentrations in Swedish drilled well water samples (µg/L).

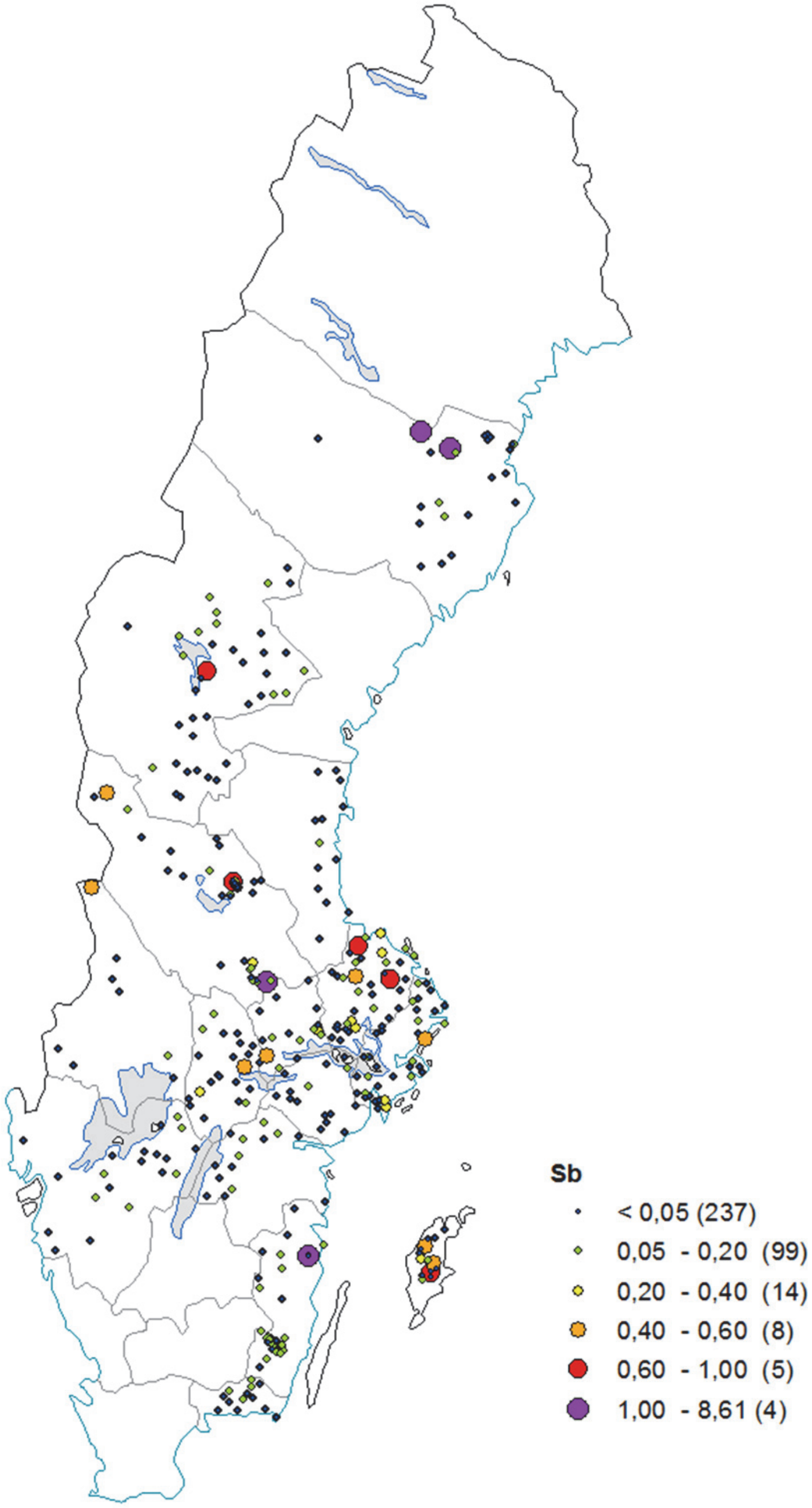

Figure 25. Antimony concentrations in Swedish drilled well water samples (µg/L).

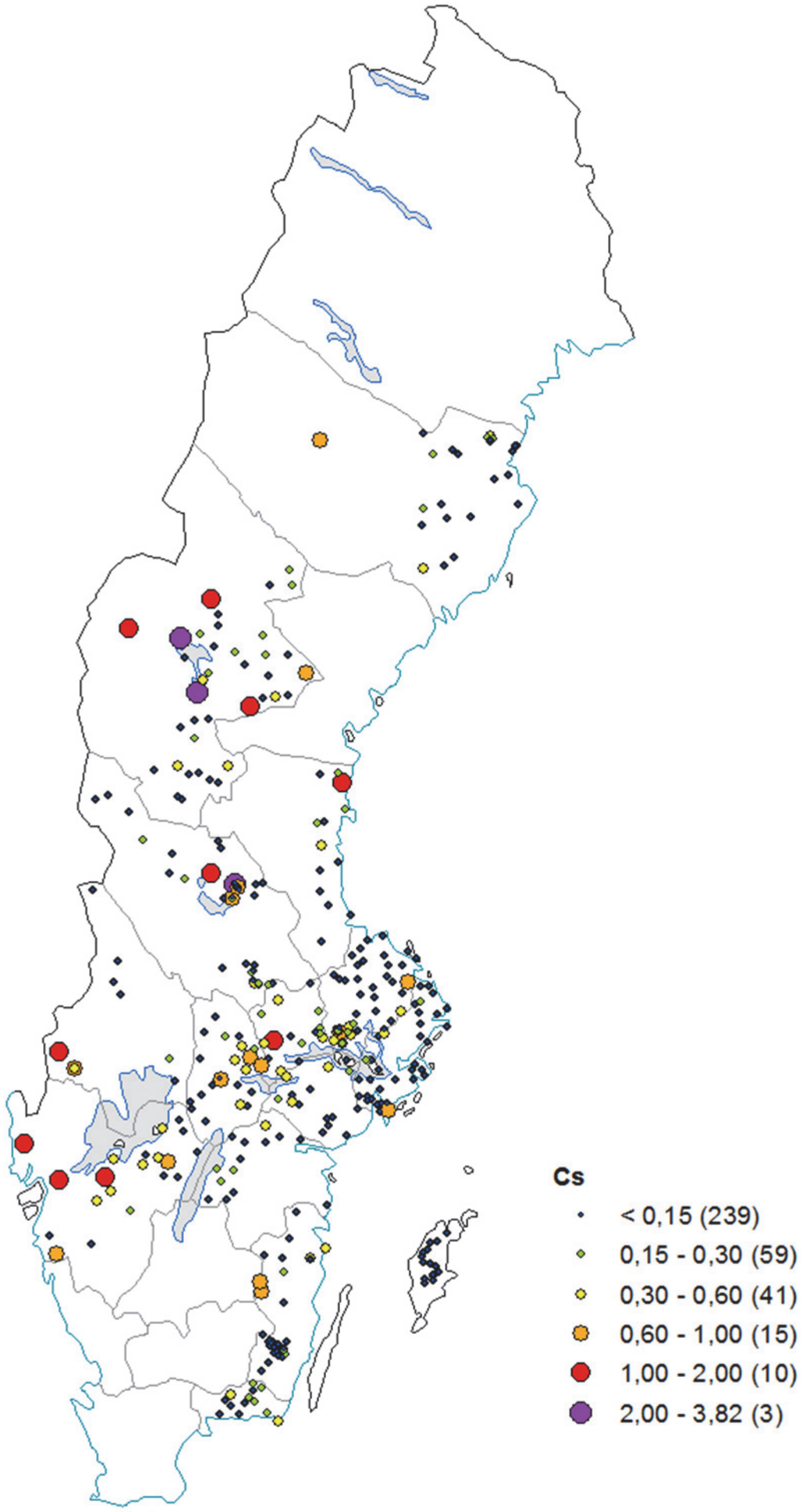

Figure 26. Cesium concentrations in Swedish drilled well water samples (μg/L).

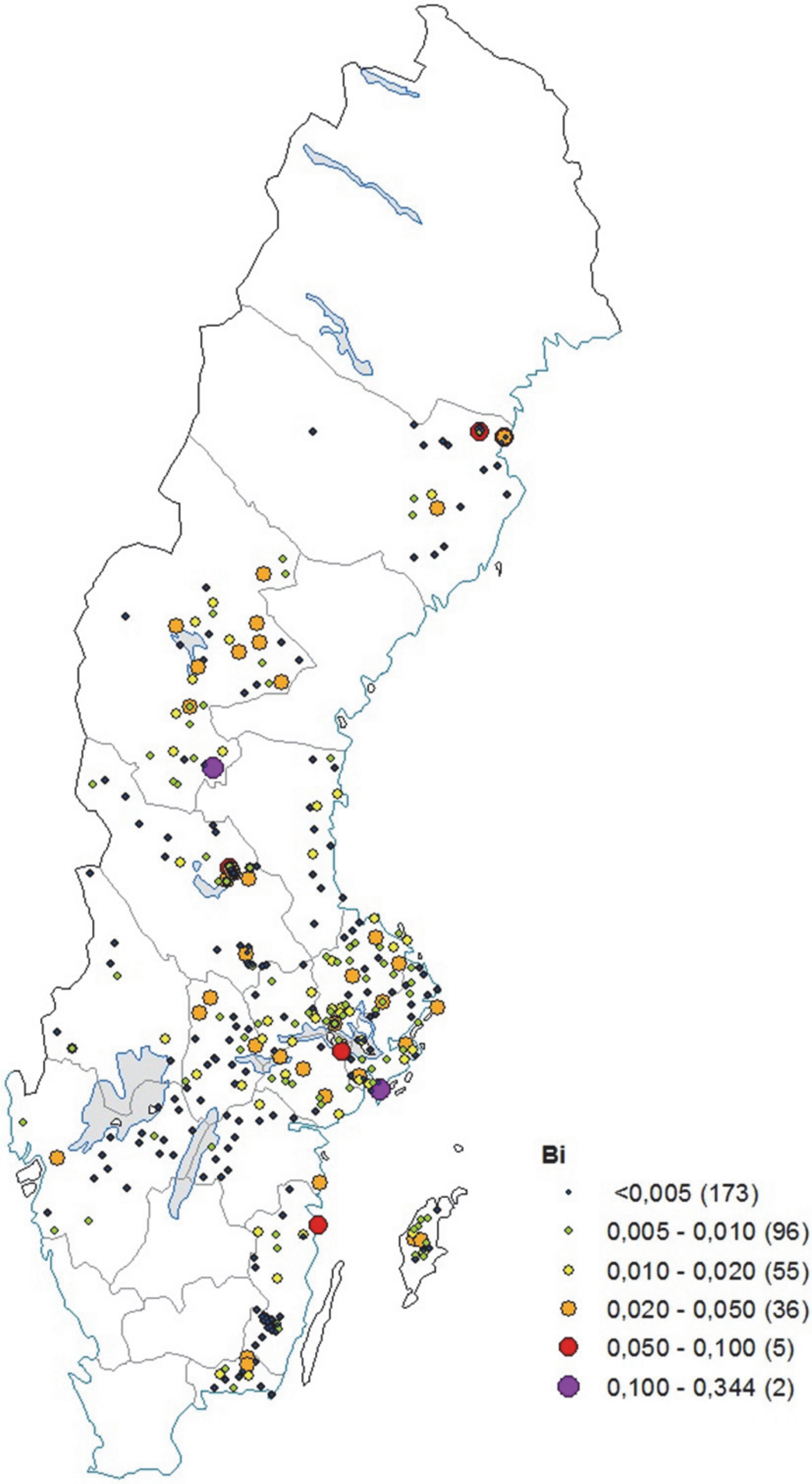

Figure 27. Bismuth concentrations in Swedish drilled well water samples (μg/L).

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