# European Ground Water Geochemistry Atlas: Using Bottled Water as a Sampling Medium


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# EUROPEAN GROUND WATER GEOCHEMISTRY USING BOTTLED WATER AS A SAMPLING MEDIUM

## Structure of Seminar presentation

- Introduction
- Development of project idea
- Issues that were considered in the use of bottled water
- European Directives
- Development of a harmonised database
- Analytical programme
- Quality control
- Leaching of elements from bottle materials
- Comparison with other data sets
- Geochemical distribution maps
- Publications
- Conclusions

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# Sample materials for the Geochemical Atlas of Europe:

- surface (stream) water
- overbank sediment
- stream sediment
- topsoil (0-25 cm)
- subsoil (C-horizon)



**Missing: Ground water**

http://www.gtk.fi/publ/foregsatlas/

http://www.gsf.fi/foregs/geochem/fieldman.pdf

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# Geochemistry of European Ground water

## Ground water

- It is a difficult medium to sample (contamination issues)
- It is difficult to map [3D-regional distribution (aquifers)]
- It has a high local variation

**Ground water is, therefore, a medium that it is *impossible to sample systematically and analyse at a reasonable cost*.**

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**The Federal Institute for Geosciences and Natural Resources (BGR) offers to analyse free of charge all water samples.**

***There is, however, an almost impossible problem:***

***No funds are available to carry out systematic sampling of ground water in Europe.***

**Does one give up?**

**or**

***is there a cost-effective method to map the geochemistry of ground water at the European scale?***

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# Atlas of European Ground water Geochemistry

Manfred Birke (BGR)

**Project idea:**

**Ground water can be bought readily sampled at the European scale – more than 1900 bottled water brands are registered in Europe.**

**According to Manfred’s experience, bottled water can be used as a “proxy“ to ground water.**

# In Europe there are 3 categories of bottled water:

(1) **Natural mineral water** (Directives 80/777/EC & 2003/40/EC, 2009/54/EC for limit values): It is drinking water of underground origin with a stable chemical composition, and it is bottled at source (spring/well/borehole). ***It is not subjected to any treatment***, and differs from common drinking water by its mineral content, trace elements or other constituents.

(2) **Spring water** (Directives 96/70/EC & 98/83/EC for limit values): It is drinking water of underground origin, and is bottled at source (spring/well/ borehole). ***It is not subjected to any treatment***. It is not required to have a stable chemical composition. Its physicochemical characteristics (parameters) must satisfy the standards of drinking water for human consumption.

(3) **Table water** (Directive 98/83/EC for limit values): It is bottled water for human consumption. Its physicochemical characteristics (parameters) satisfy those of common drinking water. It can even be normal tap water or treated and disinfected river water.

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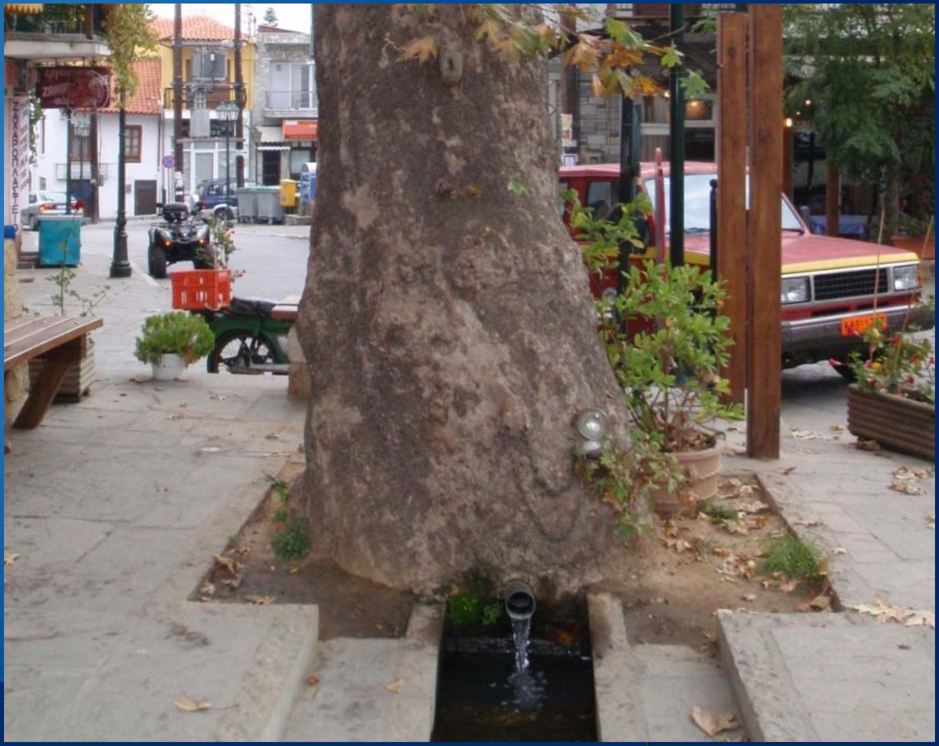
***According to European legislation, the majority of bottled water brands, sold in supermarkets, are from natural ground water.***



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***Decision time!!!***

**Do we go ahead with the project?**

**No**

**Yes**

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***It is noted that most members of the EuroGeoSurveys Geochemistry Expert Group are Applied Geochemists with experience in mineral exploration, where they learnt to sample and measure systematically any natural medium to meet project objectives in a cost-effective manner.***

**Manfred’s proposal was discussed, and although it was met with considerable skepticism, the explorationists among the group decided that it was worth a try, since the cost of sampling was minimal, *i.e.*, just the cost of purchase of bottled water and of posting to Germany.**

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# Atlas of European Ground water Geochemistry


**The sampling or purchase of bottled water from supermarkets started in November 2007 and was completed by April 2008.**

**In total, 1785 "samples" of bottled water were purchased from supermarkets all over Europe (38 countries), representing 1247 boreholes/wells at 884 unique locations.**

**Sample storage at BGR, Berlin (Reimann & Birke, 2010, Fig. 19, p.30)**

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## Issues when using ”Bottled Water” as a proxy for ground water:

- Bottled water may come from non-representative, quite special and unusual aquifers;

- Bottled water is sometimes treated prior to bottling (filtered, carbonated, de-ironed, *etc*.);

- Bottled water samples can be contaminated from well and bottling installations, and

- Bottled water samples can be contaminated from the bottle material itself.

# Atlas of European Ground water Geochemistry


**To obtain a fully harmonised data set all samples were analysed in just one laboratory for 72 parameters.**

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# Atlas of European Ground water Geochemistry

## Analytical programme, BGR-lab:

**ICP-QMS**: Ag, Al, As, B, Ba, Be, Bi, Cd, Ca, Ce, Co, Cr, Cs, Cu, Er, Eu, Fe, Ga, Gd, Ge, Hf, Hg, Ho, I, K, La, Li, Lu, Mg, Mn, Mo, Na, Nb, Nd, Ni, Pb, Pr, Rb, Sb, Sc, Se, Sm, Sn, Sr, Ta, Tb, Te, Th, Ti, Tl, Tm, U, V, W, Y, Yb, Zn, Zr

**ICP-AES**: Ba, Ca, K, Mg, Mn, Na, Sr, P, Si

**IC:** $Br^-$, $Cl^-$, $F^-$, $NO_2^-$, $NO_3^-$, $SO_4^{2-}$

**AFS**: Hg

**Titration**: tAlk - $HCO_3^-$

**Photometric**: $NH_4^+$

**Potentiometric**: pH

**Conductometric**: EC

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# QUALITY CONTROL (1/4)

A very strict quality control programme was installed:

(1) Analysis of international reference samples to document the trueness of analytical results;
(2) Frequent analysis of an in-house project standard (MinWas) to check the accuracy of determined parameters;
(3) Frequent analysis of blank samples to detect any contamination issues, and to derive reliable detection limits;
(4) Frequent analysis of sample duplicates to determine precision of measurements;

# QUALITY CONTROL (2/4)

(5) Comparison of analytical results of this study with those displayed on bottle labels;

(6) Determination of few parameters (Ba, Ca, K, Mg, Mn, Na & Sr) by ICP-QMS and ICP-AES, and Hg by ICP-QMS and AFS;

(7) Checking of bottled water samples with unusually high results for important parameters by buying another bottle and repeating the analysis.

# QUALITY CONTROL (3/4)

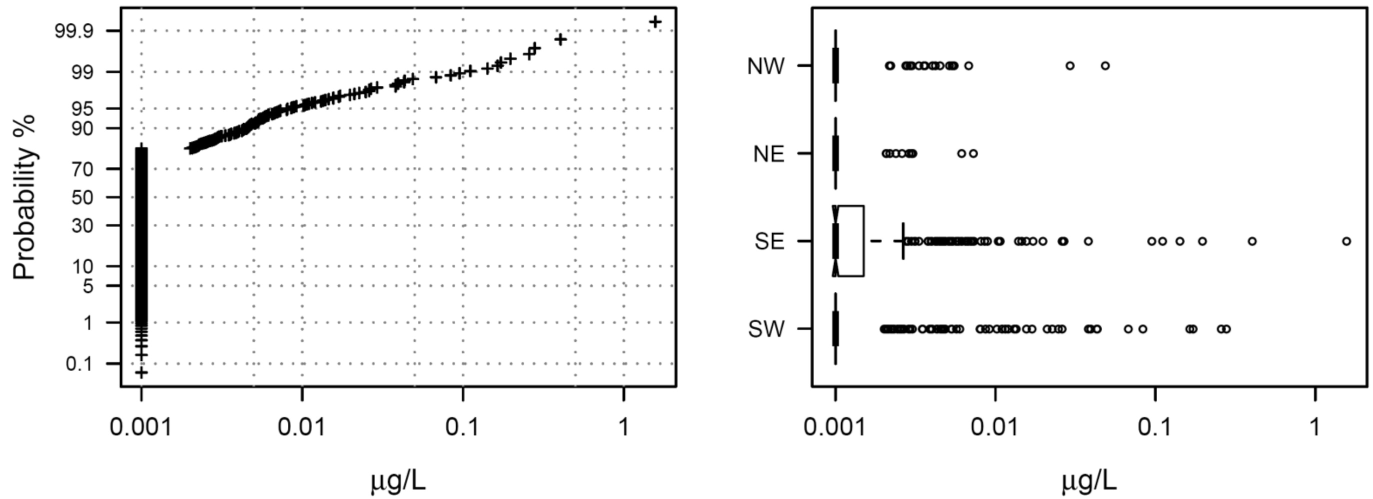
- The international sample SLRS-4 was analysed 103 times;
- The low fortified standards for trace elements TM-26.3, TM-27.2, TM-28.2 and TM-28.3 were analysed from 25 to 91 times;
- The in-house laboratory reference sample ”MinWas” was analysed 261 times;
- The blank samples of deionised water (SERALPUR-90; 18.2 MΩ) and 4 ml 69% $HNO_3$ (Roth, ultrapure) were analysed from 10 to 12 times.

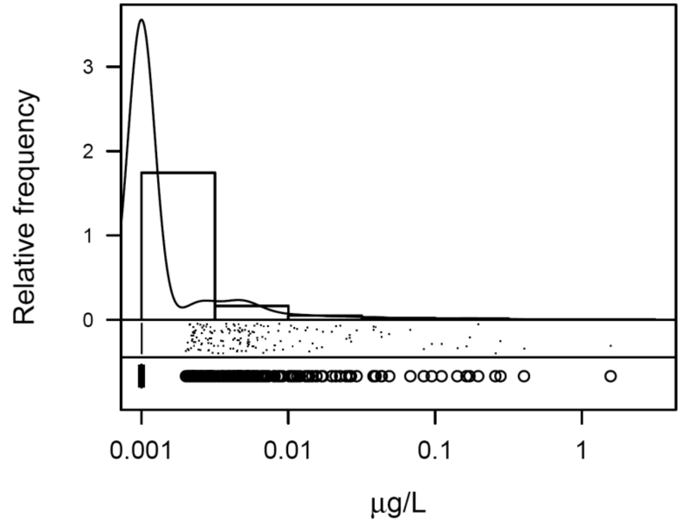
# QUALITY CONTROL PROBLEMS (4/4)

**The only problems that were detected, during the study of quality control results, concerned the reproducibility of Hf, Nb, Sn, Ta and W at low concentrations.** ***Consequently, the results of these elements up to concentrations 10 times their detection limit should be considered with some caution*****.**

**However, the results of reference samples at higher concentrations show that these are reliable.**

Drinking water standard: none defined




Analytical Method: ICP−QMS
Detection limit: 0.002 µg/L
722 (81.7%) < detection limit

| | |
|---|---|
| n: | 884 |
| Minimum: | < 0.002 µg/L |
| 5%: | < 0.002 µg/L |
| 25%: | < 0.002 µg/L |
| Median: | < 0.002 µg/L |
| 75%: | < 0.002 µg/L |
| 95%: | 0.0081 µg/L |
| Maximum: | 1.57 µg/L |

# Hafnium

# Hf

Thompson-Howarth plot

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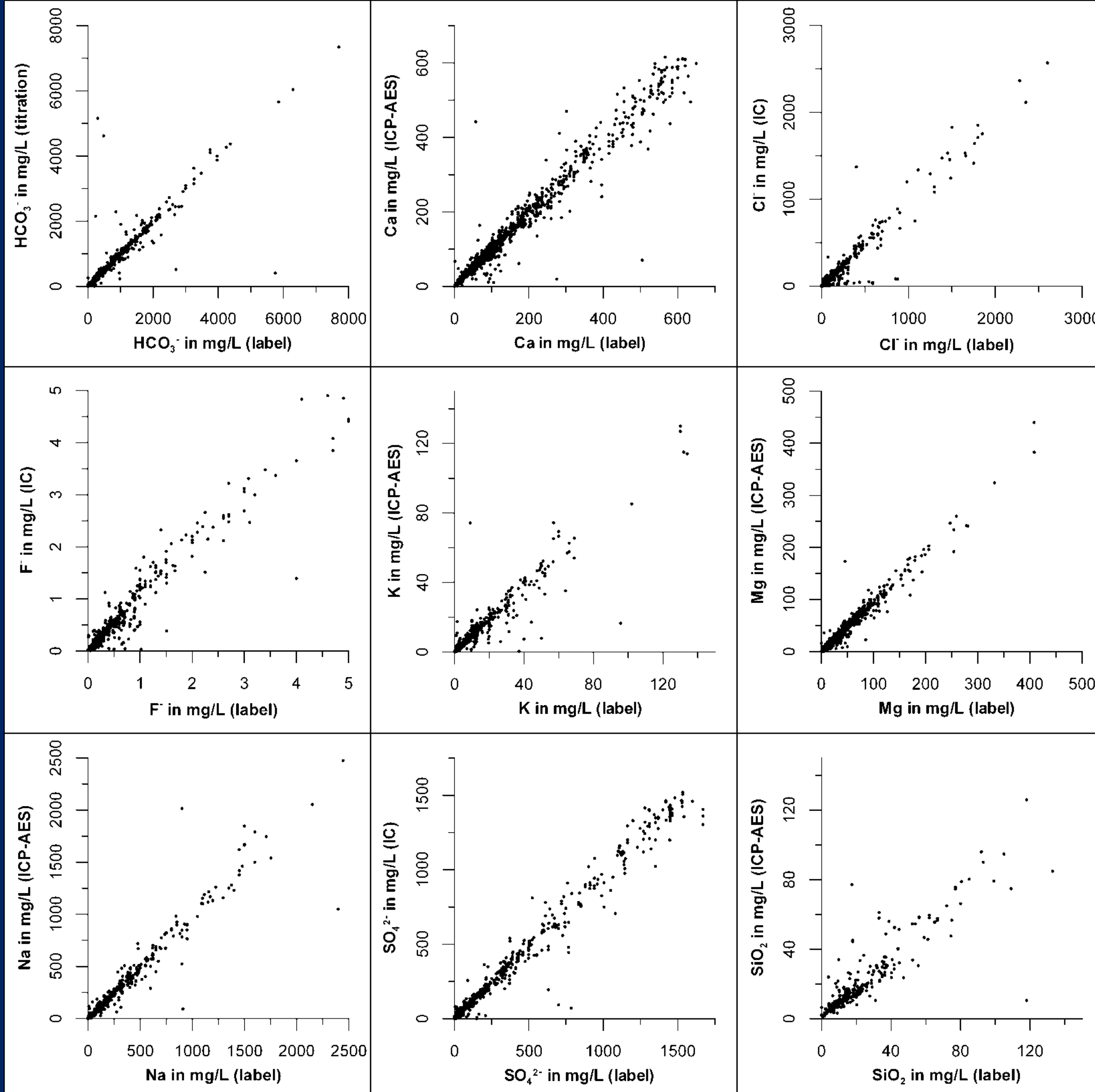


Comparison of analytical data of the EuroGeoSurveys study with those displayed on bottle labels show a fairly good linear correlation.

(Reimann & Birke, 2010, Fig. 20, p.44)

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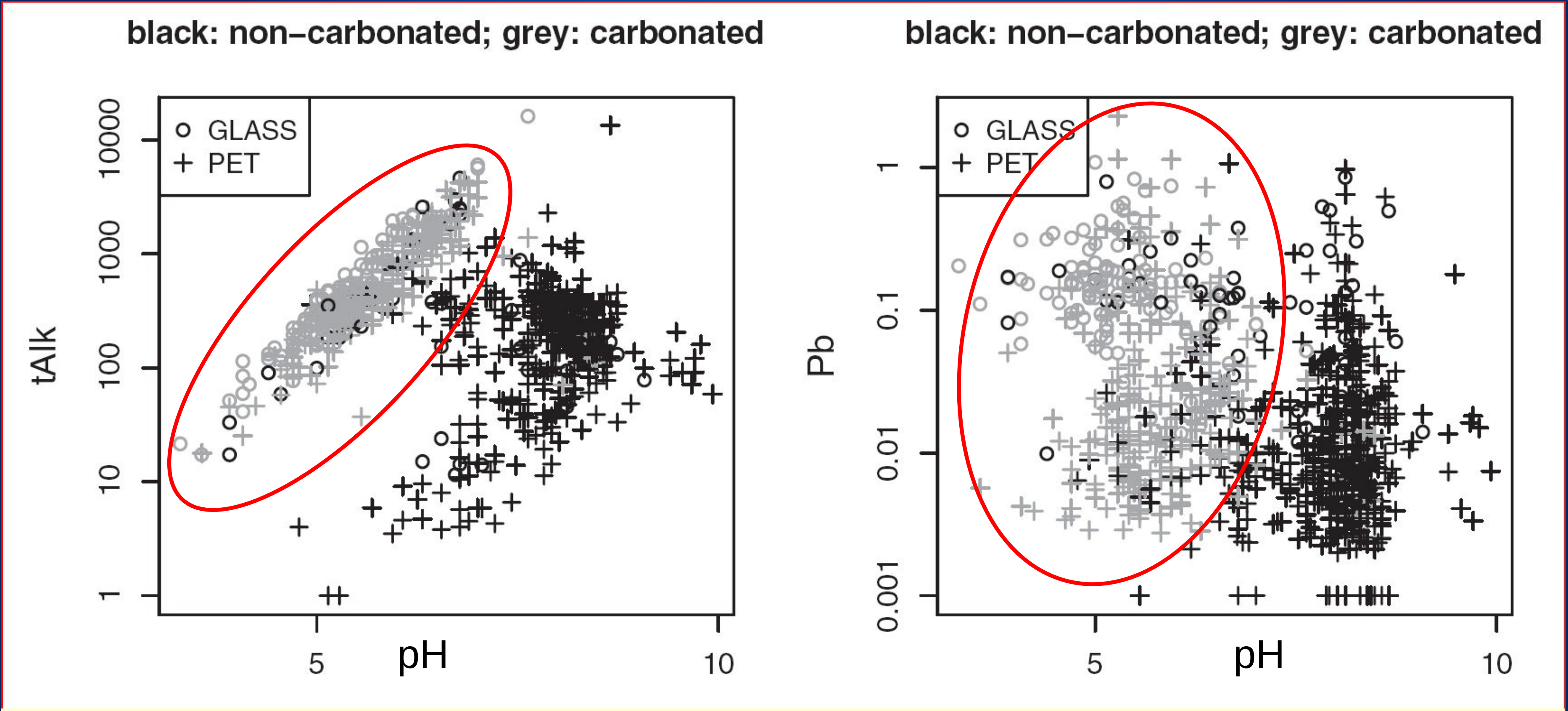
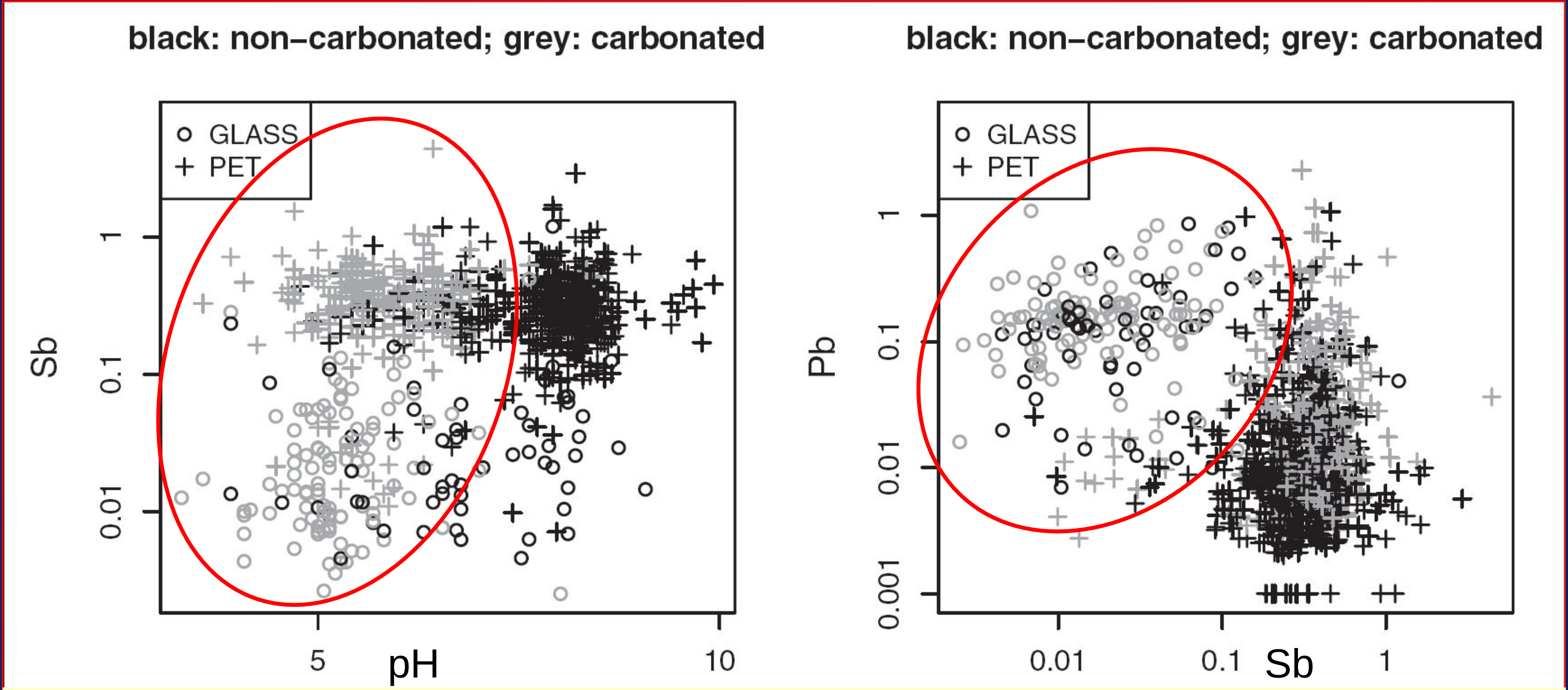
# Leaching of elements from bottle material



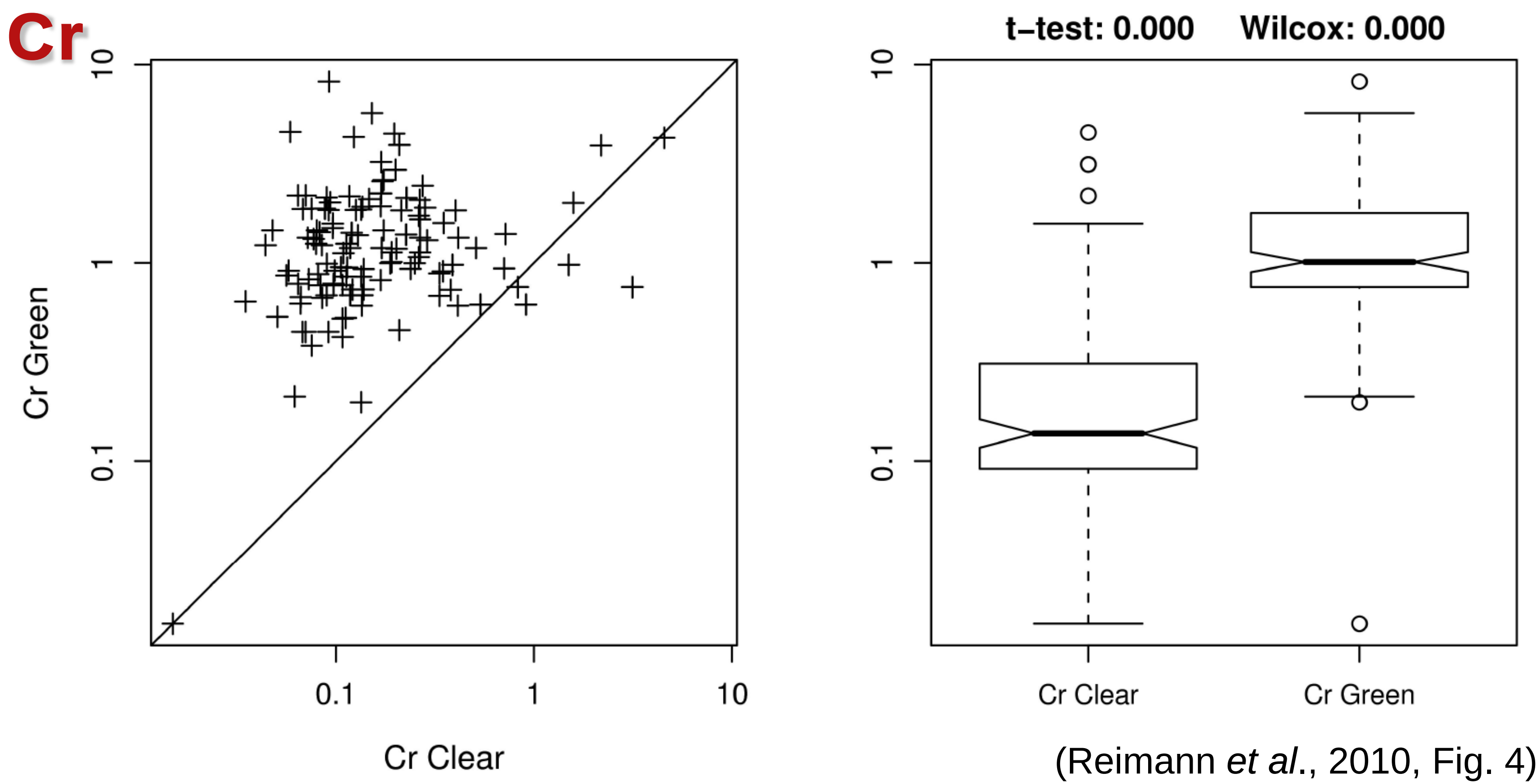

**Scattergrams showing dependence of (a) pH in relation to total alkalinity [tAlk meq/L], and (b) pH in relation to Pb [µg/L], according to bottle type [o glass & + PET. Still water in black – Carbonated water in grey] (Reimann & Birke, 2010, Fig. 21, p.47)**

# Leaching of elements from bottle material




**Scattergrams showing dependence of (a) pH in relation to Sb [µg/L], and (b) Sb [µg/L] in relation to Pb [µg/L], according to bottle type [o glass & + PET. Still water in black – Carbonated water in grey) (Reimann & Birke, 2010, Fig. 21, p.47)**

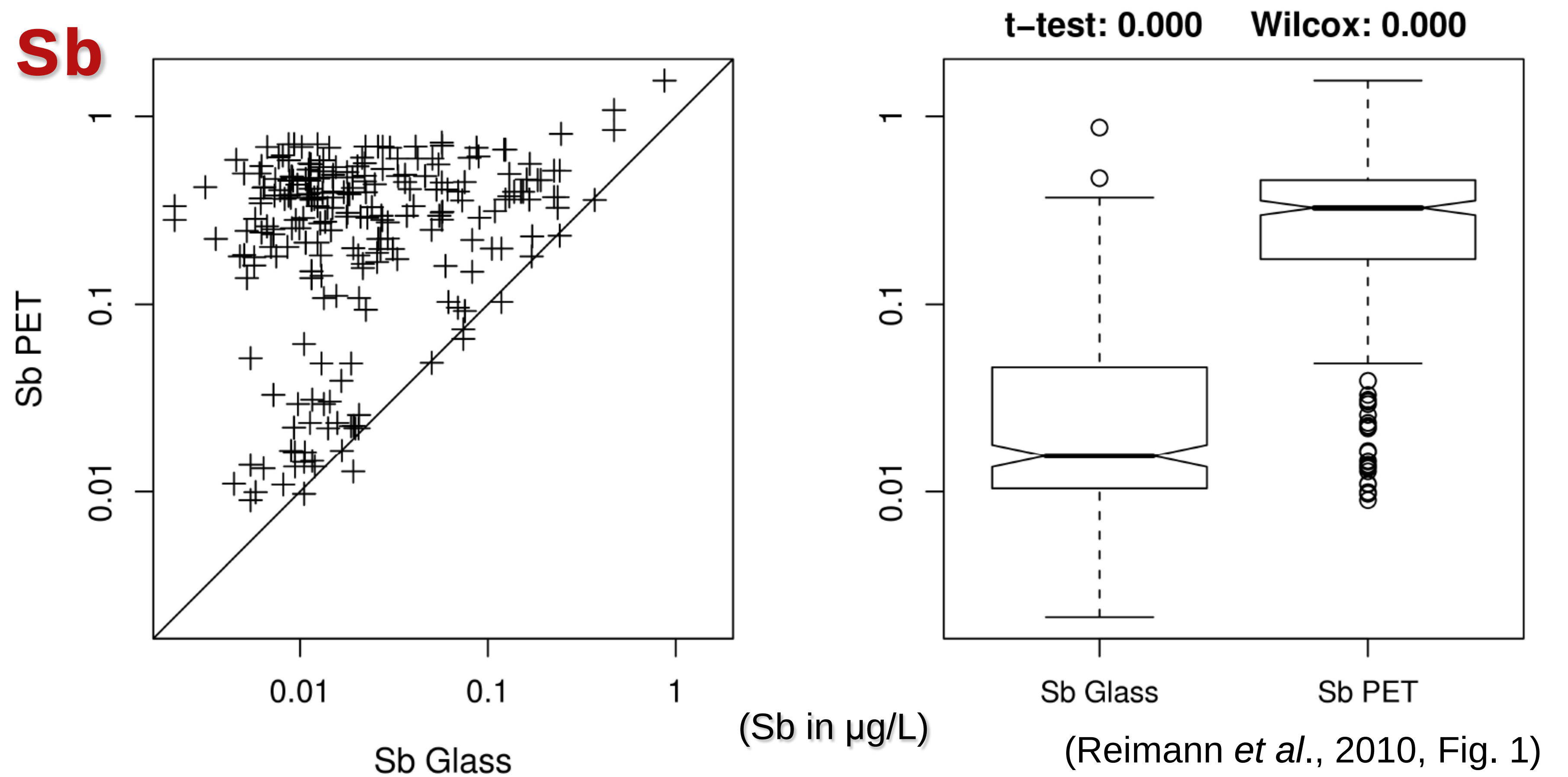
# Leaching of elements from bottle material




(Reimann *et al*., 2010, Fig. 4)

**Green glass bottles release more Cr to the water than clear bottles**

# Leaching of elements from bottle material




(Reimann *et al*., 2010, Fig. 1)

**Comparison of the same type of bottle water sold in glass and PET bottles (N=131)**

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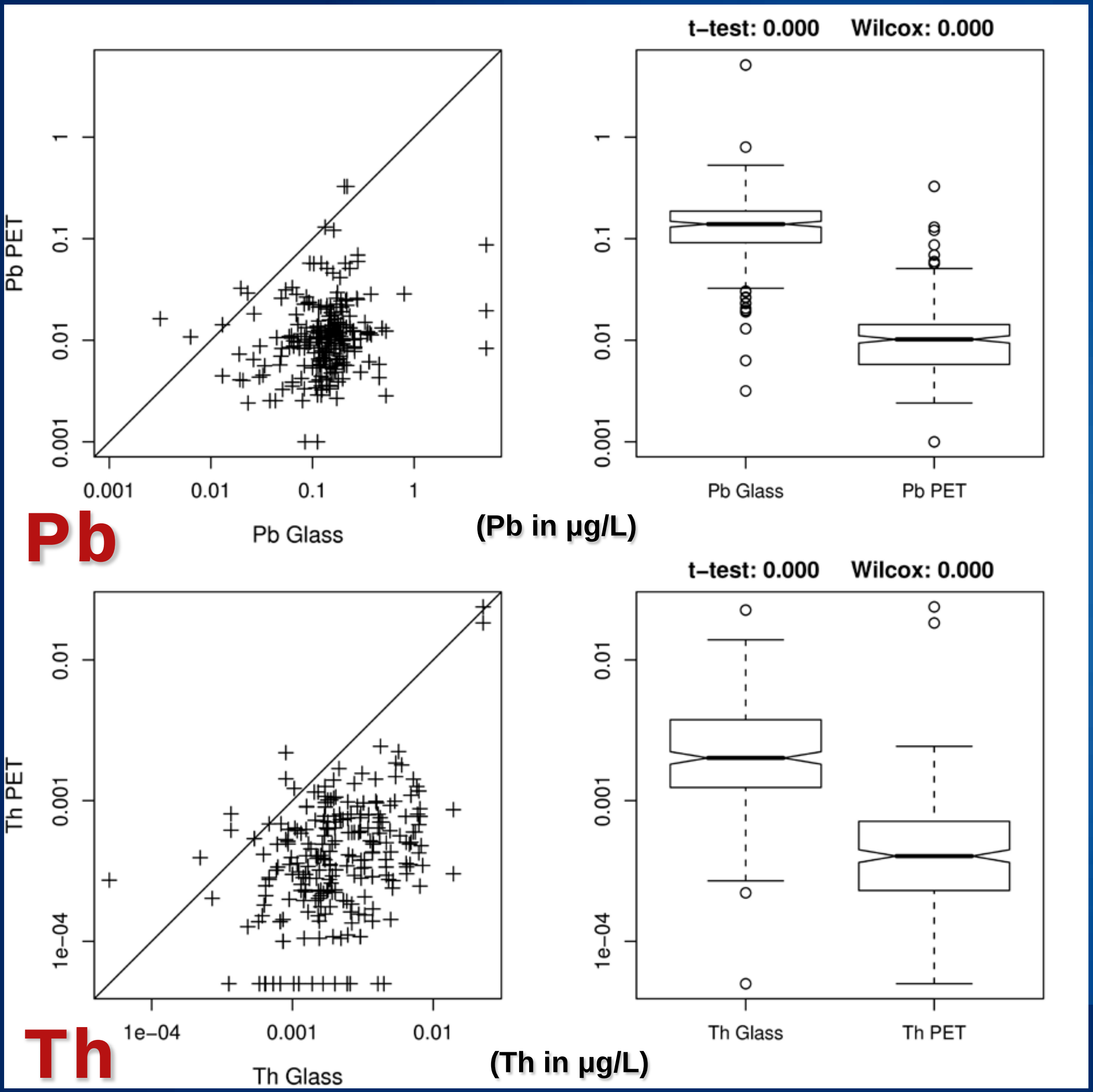


# Leaching of elements from bottle material

**Glass affects more the concentrations of elements such as Ce, Pb, Al, Zr, Ti, Hf, Th and La**

**(N=131)**

(Reimann *et al*., 2010, Fig. 2)

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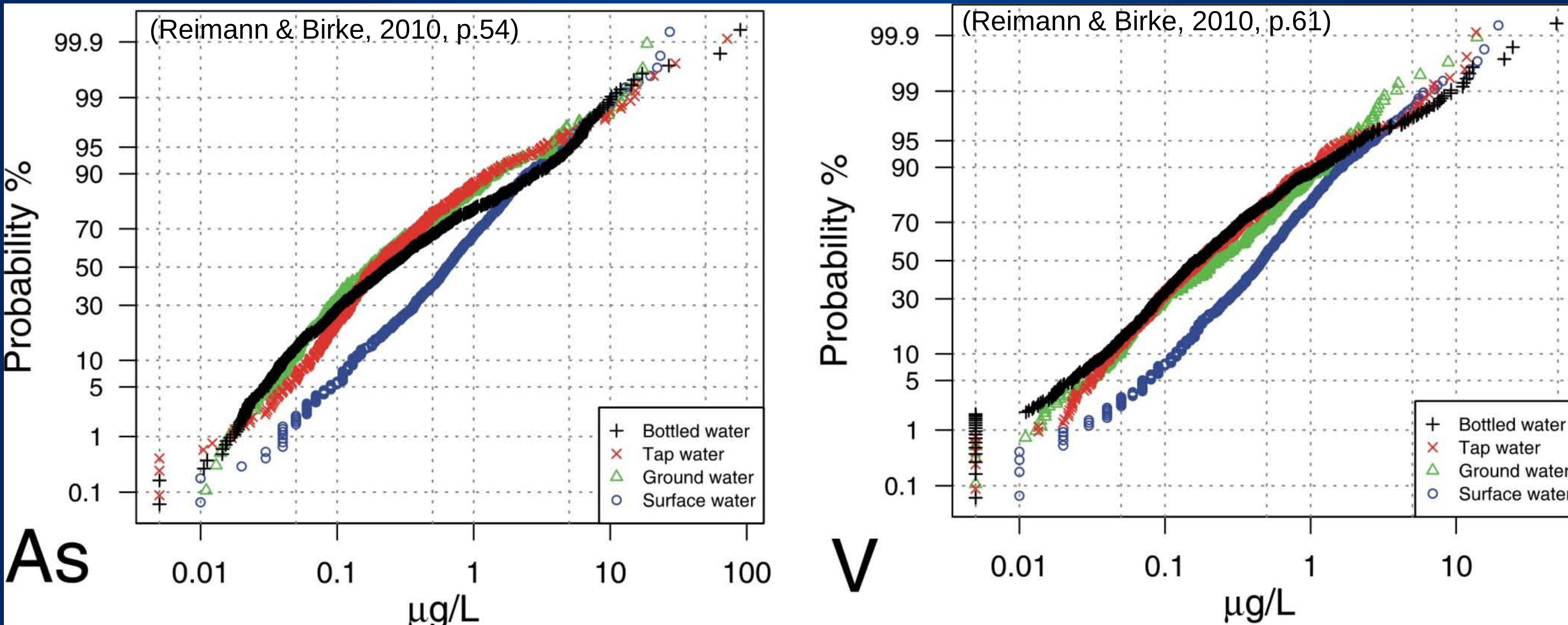
# COMPARISON OF RESULTS IN FOUR DATA SETS OF WATER SAMPLES

- **European bottled water** (+ black, N=884),
- **European tap water** (x red, N=586),
- **Norwegian ground water** (Δ green) [Frengstad *et al*., 2000],
- **European stream water** (o blue) from the FOREGS Geochemical Atlas of Europe (Salminen *et al*., 2005)




# As and V in European water

## Most elements show a surprising comparable distribution in all 4 data sets

**European bottled water (+ black, N=884), European tap water (x red, N=586), Norwegian ground water (Δ green) [Frengstad *et al*., 2000], European stream water (o blue) from the FOREGS project (Salminen *et al*., 2005)**

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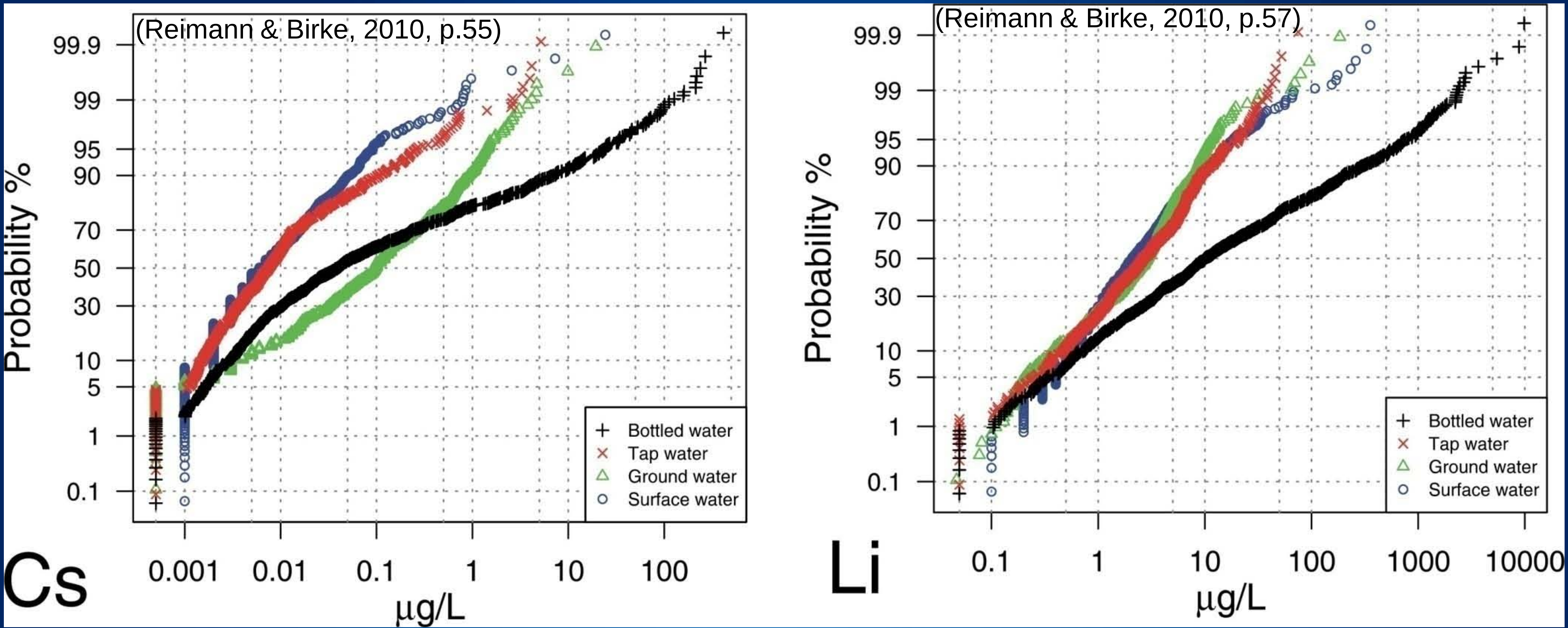
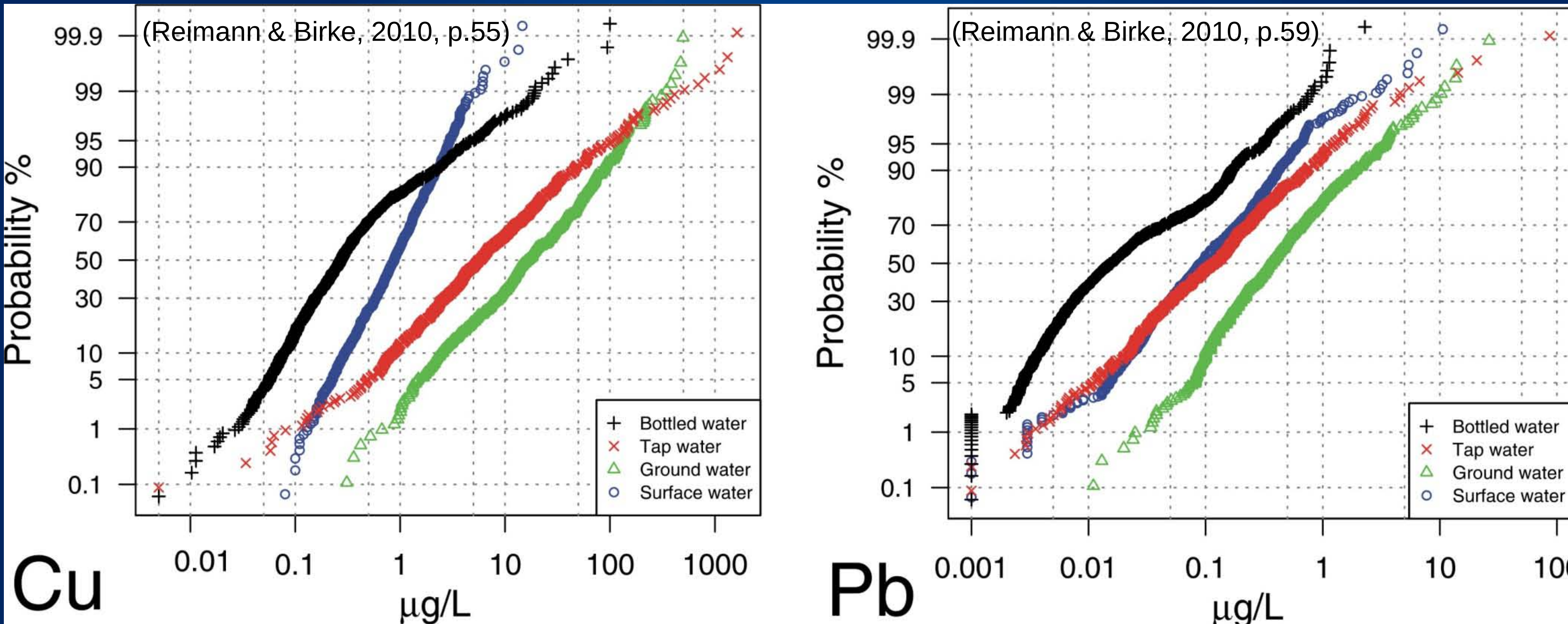



**Some elements (*Ag, B, Be, Br, Cl, Cs, F, Ge, I, K, Li, Na, Rb, Sr, Te, Tl, Zr*) show a "mineral water" specific enrichment (+1 to +2 orders of magnitude variation) – brines and hydrothermal water – deep sources**

**European bottled water (+ black, N=884), European tap water (x red, N=586), Norwegian ground water (Δ green) [Frengstad *et al*., 2000], European stream water (o blue) from the FOREGS project (Salminen *et al*., 2005)**




**Some elements (Cu, Pb, Zn) show definite indications that European tap water and Norwegian ground water are contaminated by well installations and water piping – over the whole concentration range**

**European bottled water (+ black, N=884), European tap water (x red, N=586),**

**Norwegian ground water (Δ green) [Frengstad *et al*., 2000],**

**European stream water (o blue) from the FOREGS project (Salminen *et al*., 2005)**

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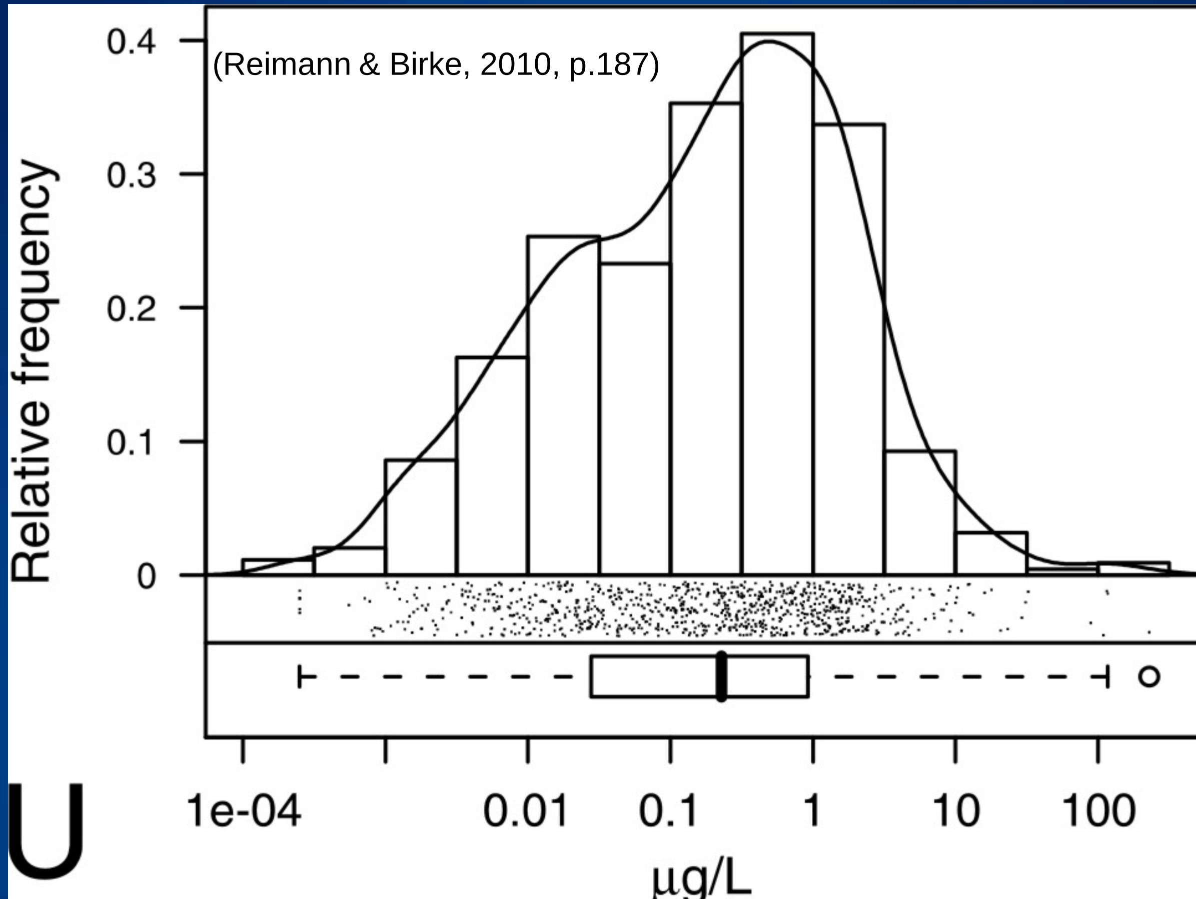
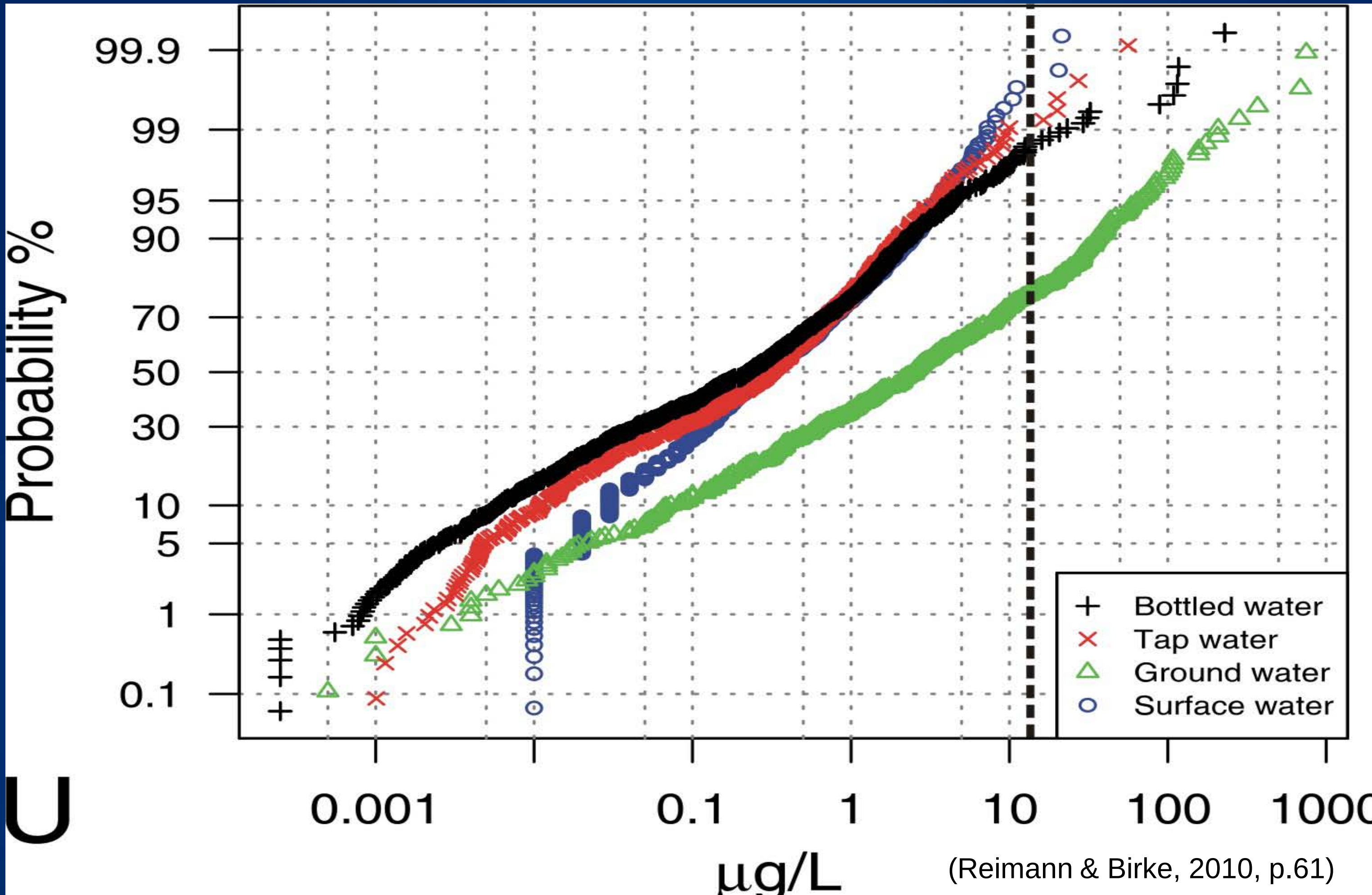



**Distribution of U in samples of bottled water (N = 884). Observe the sudden drop of values at 10 µg/L.**

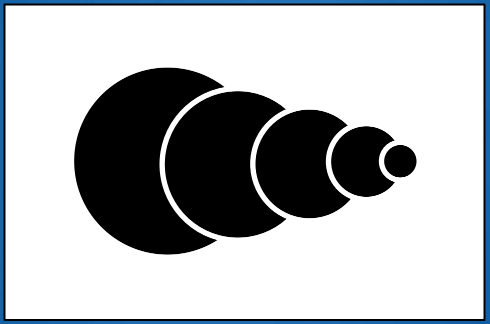
***The World Health Organisation has proposed a guideline value of 15 µg/L and it appears that the bottling companies are observing this limit.***




**European bottled water (+ black, N=884), European tap water (x red, N=586),**
**Norwegian ground water (Δ green) [Frengstad *et al*., 2000],**
**European stream water (o blue) from the FOREGS project (Salminen *et al*., 2005)**

# Distribution maps and interpretation text

Distribution maps were prepared using the ”variable-size” or ”growing dot” approach.



Clear regional patterns emerge for the majority of elements.

## Lithium

**Li**

Lithium (atomic number 3) is the lightest of all metals, occurring in nature in the +1 oxidation state. Spodumene ($LiAlSi_2O_6$) and lepidolite ($K_2Li_3Al_4Si_7O_{21}(OH,F)_3$) are but two typical Li minerals. More geochemically important, however, is the occurrence of Li in micas and in K-feldspars, amphiboles and clay minerals. Lithium is generally more enriched in acidic rocks (granites) and in many sedimentary rocks (especially marine shales and schists) than in basic (e.g. basalts) or ultrabasic rocks. Lithium is especially common in minerals crystallised from late-stage melts (e.g. in pegmatite minerals). Brines are strongly enriched in Li.

The $Li^+$ ion is generally stable and soluble in the groundwater environment, although its mobility may be modified due to sorption onto clay minerals and zeolites. Lithium is used in heat-resistant glass and ceramics, high strength-to-weight alloys used in the aircraft industry and ever increasingly in lithium batteries. It is also used in reagents and catalysts in the pharmaceuticals and organic chemical industries. Compared to freshwater, Li is strongly enriched in ocean water. Hem (1992) reports a mean value of 170 µg/l in the ocean.

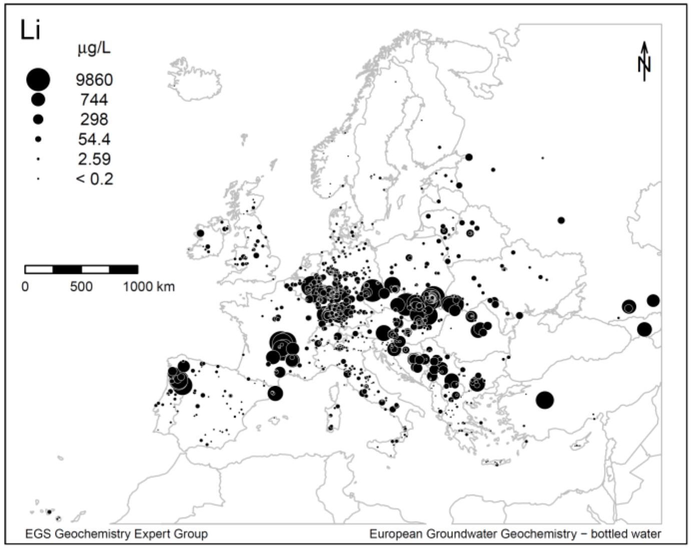
A typical concentration of Li in surface water has been estimated to be 3 µg/l (see Koljonen, 1992), a value that appears quite reasonable in the light of the results for European surface water (median = 2.1 µg/l - Salminen et al., 2005). The median value presented here for bottled water is several times higher, at 10 µg/l.

The map depicts a number of clear Li anomalies. Many of the hot spots (e.g. northern Portugal, France) are related to young granitic intrusions (compare K, Cs, Rb). Many wells from the general area of the Carpathian Mountains and the Dinarides also exhibit enhanced Li concentrations. In Germany, high Li-concentrations are associated with wells in Jurassic and Triassic sediments, but a well drawing water from an important fault zone is also indicated by an unusually high Li value. The highest value (9860 µg/l) was observed in a bottled water from Slovakia.

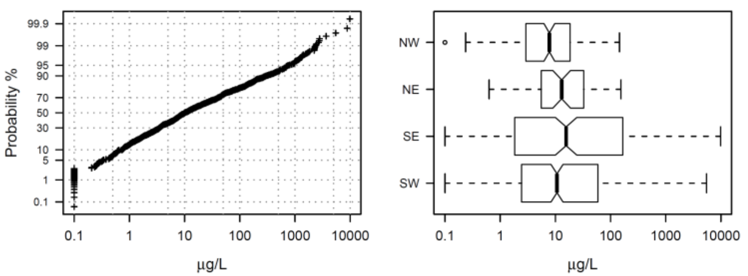
Bottle leaching has no influence on the observed Li distribution on the map at circumneutral pH, while minor leaching may occur from glass bottles at low pH (up to 3.23 µg/l).

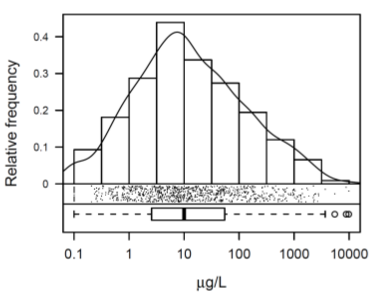
No water standard is defined in the EU for Li. However, Li is a biologically active element and Li-based drugs have been used to treat manic-depressive conditions since the 1950s. It is likely that some of the bottled waters with the highest Li concentrations have medical effects (see, e.g. Ohgami et al., 2009). Given the enormous natural variation (6 orders of magnitude) it is one of the elements where deficiency may play a role as well. According to Bradford (1963), Li can be toxic to some plants at concentrations >60 µg/l. In the CP-Diagram comparing the bottled water dataset with tap water, Norwegian bedrock groundwater and European surface water (Figure 26), Li concentrations are clearly highest in the bottled water. Indeed, Li is one of the elements (together with B, Br, Cs, K, Ge, Rb) where high concentrations are rather characteristic of "mineral water". The CP-diagram suggests that variation in Li concentrations in European bottled water spans fully 6 orders of magnitude, while the variation for European tap water, surface water and Norwegian bedrock groundwater is "only" 4-5 orders of magnitude.

(Reimann & Birke, 2010, p.130-131)




Drinking water standard: none defined



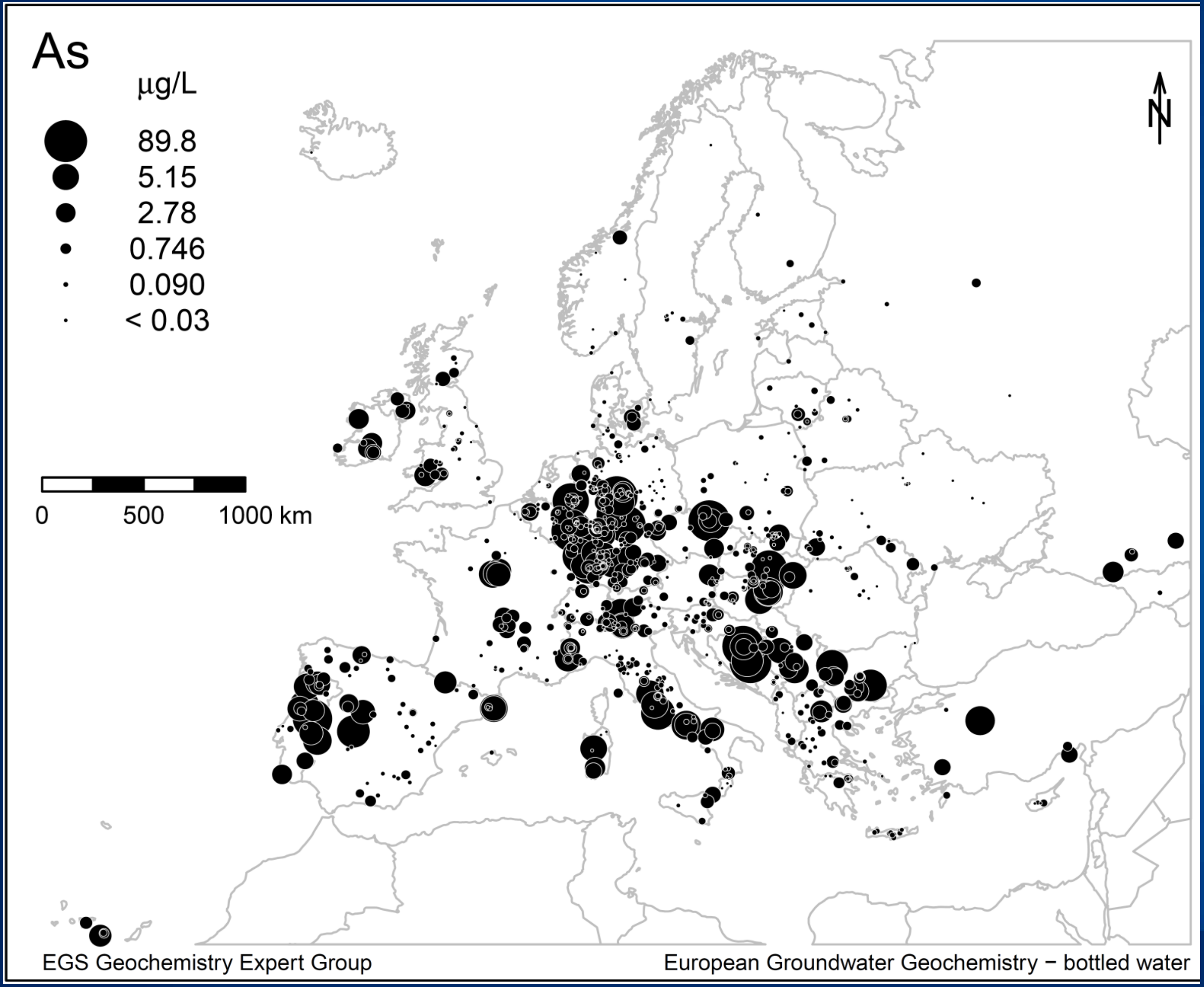




Analytical Method: ICP–QMS
Detection limit: 0.2 µg/L
24 (2.71%) < detection limit

| | |
|---|---|
| n: | 884 |
| Minimum: | < 0.2 µg/L |
| 5%: | 0.335 µg/L |
| 25%: | 2.59 µg/L |
| Median: | 10.0 µg/L |
| 75%: | 54.4 µg/L |
| 95%: | 744 µg/L |
| Maximum: | 9860 µg/L |

**Lithium** **Li**

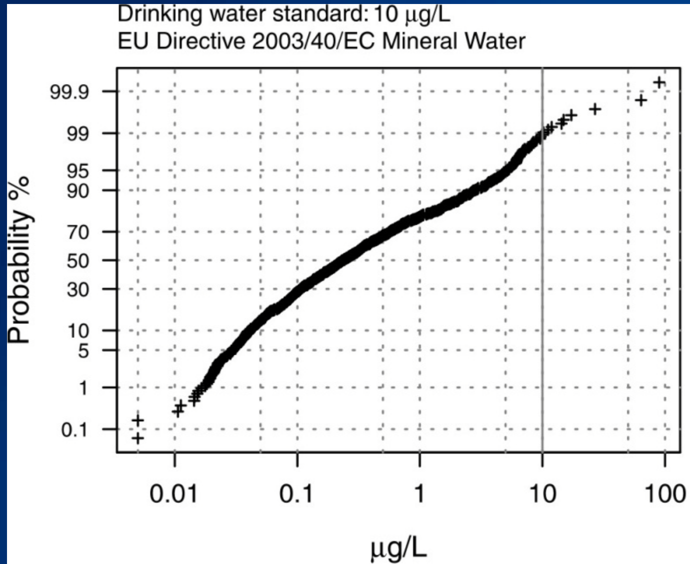
(Reimann & Birke, 2010, p.77)

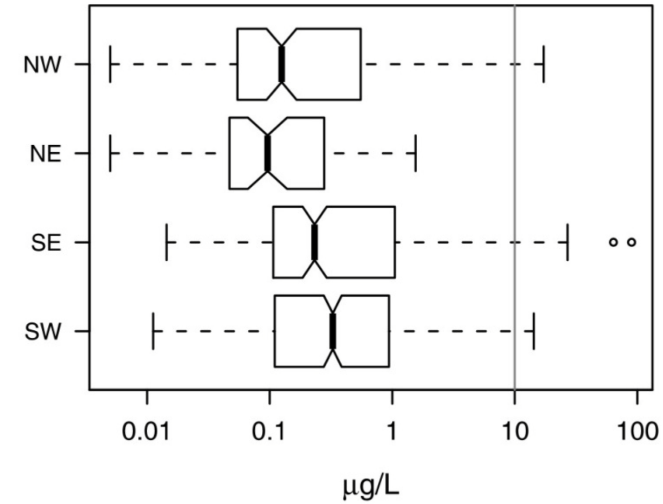


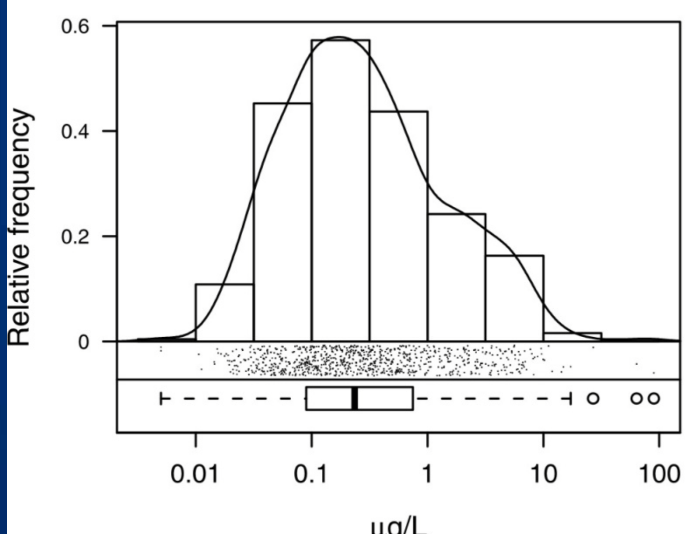

**Arsenic (As):**

**Sulphide mineralisation, fault zones**

(Reimann & Birke, 2010, p.77)




Analytical Method: ICP-QMS
Detection limit: 0.01 µg/L
2 (0.23%) < detection limit

| | |
|---|---|
| n: | 884 |
| Minimum: | < 0.01 µg/L |
| 5%: | 0.030 µg/L |
| 25%: | 0.090 µg/L |
| Median: | 0.235 µg/L |
| 75%: | 0.746 µg/L |
| 95%: | 5.15 µg/L |
| Maximum: | 89.8 µg/L |

# Arsenic

# As

# INTERPRETATION

Reimann & Birke, 2010, p.77)

**For effective interpretation a lot of background information is required:**

- **Lithological map**
- **Structural (tectonic) map**
- **Mineral deposits / outcrops map**
- **Geothermal gradient map**
- **Rainfall intensity map**
- **Land use map**
- **Municipal waste sites map**

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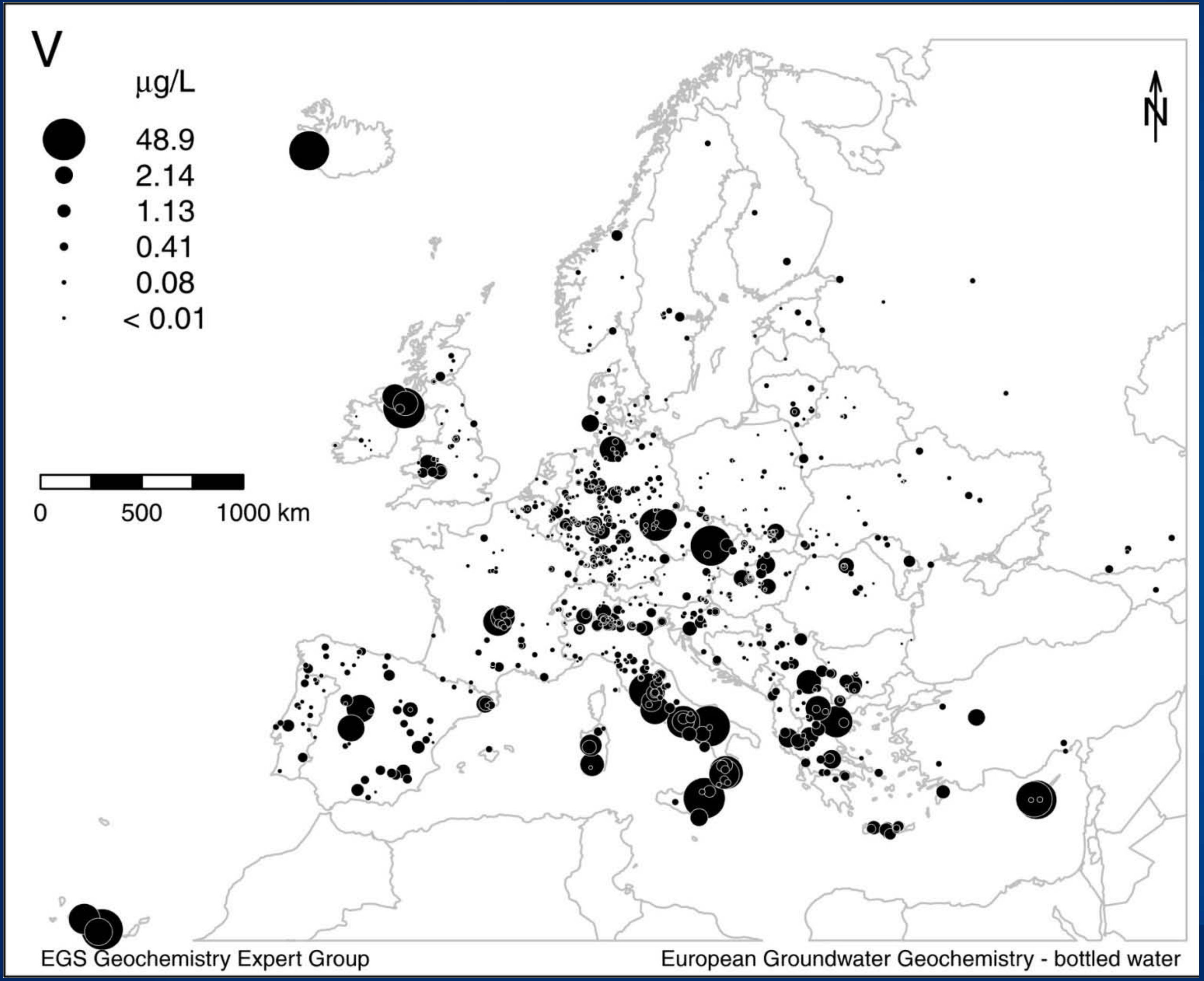
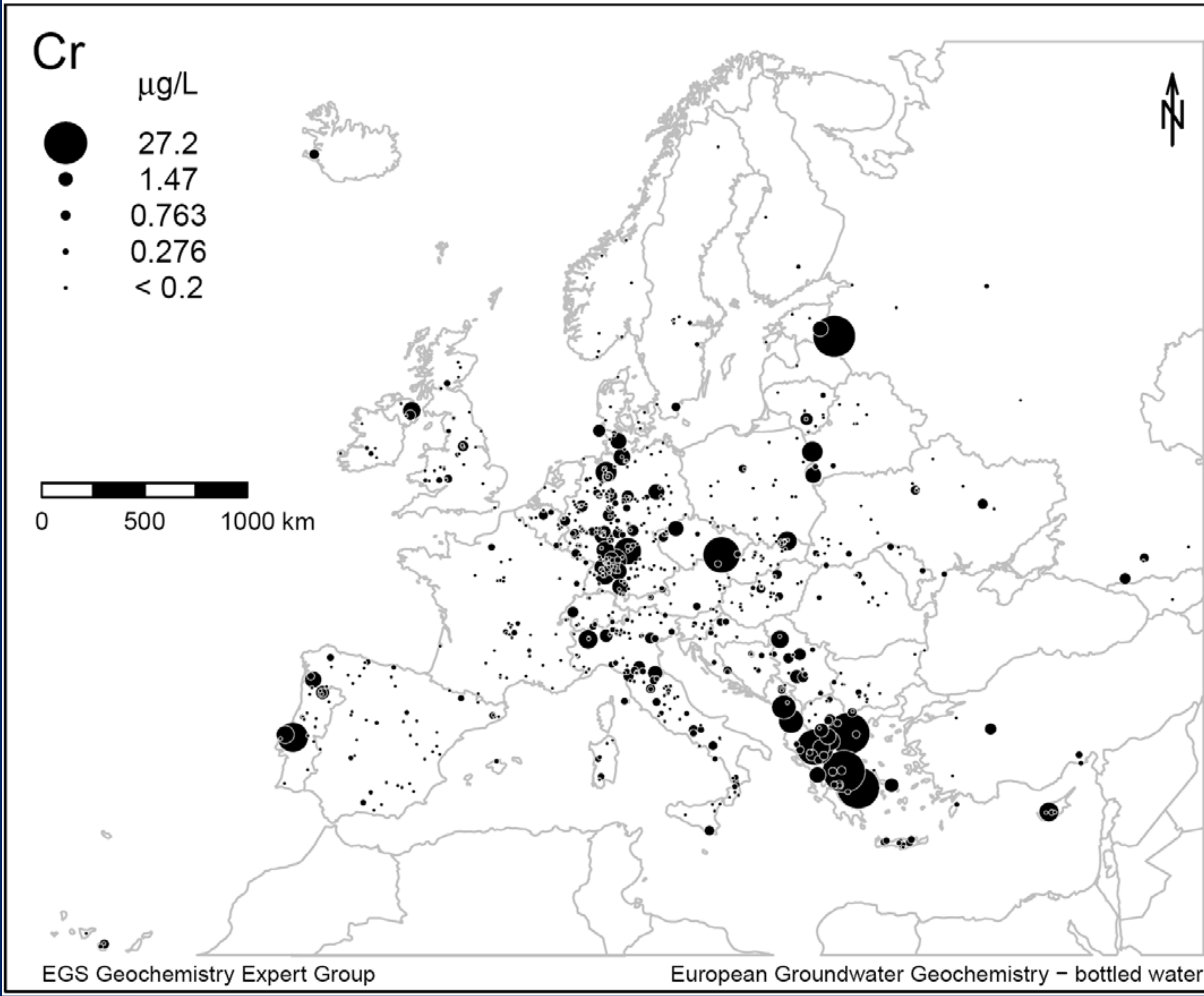



**Vanadium (V):**

**active volcanic centres and basaltic rocks are indicated**

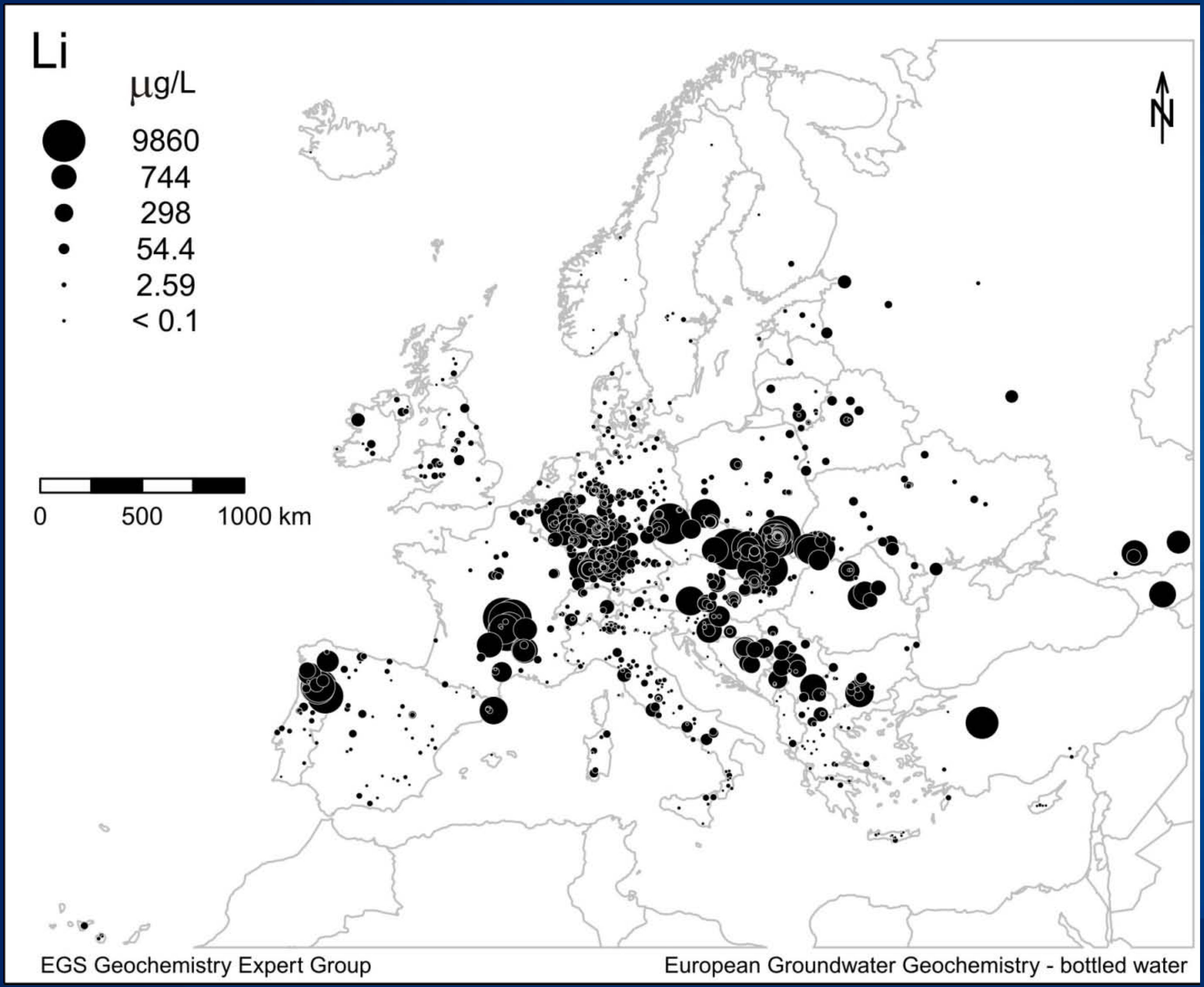
(Reimann & Birke, 2010, p.189)




# Chromium (Cr):

## Ophiolites (mafic-ultramafic rocks)

(Reimann & Birke, 2010, p.99)




# Lithium (Li):

Hercynian granites, Jurassic and Triassic sediments in Germany, and deep sourced water wells, e.g., in the Carpathian Mountain Chain, Dinarides

(Reimann & Birke, 2010, p.131)

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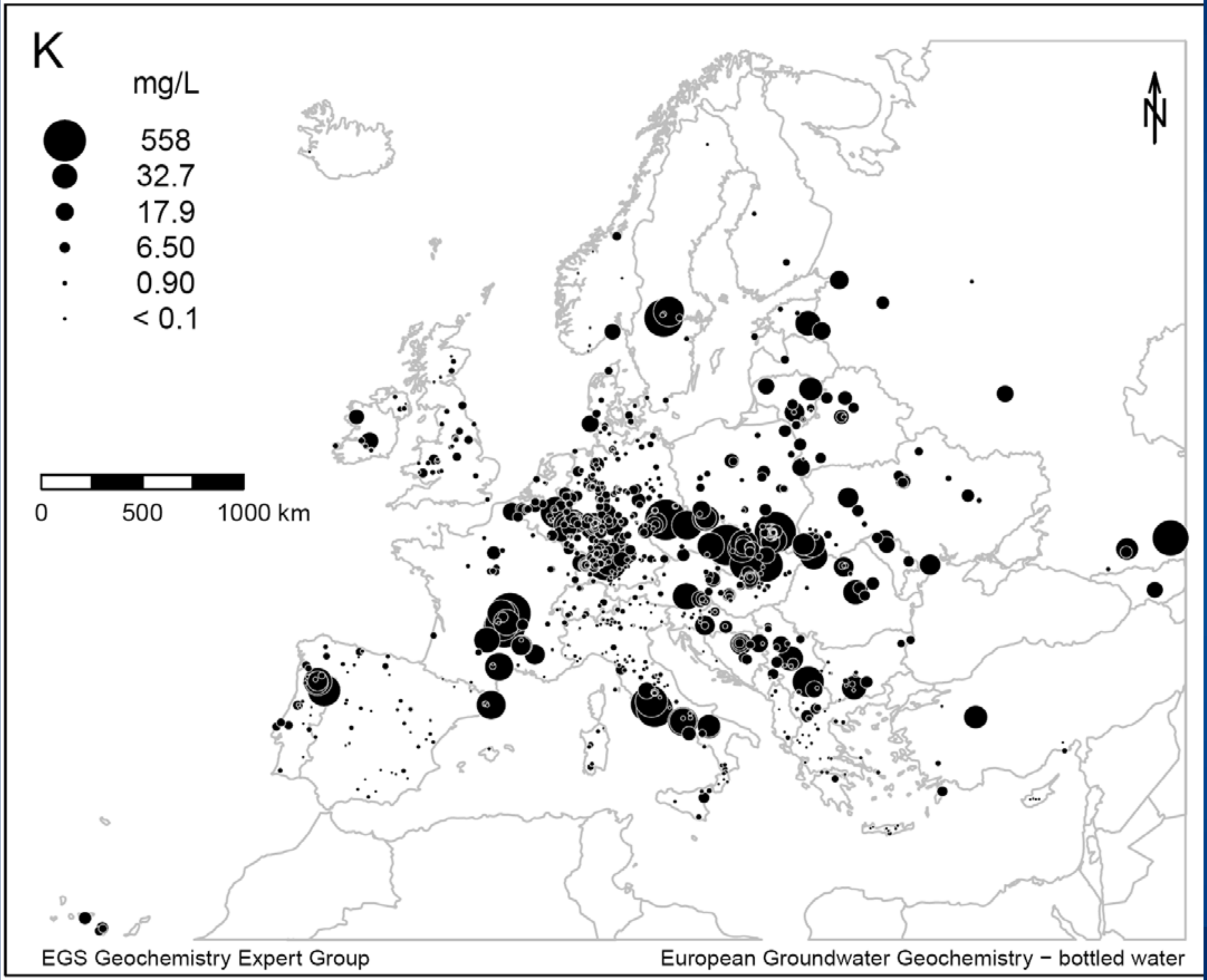


# Potassium (K):

Distribution patterns similar to Li

(Reimann & Birke, 2010, p.127)

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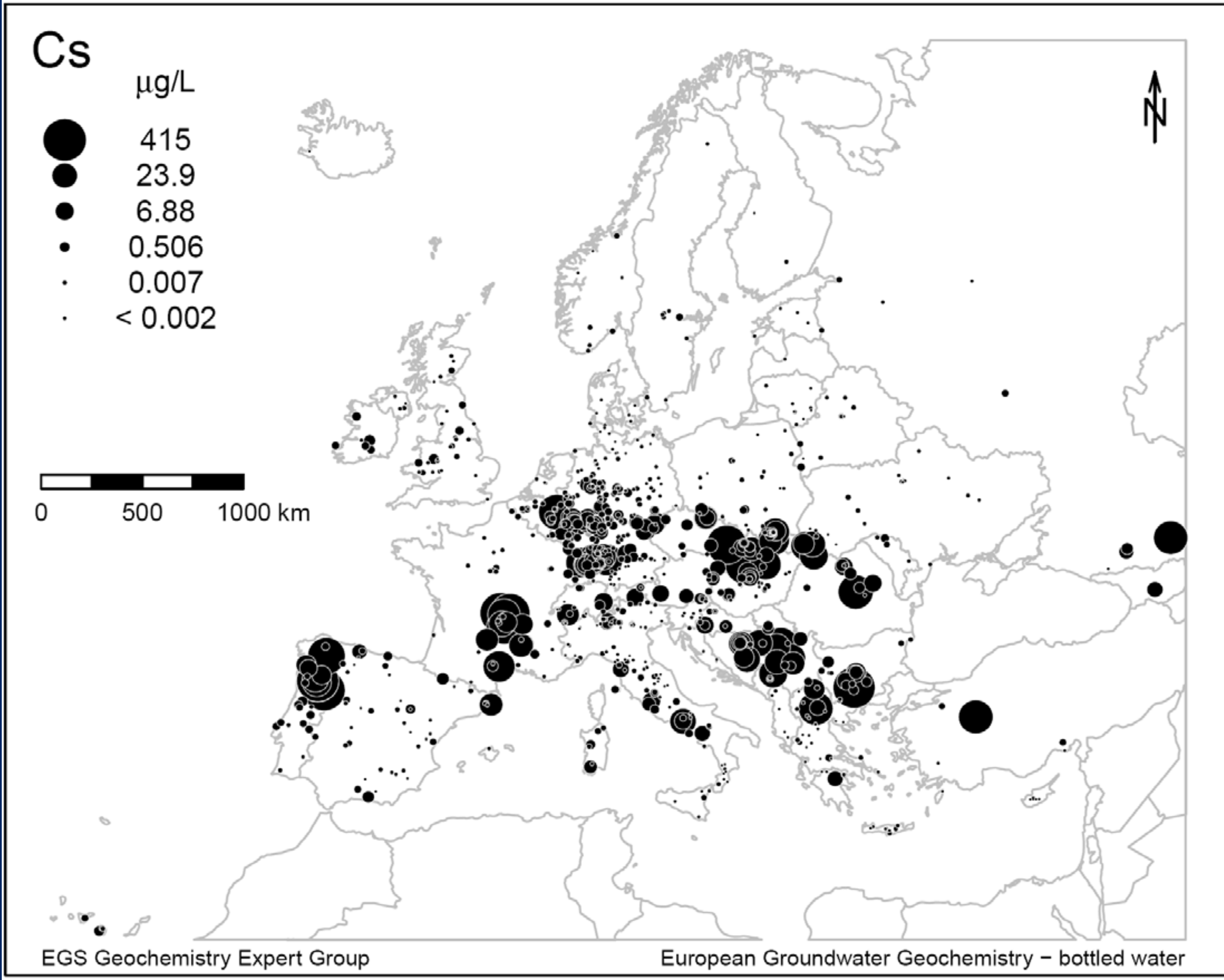
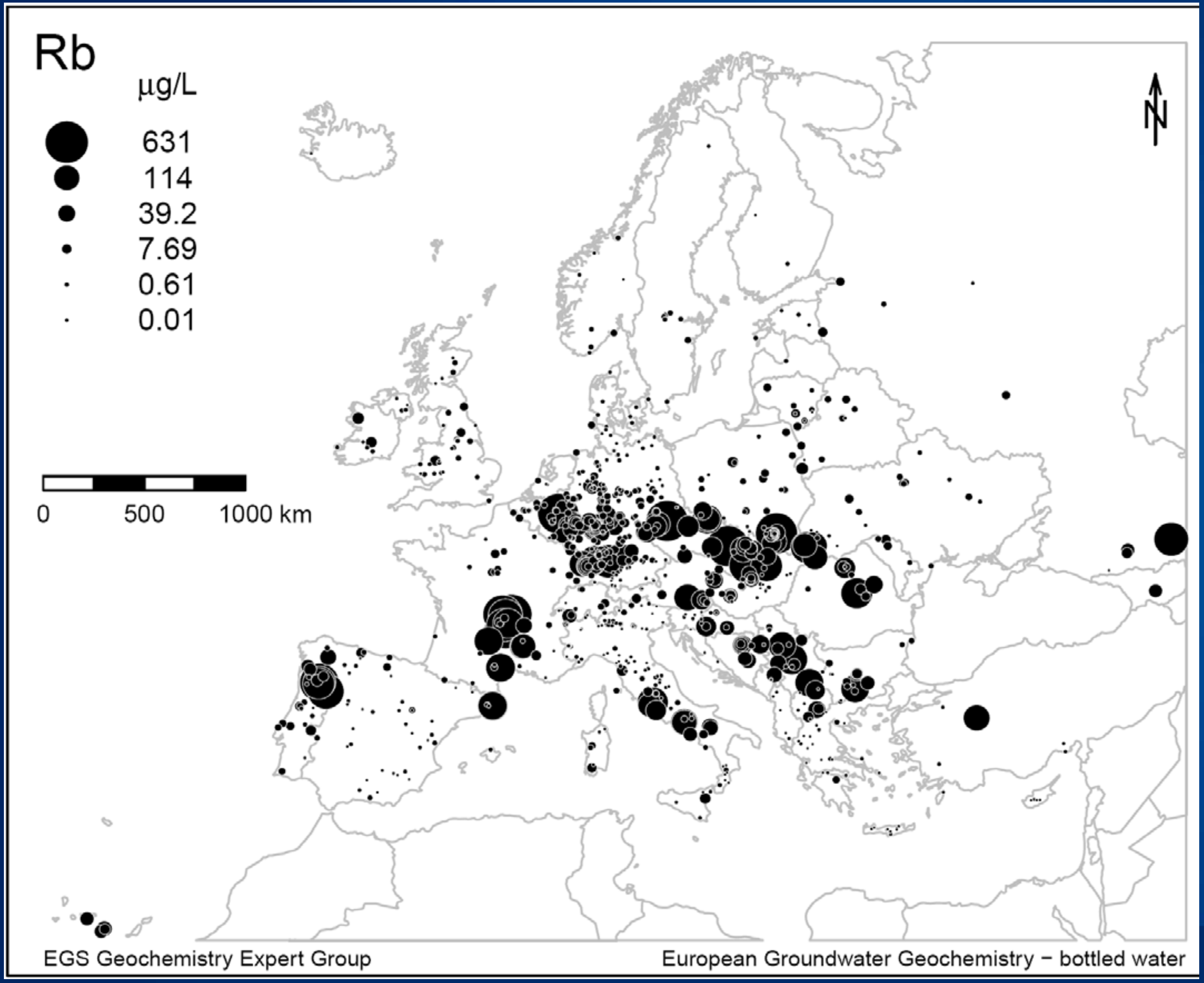



# Caesium (Cs):

Distribution patterns similar to Li and K

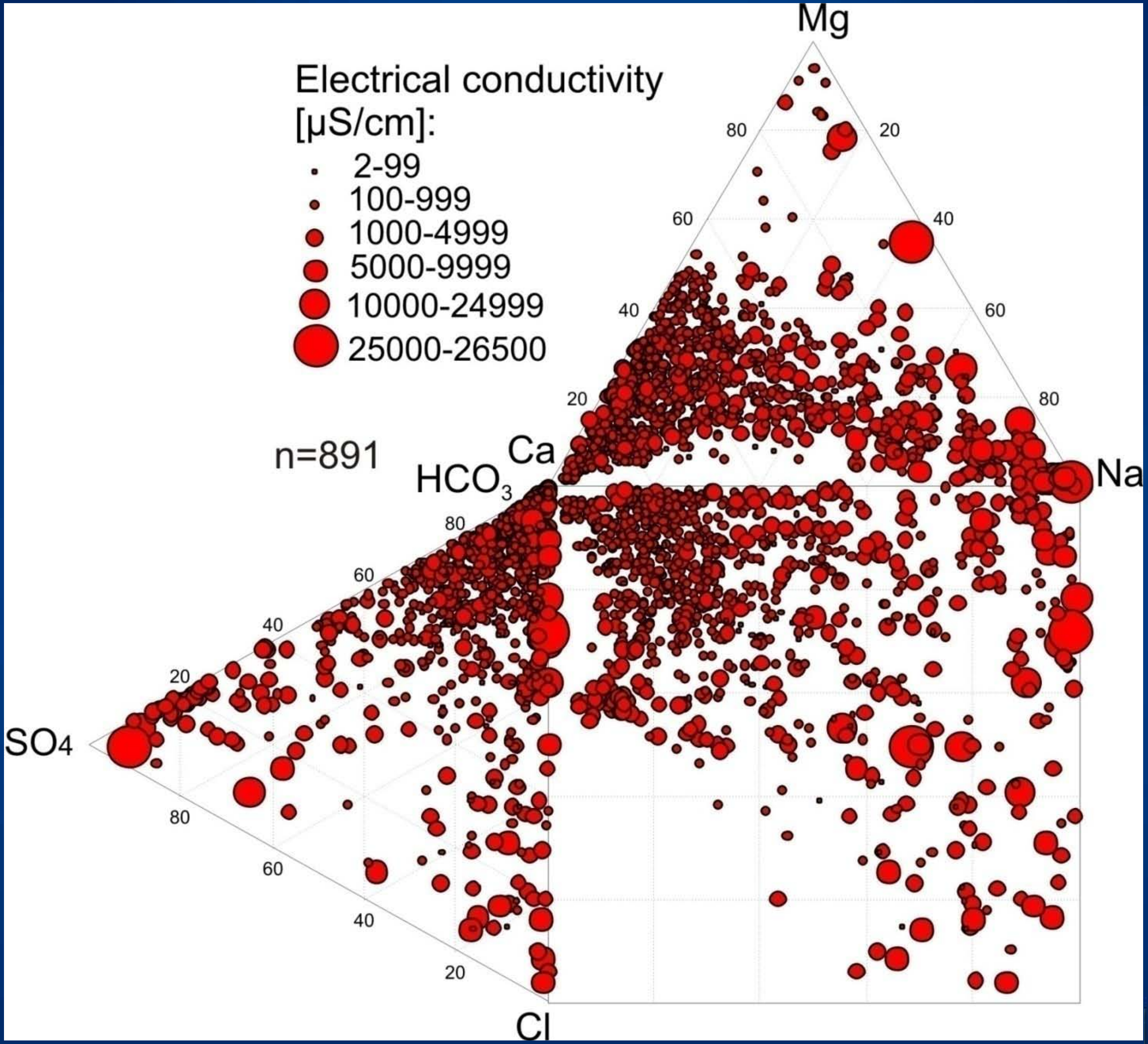
(Reimann & Birke, 2010, p.101)




# Rubidium (Rb):

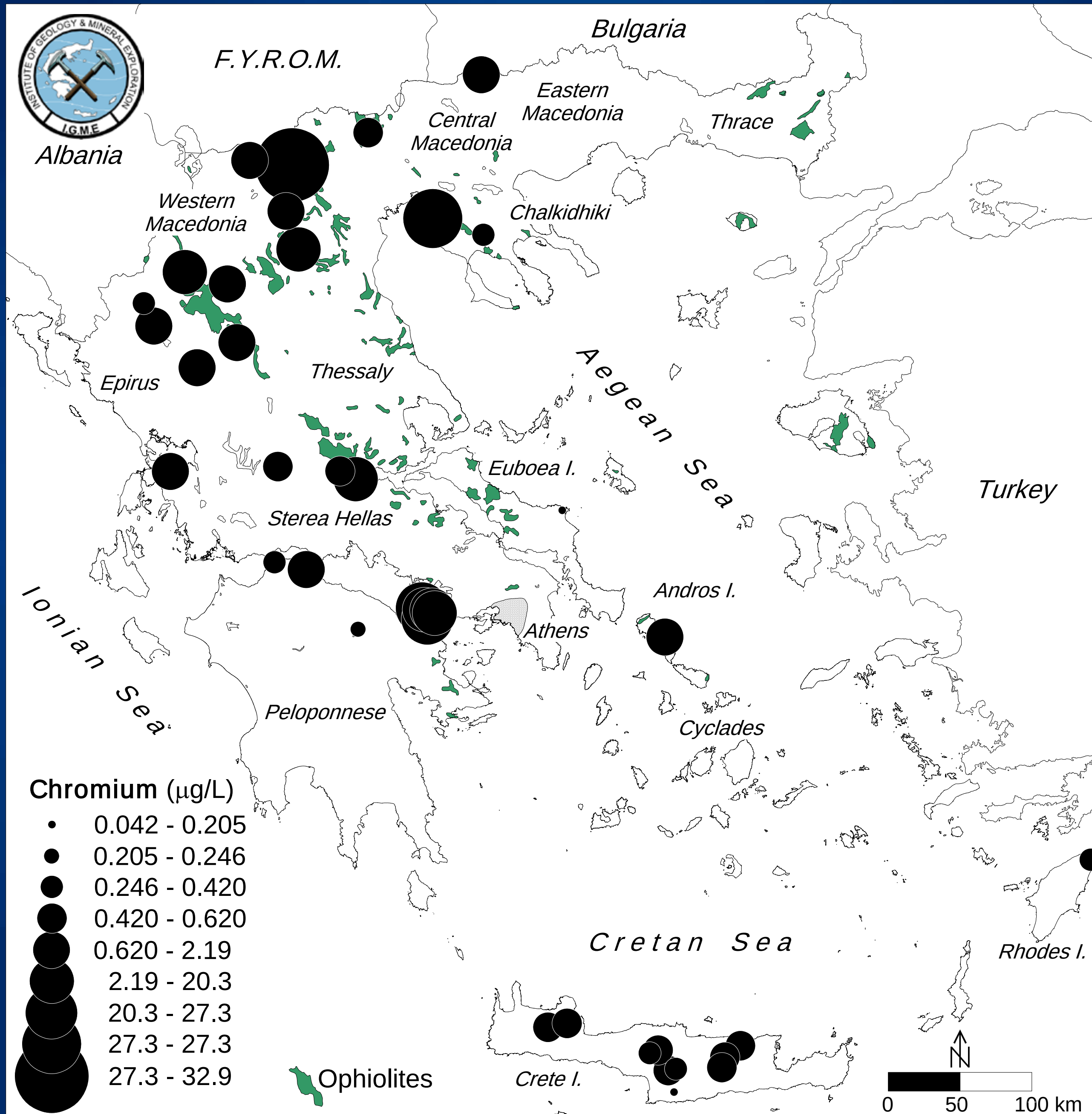
Distribution patterns similar to Li, K and Cs

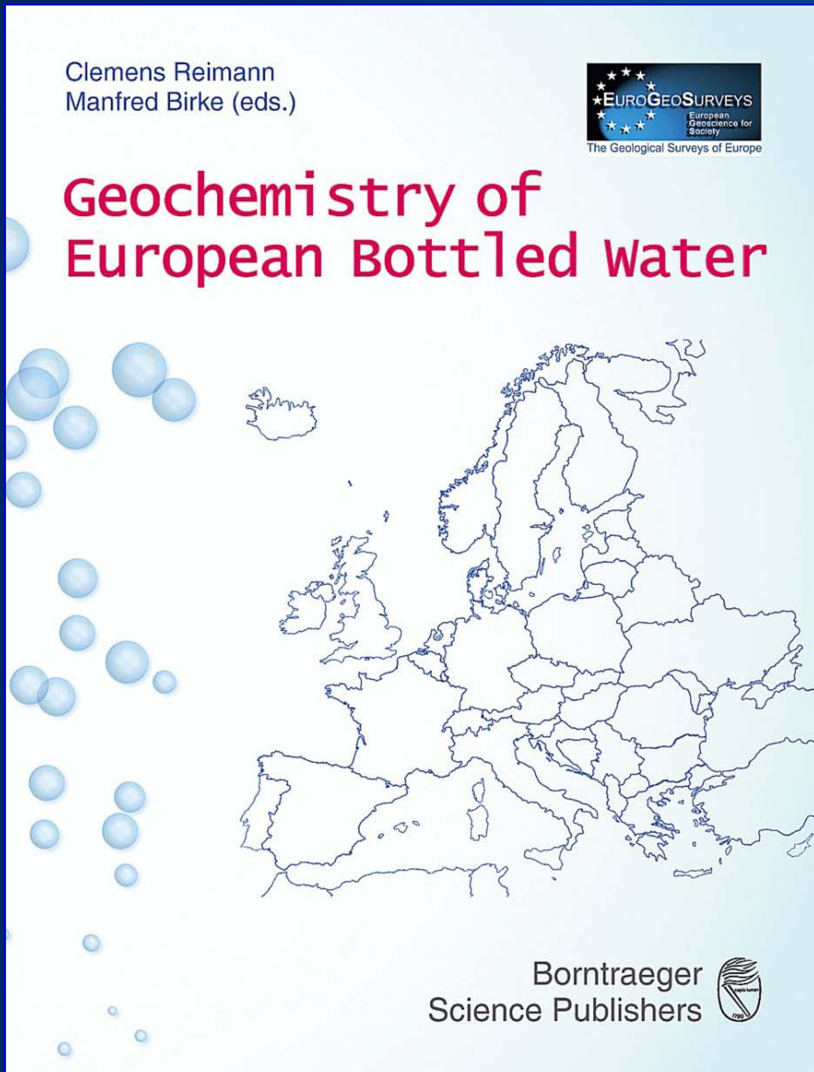
(Reimann & Birke, 2010, p.159)




(Reimann & Birke, 2010, Fig. 7, p.16)

# Distribution of chromium (Cr) in ground water, Hellas



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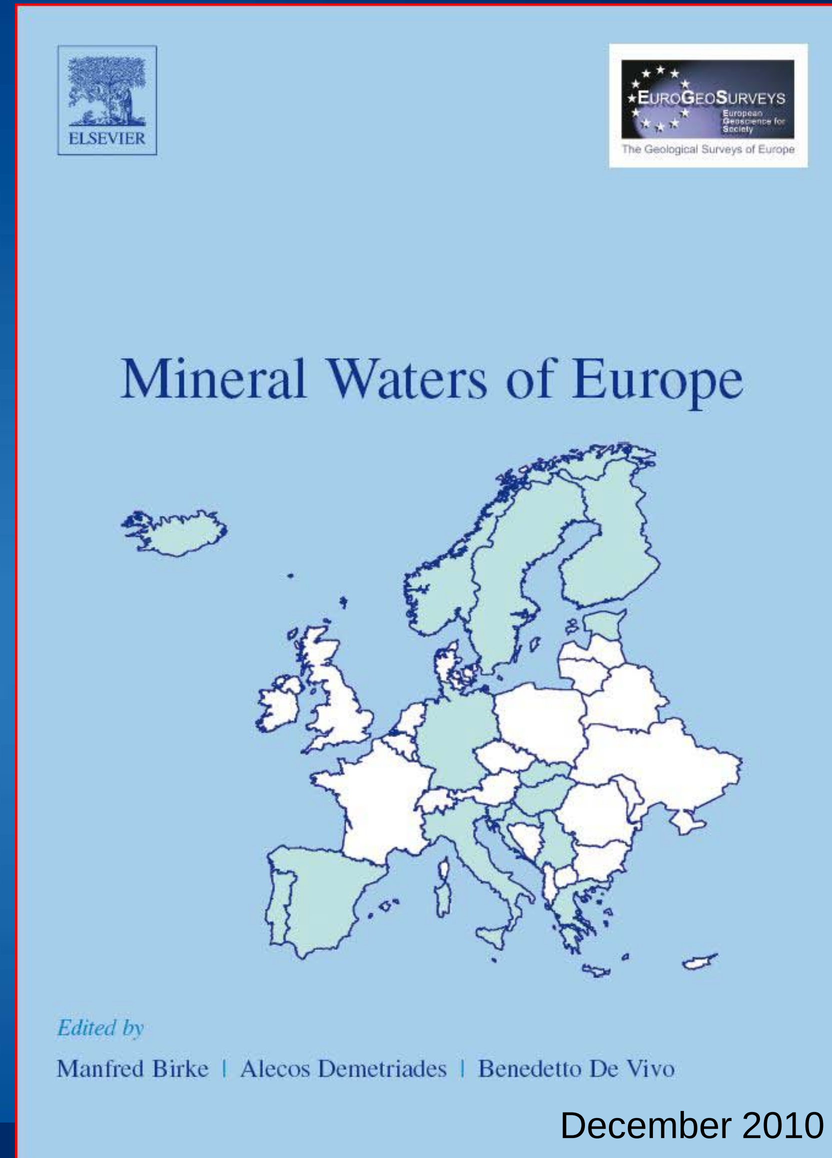


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# Conclusions (1/5)

- Bottle materials contaminate bottle water. Glass contaminates water with respect to Cu, Pb, Al, Zn, Ti, Hf, Th, La, Pr, Fe, Zn, Nd, Sn, Cr, Tb, Ag, Er, Gd, Bi, Sm, Y, Lu, Yb, Tm, Nb and Cu (also some glass bottles contaminate water with Sb. Green glass contaminates bottle water with Cr (Fe, Zr). Hence, PET bottle results were used, since the only problem is contamination of bottle water by Sb.

- Consequently, the Sb distribution map cannot be plotted.

- Some brands of “natural bottled water” are enriched in Ag, B, Be, Br, Cl, Cs, F, Ge, I, K, Li, Na, Rb, Sr, Te, Tl & Zr, and are clearly not representative of “normal” shallow ground water, and the high values are typical of “mineral water” from deep aquifers.

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# Conclusions (2/5)

- For most elements, the bottled water data set provides a realistic picture of their median value and variation in (ground)water at the European scale.

- Natural variation is enormous for most elements, usually 3-5 and for a few up to 7 orders of magnitude were observed.

- When discussing water quality, natural variation must be documented first; the present focus on ”pollution” is misleading in view of the observed high background variation.

- It may be necessary to study more seriously the problems that are related to deficiency in ground water, since water is an important source of elements, such as F, I, Se, Ba.

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# Conclusions (3/5)

**Geological characteristics which are visible on the maps include:**

- **ophiolites (Cr, V)**
- **Alkaline volcanics, and generally areas of active volcanicity (Al, As, Be, F, K, Mn, Mo, P, Rb, Se, Si, Tl, V)**
- **Hercynian granites (Al, B, Be, Cs, F, Ge, K, La, Li, Rb, Si, Sn, tAlk, Th, Ti, Zr)**
- **Deep structures (Sr)**
- **Deep sedimentary basins (B, Ba, Br, Cl, I, K, Li, Mg, Na)**

# Conclusions (4/5)

- Using bottled water as a ‘proxy’ for ground water was more successful than initially anticipated.

- ”*Low density sampling and mapping*”, as developed during the last 15-20 years for detecting large-scale geochemical processes at the Earth’s surface, can also be applied to ground water.

- Geochemical mapping at the continental scale permits cost-effective selection of scale and location of monitoring sites.

- To obtain a fully harmonised data set at the European scale high-quality measurements in a single laboratory are required.

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# Conclusions (5/5)

- **Bottled water standards: The majority of bottled water brands fulfil the requirements of European Union legislation for mineral (and drinking) water. However, for some determinands, a few brands of bottled water exceed the potable water standards, *e.g.*, the maximum values observed for Al, As, Ba, $F^-$, Mn, Ni, $NO_2^-$, $NO_3^-$, Se and U.**

- **Comparison of bottled water analytical results of the EuroGeoSurveys project with those displayed on bottle labels show a fairly good linear correlation, suggesting chemical stability of source aquifers over time, a necessary prerequisite of European legislation for bottled mineral water.**

***Finally, The new bottled-water atlas provides a valuable addition to the "Geochemical Atlas of Europe" series.***

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# The EGG atlas project: 38 European countries...

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# References

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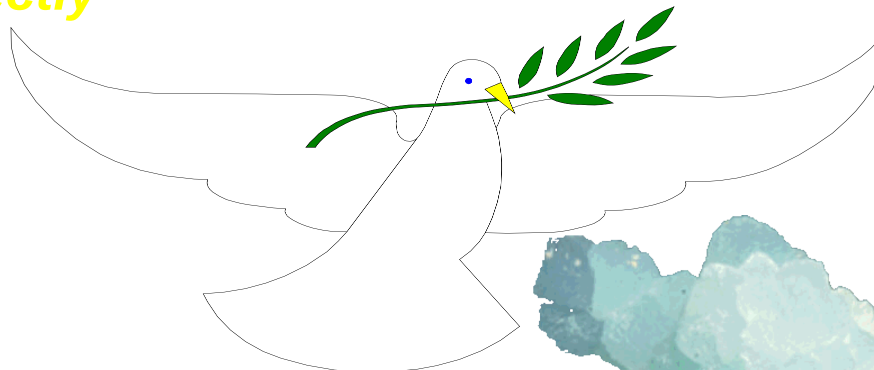

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Nanjing, P.R. China

Presentation on Thursday, 29th March 2012

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