Bottled drinking water: water contamination from bottle materials (glass, hard PET, soft PET), the influence of colour and acidification

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Abstract

A test comparing concentrations of 57 chemical elements (Ag, Al, As, B, Ba, Be, Bi, Ca, Cd, Ce, Co, Cr, Cs, Cu, Dy, Er, Eu, Fe, Ga, Gd, Ge, Hf, Ho, I, K, La, Li, Lu, Mg, Mn, Mo, Na, Nb, Nd, Ni, Pb, Pr, Rb, Sb, Se, Sm, Sn, Sr, Ta, Tb, Te, Th, Ti, Tl, Tm, U, V, W, Y, Yb, Zn and Zr) determined by inductively coupled plasma quadrupole mass spectrometry (ICP-QMS) in 294 samples of the same bottled water (predominantly mineral water) sold in the European Union in glass and PET bottles demonstrates significant (Wilcoxon rank sum test, α=0.05) differences in median concentrations for Sb, Ce, Pb, Al, Zr, Ti, Th, La, Pr, Fe, Zn, Nd, Sn, Cr, Tb, Er, Gd, Bi, Sm, Y, Lu, Dy, Yb, Tm, Nb and Cu. Antimony has a 21x higher median value in bottled water when sold in PET-bottles (0.33 vs. 0.016 µg/L). Glass contaminates the water with Ce (19x higher than in PET bottles), Pb (14 x), Al (7 x), Zr (7 x), Ti, Th (5 x), La (5 x), Pr, Fe, Zn, Nd, Sn, Cr, Tb (2 x), Er, Gd, Bi, Sm, Y, Lu, Yb, Tm, Nb and Cu (1.4x). Testing an additional 136 bottles of the same water sold in green and clear glass bottles demonstrates an important influence of colour, the water sold in green glass shows significantly higher concentrations in Cr (7.3 x, 1.0 vs. 0.14 µg/L), Th (1.9 x), La, Zr, Nd, Ce (1.6 x), Pr, Nb, Ti, Fe (1.3 x), Co (1.3 x) and Er (1.1 x).

One hundred and twenty-six bottles of 3 different materials (glass, hard PET and soft PET) in 5 principal colours (clear, light and dark green and blue, brown) were subsequently washed and then filled with high purity water (18.2 MegaΩ cm). A portion of the bottles where left at the original average pH of the water (pH 6.5) while the remaining bottles were acidified to pH 3.5 with HNO_3. Concentrations of the same 57 elements as above were determined after 1, 2, 3, 4, 5, 15, 30, 56, 80 and 150 days of leaching. Results substantiate the observations from the direct comparison of the same water sold in different bottle types (colour). For most elements leaching is enhanced at pH 3.5, and dark coloured bottles leach more than clear bottles, independent of bottle material. Values are still on the increase at the end of the test at 150 days. At that date the leachates showed a maximum concentration of 0.45 µ g/L Sb, 0.3 µ g/L Ce, 0.61 µ g/L Pb, 68 µg/L Al and 0.06 µg/L Cr (all in glass at pH 3.5). None of the leachates approaches the maximum concentrations for drinking water as defined in European jurisdiction.

Keywords: mineral water, bottled water, leaching, glass, PET, antimony, lead, cerium

1. Introduction

The European Water Framework Directive (EU directive 2000/60/EC) requires that the quality of groundwater is documented at the European scale. Though many national surveys of groundwater quality exist (e.g. Edmunds et al., 1989; Lahermo et al., 1990; Aastrup et al., 1995; Rapant et al., 1996; Frengstad et al., 2000) a directly comparable survey of European groundwater quality is missing. Due to many practical reasons it appears almost impossible to undertake a survey of groundwater quality at the European scale. It was in this situation that the EuroGeoSurveys (EGS, 2010) Geochemistry Expert Group developed the idea that instead of going out and sampling countless wells it might be possible to just buy groundwater at the European scale in the supermarket. There exist almost 2000 registered wells for producing bottled water in Europe (see http://eca.europa.eu/food/food/labellingnutrition/water/mw_eulist_en.pdf - last accessed: 05.03.2010). By analysing the majority of these bottled waters, it should at least be possible to show the overall variation of the analyzed elements in groundwater at the European scale and detect some of the main processes that determine water quality at that scale.

However, due to the low concentrations of most elements in natural water, water sampling and analysis requires great care in order to avoid contamination of the samples during sampling, storage, or analysis (e.g., Nriagu et al., 1993). Many studies have demonstrated that water samples can be severely contaminated by the storage bottles (e.g., Lloyd and Heathcote, 1985; Hall, 1998; Reimann et al., 2007), and often extreme cleansing procedures are suggested for sample bottles (e.g., Ross, 1984). The use of FEP (fluorinated ethylene propylene) or PFA (perfluoroalkoxy polymer) bottles is often suggested for scientific water sampling programs; however, these materials are prohibitively expensive costing ca. $ 25-35 per bottle.

Bottled drinking water is increasingly replacing tap water as the main intake of drinking water in several European countries (e.g., Eisenbach, 2004). It is sold in cheap plastic or glass bottles and is often stored in these bottles under unpredictable conditions for several months before consumption. Prior to using bottled water as a proxy for European groundwater quality the question of what effect the bottle materials have on bottled water quality needs to be addressed. Misund et al. (1999) reported multi-element (66 elements) concentrations in 56 European bottled mineral waters. Their study was not designed to detect contamination from bottle material, however these authors found clear indications of contamination of the water by the bottle materials, e.g., with Pb and Zr from glass bottles. At the same time this study indicated that large scale regional features will emerge when using mineral water for geochemical mapping. More recently it has been convincingly demonstrated that glass bottles will contaminate bottled water with Pb while many polyethylene terephthalate (PET) bottles contaminate the water with Sb and that the concentrations increased with storage time (Shotyk et al., 2006; Shotyk and Krachler, 2007a,b; Krachler and Shotyk, 2009). Keresztes et al. (2009) report Sb concentrations of 210 to 290 mg/kg in PET from Hungarian mineral water bottles, and significant leaching of Sb to the water depending on storage conditions and bottle volume.

Diantimony trioxide is the single most important catalyst in the production of PET (EU, 2008). Other metals are also used in the manufacture of PET for a variety of purposes (catalyst, stabilizer, colour). All 88 naturally occurring chemical elements appear in the raw materials used for glass production. For example, Pb is a common natural trace element in feldspars, one of the key raw materials for glass production. Some other elements may be added on purpose during glass production to determine colour (Fe, Cr for green colours, Co for blue colours). Water is a powerful solvent and contamination of water samples from storage containers is almost unavoidable – at least at reasonable cost. However, the contamination occurs on top of a large natural variation of all elements in the waters sold in these bottles and the relative importance of contamination in relation to natural variation needs to be established for all elements if using bottled water as a proxy for groundwater quality. Different materials (glass, soft PET, hard PET) and different bottle colours may all provide different results. It must also be assumed that the pH of the water (beverage) stored in these bottles will play an important role for the release of elements from the container walls into the fluid.

In a first step this study thus compares analytical results for 57 elements in 294 pairs of water samples sold in PET as well as in glass bottles on the European market. Significantly higher concentrations of an element in the water in a bottle type can be assumed to originate from leaching of these elements from either glass or PET into the water sold in these bottles.

Because colour could have an important effect on the leaching results, 136 water samples that were sold in both, clear and green glass bottles were analyzed for the same 57 elements and the results were directly compared.

Finally, this study reports concentrations of the same 57 elements in 126 European bottled waters, bought in supermarkets all over Europe in 5 different bottle colours (shades). Following analyses of the original water, the bottles were rinsed and subsequently filled with high purity (demineralised) water (pH 6.5). At a number of different time intervals (1, 2, 3, 4, 5, 8, 15, 38, 56, 80 and 150 days) the demineralised water stored in the bottles was analyzed for 57 elements. A selection of samples was acidified to pH 3.5 to study the effect of pH on the leaching results.

By comparing results for the same water stored and sold in different bottle materials and the original water results to the leaching results with demineralised water and acidified demineralised water this paper allows judging the relative importance of leaching from bottle materials on the element concentrations observed in natural bottled waters.

2. Materials and methods

Using the network of colleagues from EuroGeoSurveys 1785 bottles of water sold all over Europe were collected at the Federal Institute for Geosciences and Natural Resources (BGR) in Berlin. Whenever possible the same water was bought in different bottle types (glass, hard PET, soft PET), in the still and carbonated variety and in different colours, with clear, lightgreen, darkgreen, lightblue, darkblue as the main colours but also including a number of rare brown, red, or pink bottles (summarized as "brown" here). Prior to analysis all water bottles were stored refrigerated at the laboratory of BGR. The sample set contained 294 cases where water from the same well was sold in glass as well as in PET bottles. Furthermore there existed 136 cases where water from the same well was sold in clear as well as in green glass bottles (the majority from Germany).

For the leaching test 126 bottles of mineral water purchased in supermarkets all over Europe (77 Germany, 49 rest of Europe) were used. The waters were bottled in glass, hard PET and soft PET containers and represent 92 wells from 87 locations as well as the above 5 colours. Following the analysis of the original water these bottles were thoroughly rinsed with high purity (demineralised) water and then filled with demineralised water under clean room conditions. During the time of the leaching test (150 days) the water samples were kept refrigerated (+2 ^{O}C).

2.1. Laboratories and instrumentation

All work was carried out using clean room procedures in a class 10000 clean room at the laboratory of BGR. The samples were analyzed by inductively coupled plasma quadrupole mass spectrometry (ICP-QMS) using an Agilent 7500ce instrument. The instrument is equipped with a standard peristaltic pump, a MicroMist concentric nebulizer, a Peltier-cooled spray chamber, the Plasma Forward Power and the Shield Torch System. The methods used are in accordance with the German norms DIN 38406-E29 (ICP-MS).

2.2. Laboratory procedures – leaching test

The 126 bottles selected for the leaching test were completely emptied following analysis of the waters and rinsed 3 times with high purity (demineralised) water. On the next day they were filled with demineralised water (SERALPUR-90, 18.2 MegaΩ cm) and to a selection of bottles 25µL 69 % HNO_3 (Roth Suprapure, density 1.41 kg/L) was added/L to acidify the water to pH 3.5. In addition several of the mineral water bottles were filled only with demineralised water from the SERALPUR-90 system (18.2 MegaΩ) at pH 6.5. Prior to analysis, starting the next day, the mineral water bottles were vigorously shaken 10 times.

Subsequently the water stored in the bottles was analyzed after 2, 3, 4, 5, 15, 30, 56, 80 and 150 days.

2.3. Detection limits

Sample blanks were measured repeatedly over the whole time span of the project and used to determine the detection limits as used in this publication. The instrument detection limit (IDL, Table 1) was determined using 3 times the standard deviation of the sample blank determinations. For the leaching test the IDL was used as the reporting limit because all results were plotted in the form of time trends and it was thus possible to directly judge whether the results represented noise near the detection limit or clear time trends. The IDL was also used for the direct comparison of glass and PET bottles and of clear and green glass bottles. The conservative reported detection limit (RDL) was calculated at 10 times the standard deviation of the sample blank (Table 1) and is used when presenting the analytical results of the original bottled waters.

2.4. Quality control

For quality control purposes the river water reference material SLRS-4 from the National Research Council Canada, was used. In addition, an internal laboratory mineral water standard (MinWat) was used. This sample was repeatedly measured for the entire time the samples were analyzed to have control on the repeatability of all values. The internal laboratory standard was stored in a 5 L HDPE canister. Standard SLRS-4 was inserted 82 times at regular intervals during sample analyses. Results for SLRS-4 and the internal MinWat standard are presented in Table 1 and demonstrate that for the majority of elements the results are well in agreement with the certified values.

When analyzing as many elements as in this study a general problem is that there exist no suitable reference materials that are certified for all these elements. However, for SLRS-4 published values from other investigations exist (see: http://georem.mpch-mainz.gwdg.de/) and are provided in Table 1. In the vast majority of cases the median values from this study are very close to the certified and/or published results.

The laboratory standard MinWat was inserted 83 times and used to assess precision of the measurements at the concentrations of this standard. Comparison of the coefficient of variation obtained for SLRS-4 and MinWat shows that for a number of elements a high CV (>25 %) occurs at low concentrations (e.g. Ag, Ga, Hf, Nb, Sn, Te, Tl, W) but reaches better values (CV<10%) at higher concentrations (e.g. Ag, Ga, Te). X-Charts plotted for all elements, indicate in addition a few elements where there appears to be a general problem with reproducibility at the very low concentrations near the IDL of these elements in the standards: Hf, Nb, Sn, Ta and W. All results for these few elements need to be treated with care in that concentration range (up to the RDL). The element Sc was measured but is not reported here due to an interference with Si, causing values to be too high. This problem can only be overcome on a high resolution (HR-) ICP-MS. Special care was taken for the analysis of the Rare Earth Elements (REEs) and I, which provide a number of problems in routine ICP-MS analyses (REEs: interferences; I: sample stabilization, memory effects).

In addition, duplicates were analyzed at regular intervals and a number of quintuple and 10-fold determinations were also carried out. Furthermore several series of sample blanks were measured over the entire time span of the project.

2.5. Data Analysis

Exploratory data analysis techniques were used for data analysis (Tukey, 1977). All graphics and statistical calculations for the tables were prepared in R (Reimann et al., 2008). Note that these data are compositional (closed) data (Aitichison, 1986) - classical statistics are not really suitable for such data. Filzmoser et al. (2009) have recently demonstrated that especially the calculation of the mean and even more so the standard deviation of the original or log-transformed data and in consequence using statistical tests that build around these values are fraught with problems. Thus more modern techniques, as outlined in Reimann et al. (2008) are used here and no values for "mean and standard deviation" are provided. Instead the median and the median absolute deviation of the isometric logratio- (ilr-, see Egozcue et al., 2003) transformed data (MAD.ilr – see Filzmoser et al., 2009) are provided. The MAD.ilr is a measure of stability or precision; the orders of magnitude of variation, the powers, provide one of the most intuitive measures of variation.

3. Results

3.1. Direct comparison of the same water sold in glass and PET-bottles

Table 2 compares the results of the 294 pairs of water samples sold in glass and in PET bottles (median glass, median PET, the results of a Wilcoxon rank sum test, α=0.05, and the ratio glass/PET). In addition the results were directly compared in a series of XY diagrams and boxplots (Figs 1-3 provide some examples).

The water sold in PET bottles shows a significantly higher concentration of Sb than the same water sold in glass bottles – the median Sb-concentration for the water sold in PET-bottles is higher by a factor of 21 than the median for the same water sold in glass bottles (Table 2, Fig. 1). The XY-diagram, however, indicates that there exist quite a number of PET bottles where leaching is only a minor problem. Shotyk and Krachler (2007a) also describe differences in the reactivities of different bottles. Antimony is the only element where the waters stored in PET bottles show higher concentrations than the same waters sold in glass bottles (Table 2). The analytical values from the water sold in PET-bottles can – with the exception of Sb – thus be taken to represent "background" variation for bottled water.

Glass bottles, however, leach a much longer list of elements into the water, with Ce (19x higher median in glass than in PET), Pb (14x) and Al (7x) topping the list (Table 2). Furthermore Zr, Ti, Hf, Th, La, Pr, Fe, Zn, Nd, Sn and Cr show 7 to 2.4 times higher median values in the water stored in glass bottles than in the same water sold in PET bottles. Figure 2 shows this effect for the elements Pb and Th. Again it is apparent that different bottles can leach different amounts.

In addition there is a long list of elements where no significant effect of bottle material is observed (see Fig. 3 for an example): As, B, Ba, Be, Ca, Cd, Co, Cs, Eu, Ga, Ge, I, K, Li, Mg, Mn, Mo, Na, Ni, Rb, Se, Sr, Ta, Te, Tl, U, V and W (Table 2). This could be due to:
(1) the fact that there is no leaching of these elements from the containers,
(2) the bottled waters have such high concentrations of these elements that an additional leaching from one of the two bottle types remains invisible and
(3) the unlikely case that these elements leach from both bottle materials to the same extent.

3.2. Influence of colour – direct comparison of the same water sold in clear and in green glass bottles

European mineral waters are sold in bottles of principally 5 different colours (shades), with clear and green as the prevailing colours. For a direct comparison of the same water sold in bottles of different colours it was only possible to obtain a sufficient number of clear and green glass bottles (136 pairs) containing water from the same well. Median values were computed for the water stored in green and for the water stored in clear glass bottles. These were compared using a Wilcoxon rank sum test (α=0.05, Table 3). In addition the same plots as above were used to compare the waters sold in clear and green bottles. For the majority of elements there was no visible difference between the values found in clear glass compared to those from water sold in green glass. However, for Cr (Fig. 4), Th, La, Zr, Nd, Ce, Pr, Nb, Ti, Fe, and Co the Wilcoxon rank sum test indicates a significant difference (Table 3). For Cr the waters stored in the green glass bottles show a 7x higher median value than the same waters sold in clear glass bottles. For all other elements the effect is much smaller. Tungsten (Fig. 4) is the only element that returned somewhat higher concentrations from the clear glass bottles, however, W determinations are plagued by a serious precision problem in the concentration range of the majority of the mineral waters (see above), thus this observation may be an analytical artifact.

3.3, Bottle leaching

126 bottles representing the different bottle materials and variations of colours on the European market were selected for the bottle leaching test. Table 4 presents analytical results for the 126 original mineral waters sold in these bottles. The maximum admissible concentrations in mineral water sold on the European market according to EU Directive 2003/40/EC Mineral Water are also provided for comparison.

In terms of the leaching results presented here it must be noted that it was not possible to obtain factory new bottles for this test. More leaching may appear when factory new bottles are filled for the first time (most soft PET bottles are for one time use only). This test will still provide a realistic picture for leaching from return bottles and can be used to prove that the observed differences are really caused by the affected elements leaching from the container materials.

Due to the large number and variety of bottles tested, in addition to 57 elements analyzed, data analysis and presentation of the leaching results turned out to be challenging. A simple tabulation of the data according to material, colour, time and pH was practically unreadable – and, due to the different numbers of bottles tested for each case, not even reliably interpretable. Table 5 summarizes the leaching results for the 3 bottle materials (all colours included) and differentiates between the non-acidified (pH=6.5) and the acidified (pH=3.5) samples. The IDL and RDL and the original values for the water sold in these bottles (Table 4) can be used to judge the relative importance of bottle leaching on the values reported for the bottled water sold in these containers.

Furthermore a series of two different plots were used to study the data. To examine the time trends for each element 3 plots (one for each bottle material: glass, hard PET and soft PET) were combined on one page, where the time trends (1-150 days) were plotted with two different line styles for the non-acidified (dashed) and acidified (solid) water. Figure 5 shows this diagram for Pb, Figure 6 for Sb. To directly provide an impression of the significance of leaching of the element from the bottle material into the water stored in these bottles the variation in concentration observed in the original bottled water was added to the plots in the form of two boxplots – one for water sold in glass bottles and one for water sold in PET bottles. The dotted lines along the x-axis indicate the detection limit and multiples (10, 100, 1000x) of the detection limit. Note that the leaching test results were reported down to the IDL, while the RDL is used for the original samples.

All glass bottles clearly leach Pb into the water stored in the bottles (Fig. 5). The boxplots indicate that water sold in glass and in PET bottles are incomparable in terms of the observed Pb concentrations. Colour of the bottle does not determine how much Pb will leach. Acidification has a highly significant effect on leaching (Fig. 5); Pb concentrations in the acidified waters are almost an order of magnitude higher than in the non-acidified waters. Concentrations increase with time and most bottles had not reached a concentration plateau; the plots indicate that the Pb-values are still on the increase after 150 days. Especially for the non-acidified water quite drastic increases in Pb-concentrations become visible for some bottles towards the end of the test. In contrast, for hard PET no significant leaching of Pb from the bottle material is visible; Pb concentrations fluctuate very close to IDL (0.001 µg/L - Fig. 5, Table 5). For soft PET, again most values fluctuate around the detection limit, there are, however, single bottles that show Pb concentrations reaching the 75th percentile value of the original water sold in these bottles.

The diagram for Sb looks quite different. It is apparent that Sb can leach from all bottle materials, even glass, and not just PET (Fig. 6). Still, the boxplots indicate that water sold in glass and in PET bottles is incomparable in terms of Sb concentrations. The leaching effect is not as pH dependent as for Pb (see Fig. 5), and clearly most pronounced for soft PET, where the majority of bottles reach values in the range of 10 to 100 times the detection limit towards the end of the test (Fig. 6) and are well above the 75th percentile of the original water sold in glass bottles (Table 4). At the end of the 150 day period the values have not reached a plateau, they are still on the increase. For hard PET bottles only the dark blue variety shows significant leaching of Sb, but note that only a limited number of hard PET bottles were tested. The values observed for the glass bottles are usually much lower than those from the PET bottles (range DL to 10x DL) but the water in some single glass bottles reaches as high Sb values as those observed in the PET bottles. The highest Sb value found at the end of the test (150 days) was observed in a dark green glass bottle (Fig. 6). This is a general message from these graphics: there is no overall trend that is valid for all bottles, there are often only some few bottles that leach considerably, while the majority shows minimal leaching.

The time trends display considerable noise, which may partly be due to precision problems at the ultra low element levels in the demineralised water near the IDL, but can also be caused by adsorption and desorption reactions of certain elements with the bottle walls or be due to the formation and dissolution of colloids in the water during storage. Several elements (e.g. Hf, Nb, Ta, Ti, Sn, Th, W, and Zr) tend to be unstable in only slightly acidified water samples, this is the reason why they are not certified in most water standards (e.g., Salbu and Steinnes, 1995).

In order to be able to more directly compare the effects of the different bottle materials and colours a second set of diagrams was prepared. Here only the median values of the leaching results were plotted for each bottle colour and each material and a separate diagram was drawn for acidified and non-acidified samples. A boxplot showing the variation of each element in the original waters stored in the bottles was again added to the figure.

Figure 7 shows this plot for the element Al. It is visible at a glance that glass bottles appear to leach highly significant amounts of Al into the water in the bottles at pH 3.5 and usually only small amounts of Al at pH 6.5. However, it must also be noted that the caps of many glass bottles are made of Al and that opening and closing of the bottles several times could lead to Al dust from the caps dissolving over time in the water.

Figure 8 shows the same plot for the element Cr. Here the effect of dark green glass on the Cr-concentrations observed in the leachate is clearly visible. Again the effect depends strongly on the pH of the sample and is most pronounced for the acidified samples (pH 3.5) while in the non-acidified case very little is leached from the bottles.

Finally, Figure 9 shows this plot for K. Here the concentration of K in the original mineral water is so much higher that leaching from the bottles has no additional effect. These diagrams, in combination with Tables 4 and 5, now allow a rapid evaluation of the test results.

4. Discussion

In order to evaluate whether the possible bottle contamination levels summarized in Table 5 are significant for the concentrations reported as "typical" in bottled waters, it is necessary to know the range of concentrations to be expected of that element in a natural water sample, without any effect of contamination. At the concentration levels reported here for many of the elements it is actually a difficult undertaking to establish truly natural minimum concentrations. It is of course possible to compare to the levels of the original bottled water (Table 4) – but these values are plagued by the same (if not worse) contamination issues as identified in the leaching test because the water was originally sold in these bottles. Even if taking water samples for a scientific study directly at source using expensive PFA- or FEP-bottles there always remains a possible contamination issue due to differences of well installations and residence times of the water in the well. In that respect collecting bottled water may actually be an advantage, all samples come from high production wells where contamination from well installations should be small when compared to low production wells. Sampling natural springs as suggested by Shotyk and Krachler (2007b) may be a good alternative for obtaining a first estimate of "true" element levels in natural waters, it neglects, however, that there will be a difference between spring water and water from deep wells. The most practical approach to assess possible contamination of the water samples from the bottle is thus probably the direct comparison of the same water sold in different bottles in combination with a leaching test as provided here. In addition to the directly observable differences between different bottle materials, high concentrations and especially increasing concentrations over time in the leachates will provide clear evidence for leaching of the elements concerned from the bottle material. The comparison will show which elements are seriously affected and the leaching test will demonstrate for which elements one or even all bottle materials will pose a contamination issue when using these waters to indicate "groundwater quality".

Contamination from the bottle materials is not the only issue that needs to be considered in this connection. Elements can also adsorb to the bottle walls and thus be removed from the water. Such effects have, for example, been reported for Hg and Au (e.g., Feldmann, 1974; Lindquist et al., 1996). The reliable analysis of these elements requires the selection of special bottle materials and/or sample conservation procedures at the time when the sample is taken. This effect will lead to too low and/or spurious analytical results in a water sample if the proper precautions are not taken. The same is true if some elements are unstable in solution and tend to fall out (unstable in solution at the pH of bottled water are for example Hf, Nb, Ta, Ti, Sn, Th, W, and Zr) (note the poor precision for most of these elements in Table 1) or to form or adhere to colloids (e.g. Al, Fe). Colloids can sorb large concentrations of trace elements (Horowitz et al., 1996). Unfortunately colloids in a water sample can form and disappear again over time. The leaching test provides indications that such effects must be expected to take place for quite a number of elements.

Fortunately, the concentrations of only few elements appear to be seriously influenced by leaching from the bottle materials. For PET bottles Sb turned out to be the only element of real concern. Although the leaching test demonstrates that Sb can also leach from glass bottles (the highest observed leaching value is from a glass bottle) the median concentration of the waters sold in PET bottles is 21 times higher than for the same water sold in glass bottles. The leaching test demonstrates that Sb is really leaching from the PET bottles, that the effect is almost pH independent, that bottles of all colours leach and that the values are still increasing after 150 days. Less serious leaching is observed for the hard PET bottles than for the soft PET bottles. That may in part be due to the fact that most hard PET bottles are returnable and will be re-used, which should, after time, reduce the leaching from the bottle walls. For the glass bottles some single bottles show serious leaching effects. The results indicate that it will be impossible to establish the true background variation of Sb in European groundwater using commercial bottled water.

Glass bottles leach a much longer list of elements and in the direct comparison glass - PET it is Ce (19 x higher in glass bottles than in PET bottles) that tops the list, closely followed by Pb (14x) and Al (7x). The leaching tests demonstrate that leaching of Al is no issue for hard PET bottles. Some single soft PET bottles can leach significant amounts of Al at pH 3.5. Similar results are visible for Ce and Pb. It is an interesting observation that practically all the determined rare earth elements (REEs) are enriched in the water sold in glass bottles (exception: Eu). The leaching test showed that high levels of Al and Ce are already reached on day 1 in the demineralised water stored in glass bottles and that the further increase over time is comparatively small. In both cases leaching is much stronger at a lower pH. The diagram for Al is surprisingly noisy given the high concentration, an indication that Al-colloids may form and dissolve again in the bottles over time. Leaching of Zr, Ti, Hf and Th, which are also quite enriched in the glass bottles, happens very close to the detection limit. Only some bottles appear to leach significantly and pH always plays an important role (stronger leaching at pH 3.5). The results from the leaching test alone would not have shown the same significance as comparing the original water sold in different bottle materials.

At pH 6.5 the leaching plots indicate very few problems. Significant (in relation to the expected values in the water) amounts of the following few elements may leach to the water stored in the bottles:

Al: darkgreen glass

Cu: darkblue glass
Sb: soft PET, darkblue hard PET
Sn: all glass bottles
Tl: all glass bottles - especially clear glass
W: glass bottles, 1 soft PET bottle (but note poor precision of W).

At pH 3.5 the situation changes considerably. In general many more elements will leach from the bottle materials to the waters in the bottles. Glass is the bottle material displaying the most pronounced leaching effects for a long list of elements (Table 5, maximum leaching results are marked in bold). hard PET is the bottle material leaching least. Soft PET shows effects for some bottles and specific colours. When investigating the effects of colour in detail it turns out that brown bottles (glass and soft PET) leach most, closely followed by dark green glass bottles. It should be noted that a pH of 3.5 is not a realistic value for a bottled water sample, it will, however, be easily reached by some other beverages sold in similar bottles.

In terms of consumption of the bottled water none of the reported values from bottle leaching provides reason for concern, they are all considerably below the maximum acceptable concentration of these elements in drinking water (EU Directive 1998/83/EC Drinking Water) or mineral water (EU Directive 2003/40/EC Mineral Water) as provided in Table 5.

5. Conclusion

The values for elements reported as "bottle contamination" of bottled water in this study are all under the respective drinking water action levels. There are clear differences between the three bottle materials investigated. For hard PET Sb is the only element where contamination of the water stored in these bottles is a real issue. The leaching test reveals that soft PET, which is rapidly developing into the main bottle material in many European countries, is in comparison to hard PET surprisingly "dirty". However, again Sb poses the most serious contamination issue observed. In terms of obtaining an indication of natural background values of Sb in European groundwater, water sold in PET bottles cannot be used. It must be noted that even some glass bottles can leach Sb. The highest Sb value of all observed after 150 days of leaching came from a darkgreen glass bottle.

Many more elements leach from glass than from PET bottles. Comparing the same water sold in PET-bottles to results for water sold in glass bottles Ce, Pb, Al and Zr are the 4 elements that leach most from glass, but Ti, Th, La, Pr, Fe, Zn, Nd, Sn, Cr, Tb, Er, Gd, Bi, Sm, Y, Lu, Yb, Tm, Nb and Cu are all significantly enriched in the glass bottles when compared to the same water sold in PET bottles. The leaching test shows that at a lowered pH of 3.5 leaching from glass increases considerably, often by a factor 10 or more, for most elements. In terms of determining the background variation of Ce, Pb and Al in European groundwater, contamination of samples from glass is so serious that bottled water sold on the European market in glass bottles cannot be used for that purpose.

In terms of colour, dark coloured bottles (especially darkgreen and brown) leach more than clear bottles. This is true for PET and glass. The most significant influence of colour on leaching was observed for Cr from darkgreen bottles. The more rare blue bottles leach Co. Brown bottles often show strongly increased values for Fe and most REEs.

All leaching tests demonstrate large differences between single bottles. There exist bottles where leaching is a major problem and bottles of the same material and colour where leaching is insignificant. The reactivities of the bottles are clearly different, the reasons are not clear. For glass and hard PET alone these differences could be due to the fact that return bottles were used and bottle age is not known. Soft PET bottles, however, are not re-used and show the same effect.

Bottled drinking water is often stored under unpredictable conditions for prolonged times in the water bottles before it is sold and drunk and storage time and conditions will clearly have an influence on how much of an undesirable element leaches into the water. For the elements indicated in this test as leaching from the bottles care is needed in the interpretation of single high values obtained when analyzing bottled water. However, the test demonstrates that for the vast majority of elements bottled water can be used for providing a proxy for “natural” groundwater quality at the European scale.

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Table Captions

Table 1: Analytical results for standard samples analyzed repeatedly over a period of 150 days during the leaching test: SLRS-4 (National Research Council Canada River Water Reference Material for Trace Metals), and the in-house project standard "MinWat". IDL: instrumental detection limit, RDL: reported detection limit, CERT: certified reference value, PUBL.: published value, CV%: coefficient of variation in percent, SDEV: standard deviation.

Table 2: Median concentrations of the same water sold in both, glass and PET bottles (N=294 pairs), results of a Wilcoxon rank sum test (α=0.05 and the ratio Glass/PET. Bold print: significant difference, underline: the higher value. In the right hand columns the elements are sorted according to the Glass/PET-ratio. All analytical results (median glass and PET) are in µg/L.

Table 3: Median concentrations of the same water sold in both, green and clear glass bottles (N=136 pairs), results of a Wilcoxon rank sum test (α=0.05 and the ratio green glass/clear glass. Bold print: significant difference, underline: the higher value. In the right hand columns the elements are sorted according to the green/clear glass ratio. All analytical results (median clear and green glass) are in µg/L.

Table 4: Analytical results for the 126 European bottled water samples sold originally in the bottles used for the leaching test presented here. RDL: reported detection limit; Min.: minimum; Q5, Q25, Q75, Q95: percentiles (5, 25, 75 and 95 %); Max.: maximum value; MAD.ilr: median absolute deviation for the ilr-transformed data (see Filzmoser et al., 2009); Powers: powers of magnitude variation. All analytical results are in µg/L.

Table 5: Analytical results of the leaching test for the 3 principal bottle materials (Glass, hard PET and soft PET at pH 6.5 (demineralised water) and pH 3.5 (demineralised water, acidified). The highest value observed during the leaching test is marked in bold. All analytical results are in µg/L. IDL: instrumental detection limit, RDL: reported detection limit.

Figure captions

Fig. 1: XY-plot (left) and boxplots (right) used to compare analytical results for Sb in European bottled water sold in glass as well as PET bottles (294 pairs of PET and glass bottles).

Fig. 2: XY-plots (left) and boxplots (right) used to compare analytical results for Pb and Th in European bottled water sold in glass as well as PET bottles (294 pairs of PET and glass bottles).

Fig. 3: XY-plot (left) and boxplots (right) used to compare analytical results for Li in European bottled water sold in glass as well as PET bottles (294 pairs of PET and glass bottles).

Fig. 4: XY-plots (left) and boxplots (right) used to compare analytical results for Cr and W in European bottled water sold in green glass as well as in clear glass bottles (136 pairs of green and clear glass bottles).

Fig. 5: Leaching results over time for Pb in the 3 principal bottle materials and colours. Line style according to pH: dashed line pH 6.5, solid line pH 3.5, the lines represent the measurements at 1, 2, 3, 4, 5, 15, 30, 56, 80 and 150 days. The boxplots show the analytical results for the original mineral water samples sold in glass and in PET-bottles. Detection limit (DL), 10x, 100x and 1000x detection limit are marked in the plot (dotted lines along x-axis).

Fig. 6: Leaching results over time for Sb in the 3 principal bottle materials and colours. Line style according to pH: dashed line pH 6.5, solid line pH 3.5, the lines represent the measurements at 1, 2, 3, 4, 5, 15, 30, 56, 80 and 150 days. The boxplots show the analytical results for the original mineral water samples sold in glass and in PET-bottles. Detection limit (DL), 10x, 100x and 1000x detection limit are marked in the plot (dotted lines along x-axis).

Fig. 7: Summary of leaching results (median values only) for acidified (ACID= Y, pH 3.5) and non-acidified (ACID = N, pH 6.5) samples according to colour and bottle material (individual results indicated by different symbols for different bottle materials, see plot) for Al. The boxplots show the analytical results for the original mineral water samples sold in glass and in PET-bottles. Median value for all leaching results, 10x median and 100x median are marked in the plot.

Fig. 8: Summary of leaching results (median values only) for acidified (ACID= Y, pH 3.5) and non-acidified (ACID = N, pH 6.5) samples according to colour and bottle material for Cr. The boxplots show the analytical results for the original mineral water samples sold in glass and in PET-bottles. Median value for all leaching results, 10x median and 100x median are marked in the plot.

Fig. 9: Summary of leaching results (median values only) for acidified (ACID= Y, pH 3.5) and non-acidified (ACID = N, pH 6.5) samples according to colour and bottle material for K. The boxplots show the analytical results for the original mineral water samples sold in glass and in PET-bottles. Median value for all leaching results, 10x, 100x, 1000, and 10000x median are marked in the plot.

	IDL	RDL	SLSR-4				MinWat		
			CERT.	PUBL.	MEAN (N=82)	CV	MEAN (N=83)	SDEV	CV
	μg/L	μg/L	μg/L	μg/L	μg/L	%	μg/L	μg/L	%
Ag	0.001	0.002		0.035	0.00061	42.9	0.942	0.029	3.1
Al	0.2	0.5	54	53.5	53.1	4.6	5.03	0.549	10.9
As	0.01	0.03	0.68	0.68 - 0.7	0.689	3.5	9.83	0.293	3.0
B	0.1	2		5.7- 6.3	5.42	6.5	35.5	1.36	3.8
Ba	0.005	0.05	12.2	12.2- 12.6	12.7	1.5	35.7	1.02	2.8
Be	0.001	0.01	0.007	0.007	0.0071	21.8	8.57	0.336	3.9
Bi	0.0005	0.005	nb	0.0022	0.0028	20.7	0.989	0.036	3.6
Ca	0.005	0.01	6200	5200- 6200	5365	2.7	80199	2798	3.5
Cd	0.001	0.003	0.012	0.012	0.013	12.5	0.998	0.028	2.8
Ce	0.0005	0.001		0.360	0.365	1.6	0.014	0.00094	6.5
Co	0.002	0.01	0.033	0.033- 0.048	0.036	12.1	0.946	0.0390	4.1
Cr	0.01	0.2	0.33	0.312	0.332	11.2	1.18	0.055	4.6
Cs	0.0005	0.002		0.007- 0.009	0.0072	11.0	0.010	0.0011	10.3
Cu	0.01	0.1	1.81	1.86	1.79	3.7	1.42	0.058	4.1
Dy	0.0002	0.001		0.0242	0.0236	4.7	0.0038	0.00048	12.7
Er	0.0002	0.001		0.0134	0.0134	6.0	0.0030	0.00036	11.9
Eu	0.0002	0.001		0.008	0.0086	6.6	0.0027	0.00045	16.7
Fe	0.01	0.5	103	95- 117	100	3.1	11.2	0.482	4.3
Ga	0.0005	0.005		0.012	0.021	35.2	0.898	0.061	6.8
Gd	0.0002	0.002		0.0342	0.038	5.3	0.0037	0.00064	17.5
Ge	0.005	0.03		0.010	0.022	17.6	0.0113	0.0025	21.7
Hf	0.0001	0.002		0.0033	0.0040	33.0	0.00063	0.00022	35.5
Ho	0.0001	0.001		0.0047	0.0048	5.6	0.0013	0.00017	12.5
I	0.01	0.2		nb	1.54	8.2	2.31	0.188	8.1
K	0.05	0.1	680	597- 712	503	9.7	893	79.1	8.9
La	0.0001	0.001	0.287	0.287	0.291	1.8	0.011	0.00069	6.2
Li	0.01	0.2		0.54	0.506	11.5	8.22	0.454	5.5
Lu	0.00005	0.001		0.0019	0.0020	9.1	0.00056	0.000079	14.3
Mg	0.005	0.01	1600	1600- 1624	1519	3.3	14732	661	4.5
Mn	0.005	0.1	3.37	3.37	3.36	3.1	1.07	0.0514	4.8
Mo	0.001	0.02	0.21	0.21	0.207	4.8	1.35	0.048	3.6
Na	0.02	0.1	2400	2400- 2500	2053	4.1	9271	406	4.4
Nb	0.001	0.01		0.0041	0.0053	29.1	0.0014	0.00054	38.7
Nd	0.0001	0.001		0.269	0.265	2.3	0.011	0.0011	9.7
Ni	0.005	0.02	0.67	0.67- 0.82	0.700	6.1	1.19	0.0645	5.4
Pb	0.001	0.01	0.086	0.084- 0.086	0.084	8.5	1.09	0.0327	3.0
Pr	0.00005	0.001		0.0693	0.069	2.0	0.0025	0.00021	8.6
Rb	0.001	0.01		1.53	1.52	2.1	2.05	0.0600	2.9
Sb	0.001	0.01	0.23	0.23-0.27	0.245	2.4	0.072	0.0036	5.0
Se	0.005	0.02		0.23	0.089	12.3	9.81	0.332	3.4
Sm	0.0001	0.001		0.0574	0.059	4.1	0.0029	0.00061	21.2
Sn	0.001	0.02		0.008- 0.010	0.026	70.8	0.019	0.015	77.7
Sr	0.3	1	26.3	26.3- 28.2	26.5	2.4	1242	37.1	3.0
Ta	0.0005	0.005		0.0003	0.0010	89.2	0.00043	0.00036	82.4

			SLSR-4				MinWat		
	IDL	RDL	CERT.	PUBL.	MEAN (N=82)	CV	MEAN (N=83)	SDEV	CV
Tb	0.00005	0.001		0.0043	0.0046	4.9	0.00054	0.00010	19.0
Te	0.001	0.03		0.004	0.0080	45.9	0.985	0.044	4.5
Th	0.0001	0.001		0.019- 0.022	0.020	7.3	0.0025	0.00065	25.7
Ti	0.005	0.08		1.31- 1.56	1.05	14.0	0.060	0.046	76.2
Tl	0.0005	0.002		0.0076	0.0080	42.4	0.984	0.033	3.4
Tm	0.00005	0.001		0.0017	0.0019	8.6	0.00048	0.00010	21.6
U	0.00005	0.001	0.050	0.047- 0.053	0.049	3.4	1.53	0.047	3.1
V	0.01	0.1	0.32	0.32- 0.35	0.360	9.9	1.16	0.048	4.1
W	0.002	0.05		0.013	0.012	73.2	0.020	0.011	54.8
Y	0.00005	0.001		0.146	0.138	2.8	0.048	0.0027	5.7
Yb	0.0001	0.001		0.0120	0.012	6.4	0.0031	0.00043	13.5
Zn	0.01	0.2	0.93	0.93- 1.24	1.08	16.1	21.2	0.591	2.8
Zr	0.0001	0.001		0.12	0.097	4.8	0.026	0.0034	13.2

	MEDIAN					sorted according to ratio	
	Glass	PET	WILCOXON	Glass/PET	PET/Glass		Glass/PET
Ag	**0.00105**	**<0.001**	**0.000**	**2***		Sb	0.048
Al	**6.09**	**0.863**	**0.000**	**7**		Ta	0.9
As	0.204	0.1975	0.663	1		U	0.9
B	53.65	57.3	0.940	1		B	0.9
Ba	0.032	0.0325	0.946	1		Ca	1.0
Be	0.005105	0.00372	0.169	1		Rb	1.0
Bi	**0.000919**	**0.000546**	**0.000**	**2**		K	1.0
Ca	102.5	106	0.903	1		Mo	1.0
Cd	0.00372	0.00325	0.088	1		Ba	1.0
Ce	**0.0178**	**0.000942**	**0.000**	**19**		Na	1.0
Co	0.0227	0.0199	0.436	1		Li	1.0
Cr	**0.209**	**0.0871**	**0.000**	**2**		Ga	1.0
Cs	0.07245	0.0711	0.801	1		Mn	1.0
Cu	**0.2675**	**0.1945**	**0.000**	**1**		Cs	1.0
Dy	**0.001695**	**0.00109**	**0.000**	**2**		Mg	1.0
Er	**0.00152**	**0.000763**	**0.000**	**2**		As	1.0
Eu	0.00281	0.002525	0.136	1		I	1.0
Fe	**3.66**	**0.871**	**0.000**	**4**		Tl	1.0
Ga	<0.0005	<0.0005	0.986	1		Se	1.0
Gd	**0.002265**	**0.00134**	**0.000**	**2**		V	1.1
Ge	0.03395	0.03105	0.550	1		Ni	1.1
Hf	**0.00116**	**<0.0005**	**0.000**	**5***		W	1.1
Ho	**0.000736**	**0.000555**	**0.000**	**1**		Sr	1.1
I	4.215	4.04	0.910	1		Ge	1.1
K	2.45	2.5	0.927	1		Eu	1.1
La	**0.00843**	**0.00172**	**0.000**	**5**		Co	1.1
Li	31.2	31.35	0.932	1		Cd	1.1
Lu	**0.000298**	**0.00019**	**0.000**	**2**		Te	1.2
Mg	22.75	22.2	0.975	1		Ho	1.3
Mn	0.003	0.003	0.858	1		Be	1.4
Mo	0.2875	0.2925	0.885	1		Cu	1.4
Na	17.8	18.05	0.897	1		Nb	1.4
Nb	**0.00553**	**0.00393**	**0.000**	**1**		Tm	1.5
Nd	**0.006245**	**0.00183**	**0.000**	**3**		Yb	1.5
Ni	0.2055	0.19	0.403	1		Dy	1.6
Pb	**0.139**	**0.0102**	**0.000**	**14**		Lu	1.6
Pr	**0.00173**	**0.000392**	**0.000**	**4**		Y	1.6
Rb	3.56	3.64	0.944	1		Sm	1.7
Sb	**0.01555**	**0.326**	**0.000**	**0.048**	**21**	Bi	1.7
Se	0.0349	0.03325	0.601	1		Gd	1.7
Sm	**0.00185**	**0.00111**	**0.000**	**2**		Er	2.0
Sn	**0.0112**	**0.00424**	**0.000**	**3**		Ag	2.0
Sr	0.63	0.58	0.940	1		Tb	2.0
Ta	0.00152	0.00169	0.926	1		Cr	2.4
Tb	**0.000326**	**0.000163**	**0.000**	**2**		Sn	2.6

	MEDIAN						sorted according to ratio
	Glass	PET	WILCOXON	Glass/PET	PET/Glass		Glass/PET
Te	0.0114	0.00918	0.097	1		Nd	3.4
Th	**0.00201**	**0.000404**	**0.000**	**5**		Zn	4.1
Ti	**0.16**	**0.0294**	**0.000**	**5**		Fe	4.2
Tl	0.00512	0.0049	0.762	1		Pr	4.4
Tm	**0.000254**	**0.000172**	**0.000**	**1**		La	4.9
U	0.34	0.364	0.973	1		Th	5.0
V	0.1355	0.129	0.597	1		Hf	5.0
W	0.0154	0.0142	0.152	1		Ti	5.4
Y	**0.0222**	**0.0138**	**0.000**	**2**		Zr	6.6
Yb	**0.00161**	**0.00107**	**0.000**	**2**		Al	7.1
Zn	**2.53**	**0.621**	**0.000**	**4**		Pb	13.6
Zr	**0.05615**	**0.008515**	**0.000**	**7**		Ce	18.9

GLASS:	MEDIAN CLEAR	MEDIAN GREEN	WILCOXON	green/ clear	sorted according to ratio green/clear	
Ag	**<0.001**	**0.00123**	**0.013**	**2.5***	Mn	0.6
Al	5.17	5.35	0.572	1.0	Se	0.7
As	0.0972	0.0909	0.801	0.9	**W**	**0.8**
B	79.1	74.6	0.567	0.9	Sb	0.8
Ba	0.04	0.036	0.997	0.9	Ta	0.9
Be	0.00242	0.00243	0.986	1.0	Cs	0.9
Bi	0.000834	0.000898	0.349	1.1	Ba	0.9
Ca	82.3	85.4	0.805	1.0	Ge	0.9
Cd	0.00292	0.0029	0.715	1.0	Cu	0.9
Ce	**0.0117**	**0.0182**	**0.000**	**1.6**	As	0.9
Co	**0.0222**	**0.029**	**0.000**	**1.3**	U	0.9
Cr	**0.138**	**1.01**	**0.000**	**7.3**	Rb	0.9
Cs	0.0192	0.0171	0.700	0.9	Pb	0.9
Cu	0.243	0.226	0.953	0.9	K	0.9
Dy	0.0011	0.00148	0.078	1.3	V	0.9
Er	**0.0011**	**0.00119**	**0.003**	**1.1**	B	0.9
Eu	0.00318	0.00315	0.617	1.0	Mo	1.0
Fe	**3.09**	**4.09**	**0.000**	**1.3**	Li	1.0
Ga	<0.0005	0.00197	0.079	DL	Eu	1.0
Gd	0.00208	0.00227	0.080	1.1	Te	1.0
Ge	0.0194	0.0176	0.962	0.9	Cd	1.0
Hf	0.00101	0.00109	0.266	1.1	I	1.0
Ho	0.000666	0.000708	0.248	1.1	Be	1.0
I	7.52	7.5	0.986	1.0	Zn	1.0
K	3.4	3.2	0.878	0.9	Mg	1.0
La	**0.00455**	**0.00841**	**0.000**	**1.8**	Sr	1.0
Li	11.4	11.2	0.614	1.0	Al	1.0
Lu	0.000261	0.000271	0.620	1.0	Ca	1.0
Mg	20.1	20.4	0.859	1.0	Lu	1.0
Mn	0.005	0.003	0.667	0.6	Na	1.1
Mo	0.223	0.214	0.645	1.0	Tb	1.1
Na	35.7	37.5	0.845	1.1	Ho	1.1
Nb	**0.00437**	**0.00649**	**0.000**	**1.5**	Tl	1.1
Nd	**0.00405**	**0.00633**	**0.000**	**1.6**	Bi	1.1
Ni	0.273	0.297	0.502	1.1	Hf	1.1
Pb	0.135	0.127	0.890	0.9	Yb	1.1
Pr	**0.00102**	**0.00153**	**0.000**	**1.5**	**Er**	**1.1**
Rb	2.32	2.18	0.780	0.9	Ni	1.1
Sb	0.0185	0.015	0.620	0.8	Gd	1.1
Se	0.0321	0.0231	0.038	0.7	Y	1.1
Sm	0.00164	0.00196	0.012	1.2	Tm	1.2
Sn	0.0107	0.0131	0.019	1.2	Sm	1.2
Sr	0.53	0.539	0.852	1.0	Sn	1.2
Ta	0.00189	0.00168	0.484	0.9	**Co**	**1.3**
Tb	0.000238	0.000251	0.335	1.1	**Fe**	**1.3**

GLASS:	MEDIAN CLEAR	MEDIAN GREEN	WILCOXON	green/ clear
Te	0.00963	0.00954	0.982	1.0
Th	**0.00132**	**<u>0.00249</u>**	**0.000**	**1.9**
Ti	**0.123**	**<u>0.174</u>**	**0.000**	**1.4**
Tl	0.00185	0.00199	0.804	1.1
Tm	0.000208	0.000243	0.074	1.2
U	0.126	0.118	0.879	0.9
V	0.137	0.129	0.661	0.9
W	**<u>0.0172</u>**	**0.0137**	**0.004**	**0.8**
Y	0.0127	0.0146	0.396	1.1
Yb	0.00111	0.0012	0.547	1.1
Zn	2.27	2.3	0.408	1.0
Zr	**0.036**	**<u>0.061</u>**	**0.000**	**1.7**

sorted according to ratio green/clear	
Dy	1.3
Ti	**1.4**
Nb	**1.5**
Pr	**1.5**
Ce	**1.6**
Nd	**1.6**
Zr	**1.7**
La	**1.8**
Th	**1.9**
Ag	**2.5**
Cr	**7.3**
Ga	DL

Element	RDL	Min	Q5	Q25	MEDIAN	Q75	Q95	Max	MAD.ilr	Powers
Ag	0.002	<0.002	<0.002	<0.002	<0.002	0.0030	0.010	1.2		5
Al	0.5	<0.5	<0.5	0.845	2.7	8.1	21	132	1.18	4
As	0.03	<0.03	<0.03	0.058	0.156	0.44	2.6	4.8	1.08	3
B	2	2.9	6.4	21	51	255	2100	120000	1.31	6
Ba	0.05	0.787	1.7	18	51	72	441	26800	0.606	6
Be	0.01	<0.01	<0.01	<0.01	<0.01	0.038	0.62	38		6
Bi	0.005	<0.005	<0.005	<0.005	<0.005	<0.005	0.0056	0.042		3
Ca	0.01	1370	4700	49600	86600	130000	376000	564000	0.533	3
Cd	0.003	<0.003	<0.003	<0.003	0.0038	0.0087	0.037	0.141	0.822	4
Ce	0.001	<0.001	<0.001	<0.001	0.0060	0.022	0.092	0.30	1.9	4
Co	0.01	<0.01	<0.01	0.016	0.029	0.065	0.555	16.4	0.699	5
Cr	0.2	<0.2	<0.2	<0.2	<0.2	0.26	1.8	7.4		3
Cs	0.002	<0.002	0.0034	0.015	0.071	3.9	24	276	2.31	7
Cu	0.1	0.0192	0.036	0.126	0.273	0.537	5.1	20	0.748	4
Dy	0.001	<0.001	<0.001	<0.001	0.0015	0.0053	0.022	0.103		4
Er	0.001	<0.001	<0.001	<0.001	0.0014	0.0050	0.035	0.118		5
Eu	0.001	<0.001	<0.001	0.0017	0.0038	0.0059	0.026	1.55	0.624	5
Fe	0.5	<0.5	<0.5	0.50	1.55	7.9	92.1	13500	1.35	7
Ga	0.005	<0.005	<0.005	<0.005	<0.005	0.0050	0.0112	0.0277		3
Gd	0.002	<0.002	<0.002	<0.002	0.0020	0.0067	0.023	0.174		4
Ge	0.03	<0.03	<0.03	<0.03	0.038	0.153	0.851	110		6
Hf	0.002	<0.002	<0.002	<0.002	<0.002	0.0044	0.096	1.57	1.3	5
Ho	0.001	<0.001	<0.001	<0.001	<0.001	0.0021	0.0079	0.023		4
I	0.2	0.41	0.79	3.0	5.4	10	95	4030	0.65	5
K	0.1	<0.1	300	1520	3500	13200	32300	177000	1.3	5
La	0.001	<0.001	<0.001	0.0012	0.0051	0.014	0.061	0.175	1.24	4
Li	0.2	<0.2	0.752	6.7	28.2	175	857	9860	1.73	6
Lu	0.001	<0.001	<0.001	<0.001	<0.001	0.0011	0.0084	0.030		4
Mg	0.01	0.39	1020	6180	21000	49000	106000	250300	1.02	5
Mn	0.1	<0.1	<0.1	0.362	1.4	28	376	1740	2.36	6
Mo	0.02	<0.02	<0.02	0.048	0.2	0.479	3.4	50.5	1.23	5
Na	0.1	400	2650.0	8800	25200	276000	799000	816000	1.59	5
Nb	0.01	<0.01	<0.01	<0.01	<0.01	<0.01	0.057	0.262		4
Nd	0.001	<0.001	<0.001	0.0011	0.0044	0.013	0.060	0.148	1.31	4
Ni	0.02	<0.02	0.020	0.090	0.369	1.2	8.7	27	1.42	5
Pb	0.01	<0.01	<0.01	<0.01	0.0338	0.162	0.402	1.1		4
Pr	0.001	<0.001	<0.001	<0.001	<0.001	<0.001	0.012	0.035		4
Rb	0.01	0.108	0.402	1.3	4.3	26	80	631	1.71	4
Sb	0.01	<0.01	<0.01	0.020	0.114	0.414	0.58	0.931	1.41	3
Se	0.02	<0.02	<0.02	<0.02	0.023	0.11	0.44	32.9		5
Sm	0.001	<0.001	<0.001	0.0011	0.0016	0.0048	0.023	0.132	0.666	4
Sn	0.02	<0.02	<0.02	<0.02	<0.02	0.041	0.29	6.24		5
Sr	1	8	29	169	366	1600	10900	26	1.01	5
Ta	0.005	<0.005	<0.005	<0.005	<0.005	<0.005	0.0050	0.0093		2
Tb	0.001	<0.00005	<0.00005	0.0001	0.00023	0.00077	0.0039	0.016	1.03	4
Te	0.03	<0.03	<0.03	<0.03	<0.03	<0.03	0.106	0.208		3

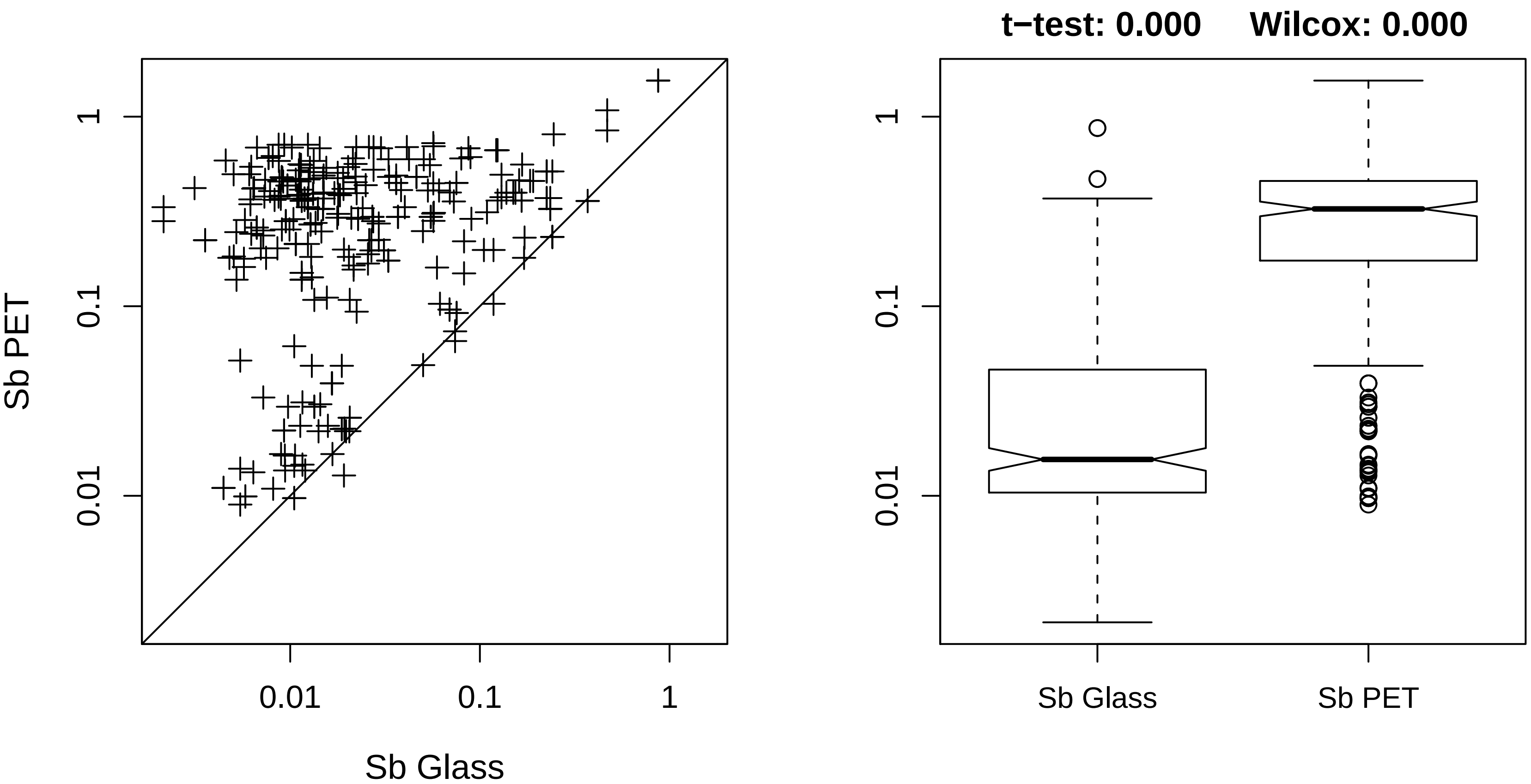
Element	RDL	Min	Q5	Q25	MEDIAN	Q75	Q95	Max	MAD.ilr	Powers
Th	0.001	<0.001	<0.001	<0.001	<0.001	0.0020	0.0056	0.059		4
Ti	0.08	<0.08	<0.08	<0.08	<0.08	0.2	1.1	6		4
Tl	0.002	<0.002	<0.002	<0.002	0.0044	0.0083	0.0782	0.634	0.913	4
Tm	0.001	<0.001	<0.001	<0.001	<0.001	<0.001	0.0058	0.0088		3
U	0.001	<0.001	0.0026	0.0218	0.135	1.0	5.5	229	2.07	7
V	0.1	<0.1	<0.1	<0.1	0.14	0.233	1.1	11.8	0.582	4
W	0.05	<0.05	<0.05	<0.05	<0.05	<0.05	0.21	2.3		4
Y	0.001	<0.001	0.0024	0.0066	0.019	0.069	0.37	0.97	1.26	4
Yb	0.001	<0.001	<0.001	<0.001	0.0016	0.0067	0.044	0.093	1.13	3
Zn	0.2	<0.2	<0.2	0.39	1.3	4.0	11	113	1.22	5
Zr	0.001	<0.001	0.0015	0.0050	0.032	0.21	12	165	1.94	7

			EU Dir.	EU Dir.	GLASS				hard PET				soft PET			
			2003/40	1998/83	pH 6.5 (N=10)		pH 3.5 (N=42)		pH 6.5 (N=5)		pH 3.5 (N=10)		pH 6.5 (N=10)		pH 3.5 (N=49)	
	IDL	RDL	Miwa	Driwa	MEDIAN	MAX	MEDIAN	MAX	MEDIAN	MAX	MEDIAN	MAX	MEDIAN	MAX	MEDIAN	MAX
Ag	0.001	0.002			<0.001	0.0015	<0.001	0.0067	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	**0.62**
Al	0.2	0.5			0.38	24	8.3	**68**	<0.3	0.61	<0.3	0.61	<0.3	0.96	<0.3	36
As	0.01	0.03	10	10	<0.01	0.090	<0.01	**0.074**	<0.01	0.010	<0.01	0.010	<0.01	0.011	<0.01	0.037
B	0.1	2	*	1000	0.30	1.1	0.25	**8.5**	<0.1	0.31	0.00050	0.31	<0.1	0.385	<0.1	0.43
Ba	0.005	0.05	1000		0.034	2.0	0.15	**261**	0.0058	2.4	0.012	2.4	0.0088	1.3	0.029	36
Be	0.001	0.01			<0.001	0.0039	0.0011	**0.44**	<0.001	0.0021	<0.001	0.0021	<0.001	0.0019	<0.001	0.25
Bi	0.0005	0.005			<0.0005	0.0020	<0.0005	**0.022**	<0.0005	0.00068	<0.0005	0.00068	<0.0005	0.00075	<0.0005	0.0040
Ca	0.005	0.01			11	550	54	**29200**	7.1	16	9.9	16	5.4	38	16	4300
Cd	0.001	0.003	3	5	<0.001	0.0021	<0.001	**0.0076**	<0.001	0.0021	<0.001	0.0021	<0.001	0.0024	<0.001	0.0029
Ce	0.0005	0.001			<0.0005	0.0041	0.0029	**0.30**	<0.0005	0.00054	<0.0005	0.00054	<0.0005	0.00096	<0.0005	0.042
Co	0.002	0.01			<0.002	0.0088	0.0028	**0.43**	<0.002	0.0032	<0.002	0.0032	<0.002	0.011	<0.002	0.16
Cr	0.01	0.2	50	50	0.046	0.21	0.063	**2.0**	0.042	0.17	0.044	0.17	0.042	0.22	0.041	0.21
Cs	0.0005	0.002			0.0022	0.013	0.0034	**0.37**	<0.001	0.0020	<0.001	0.0020	<0.001	0.0051	<0.001	0.015
Cu	0.01	0.1	1000	2000	0.023	0.66	0.050	**1.8**	0.014	0.045	0.017	0.045	0.013	0.057	0.022	0.68
Dy	0.0002	0.001			0.00017	0.00064	0.00038	0.0038	0.00019	0.00071	0.00021	0.00071	0.00018	0.00065	0.00025	**0.052**
Er	0.0002	0.001			0.00014	0.00051	0.00036	0.0094	0.00013	0.00053	0.00015	0.00053	0.00013	0.00054	0.00019	**0.032**
Eu	0.0002	0.001			<0.0001	0.00034	0.00017	**0.020**	<0.0001	0.00053	<0.0001	0.00053	<0.0001	0.00040	0.00012	0.0087
Fe	0.01	0.5		200(g)	<0.1	2.0	0.91	19	<0.1	0.24	<0.1	0.24	<0.1	0.30	<0.1	**599**
Ga	0.0005	0.005			<0.0005	0.013	0.0017	**0.020**	<0.0005	0.0055	<0.0005	0.0055	<0.0005	0.0081	<0.0005	0.0072
Gd	0.0002	0.002			0.00029	0.0013	0.00051	0.0042	0.00026	0.0011	0.00027	0.0011	0.00027	0.00091	0.00035	**0.048**
Ge	0.005	0.03			<0.005	0.0082	<0.005	0.025	<0.005	0.0055	<0.005	0.0055	<0.005	0.014	<0.005	**0.14**
Hf	0.0001	0.002			<0.0005	0.00089	<0.0005	**0.0014**	<0.0005	0.00082	<0.0005	0.00082	<0.0005	0.00069	<0.0005	0.0013
Ho	0.0001	0.001			0.00019	0.00054	0.00025	0.0011	0.00018	0.00050	0.00019	0.00050	0.00019	0.00062	0.00021	**0.011**
I	0.01	0.2			<0.2	<0.2	<0.2	**86**	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	4.0
K	0.05	0.1			8.2	26	14	**57**	11	25	11	25	10	26	11	27
La	0.0001	0.001			<0.0005	0.0019	0.0011	**0.022**	<0.0005	<0.0005	<0.0005	<0.0005	<0.0005	0.00051	<0.0005	0.0204
Li	0.01	0.2			<0.1	0.16	<0.1	**3.23**	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
Lu	0.00005	0.001			0.000051	0.00023	0.000079	0.00074	<0.00005	0.00014	0.000052	0.00014	<0.00005	0.00017	0.000063	**0.00528**
Mg	0.005	0.01			1.5	73	5.6	**1290**	0.50	1.6	0.51	1.6	0.25	7.1	0.64	76
Mn	0.005	0.1	500	50(g)	0.026	0.28	0.071	**94**	0.0043	0.078	0.0065	0.078	0.0048	0.26	0.014	6.6
Mo	0.001	0.02			0.0016	0.0042	0.0015	**0.0089**	<0.001	0.0030	0.0013	0.0030	<0.001	0.0044	0.0011	0.0060
Na	0.02	0.1		200(g)	25	353	34	**780**	3.1	16	4.6	16	4.6	18	2.5	27
Nb	0.001	0.01			<0.001	0.0067	<0.001	**0.020**	<0.001	0.0016	<0.001	0.0016	<0.001	0.0014	<0.001	0.011
Nd	0.0001	0.001			0.00035	0.0032	0.0012	0.017	0.00032	0.00098	0.00032	0.00098	0.00033	0.0015	0.00043	**0.043**
Ni	0.005	0.02	20	20	<0.01	0.031	0.010	**0.46**	<0.01	0.012	<0.01	0.012	<0.01	0.017	<0.01	0.13

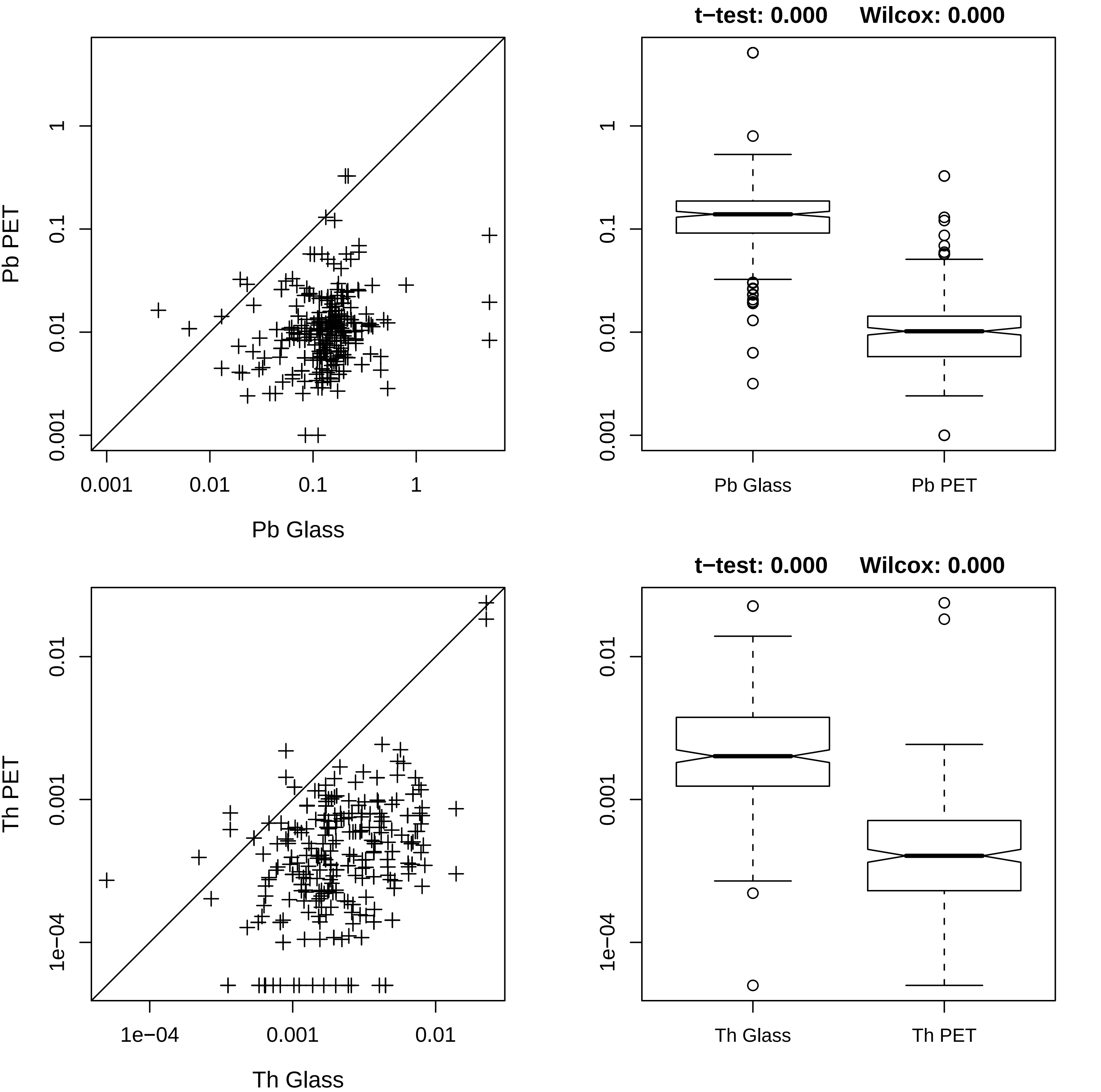
	IDL	RDL	EU Dir. 2003/40 Miwa	EU Dir. 1998/83 Driwa	GLASS pH 6.5 (N=10) MEDIAN	MAX	pH 3.5 (N=42) MEDIAN	MAX	hard PET pH 6.5 (N=5) MEDIAN	MAX	pH 3.5 (N=10) MEDIAN	MAX	soft PET pH 6.5 (N=10) MEDIAN	MAX	pH 3.5 (N=49) MEDIAN	MAX
Pb	0.001	0.01	10	10	0.0047	0.112	0.038	**0.611**	<0.002	0.0044	<0.002	0.0044	<0.002	0.016	<0.002	0.064
Pr	0.00005	0.001			0.000058	0.00076	0.00026	0.00461	<0.00005	0.00022	0.000061	0.00022	<0.00005	0.00019	0.000065	**0.00706**
Rb	0.001	0.01			0.00955	0.10	0.016	**0.20**	0.0015	0.0051	0.0015	0.0051	0.0015	0.0095	0.0014	0.011
Sb	0.001	0.01			<0.002	0.093	<0.002	**0.45**	0.0030	0.12	0.0085	0.12	0.025	0.21	0.027	0.25
Se	0.005	0.02	10	10	<0.01	0.013	<0.01	**0.018**	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	0.012
Sm	0.0001	0.001			0.00033	0.0012	0.00061	0.0045	0.00035	0.0011	0.00035	0.0011	0.00031	0.0011	0.00041	**0.025**
Sn	0.001	0.02			0.0068	0.51	0.0079	**3.81**	0.0018	0.29	0.0026	0.29	0.0021	1.09	0.00263	0.24
Sr	0.3	1			0.13	0.55	0.28	**227**	0.15	1.8	0.12	1.8	0.10	1.3	0.15	37
Ta	0.0005	0.005			<0.001	0.011	<0.001	**0.031**	<0.001	0.0044	<0.001	0.0044	<0.001	<0.001	<0.001	0.0053
Tb	0.00005	0.001			<0.00005	0.00020	0.000079	0.00071	<0.00005	0.00016	<0.00005	0.00016	0.000051	0.00019	0.000056	**0.0085**
Te	0.001	0.03			<0.005	0.012	<0.005	**0.020**	<0.005	0.012	<0.005	0.012	<0.005	0.013	<0.005	0.016
Th	0.0001	0.001			0.00010	0.00097	0.00018	**0.0021**	0.00012	0.00062	0.00011	0.00062	0.00011	0.00072	0.00011	0.00099
Ti	0.005	0.08			<0.01	0.086	0.0016	**0.53**	<0.01	0.030	<0.01	0.030	<0.01	0.031	<0.01	0.073
Tl	0.0005	0.002			0.003265	0.026	0.00088	**0.059**	0.00056	0.0032	0.00088	0.0032	0.00072	0.011	<0.0005	0.0055
Tm	0.00005	0.001			<0.00005	0.00022	0.000076	0.00063	<0.00005	0.00020	<0.00005	0.00020	<0.00005	0.00020	0.000063	**0.0048**
U	0.00005	0.001			<0.0005	0.012	0.00056	**0.099**	<0.0005	0.017	0.00084	0.017	<0.0005	0.00076	<0.0005	0.061
V	0.01	0.1			0.012	0.047	0.016	**0.078**	0.014	0.0646	0.015	0.065	0.016	0.064	0.017	**0.078**
W	0.002	0.05			0.015	0.70	0.0099	**1.39**	0.0063	0.16	0.0065	0.16	0.0064	0.49	0.0067	0.58
Y	0.00005	0.001			<0.0005	0.0017	0.0011	0.054	<0.0005	0.00076	<0.0005	0.00076	<0.0005	0.00059	<0.0005	**0.47**
Yb	0.0001	0.001			0.00022	0.00091	0.00035	0.0033	0.00024	0.00086	0.00021	0.00086	<0.0002	0.00064	0.00027	**0.031**
Zn	0.01	0.2			0.11	0.71	0.37	**12**	<0.05	0.14	0.052	0.14	<0.05	1.0	0.0841	1.1
Zr	0.0001	0.001			<0.001	0.023	<0.001	0.052	<0.001	0.0016	<0.001	0.0016	<0.001	0.0040	<0.001	**0.073**

*:pending

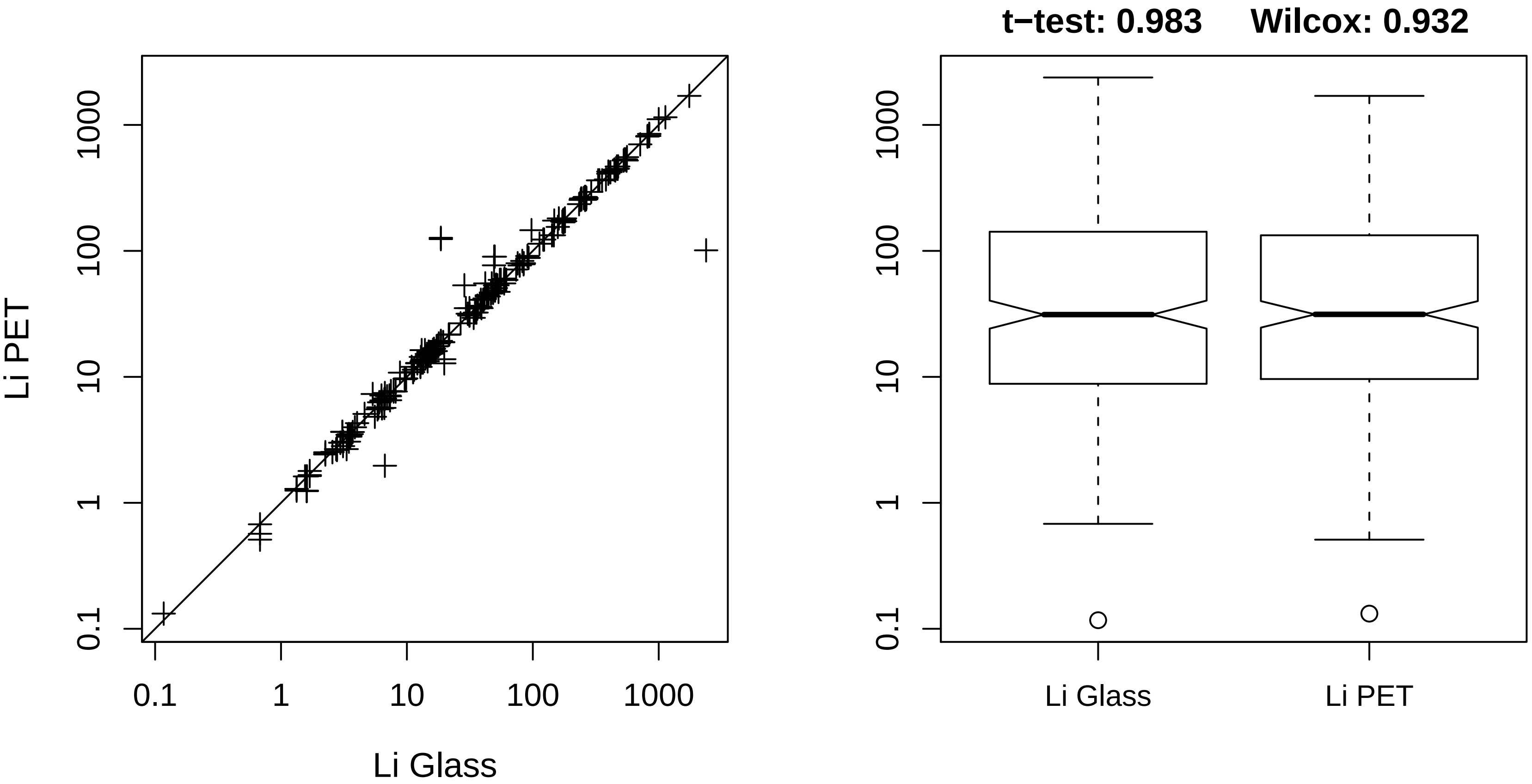
(g): guideline value

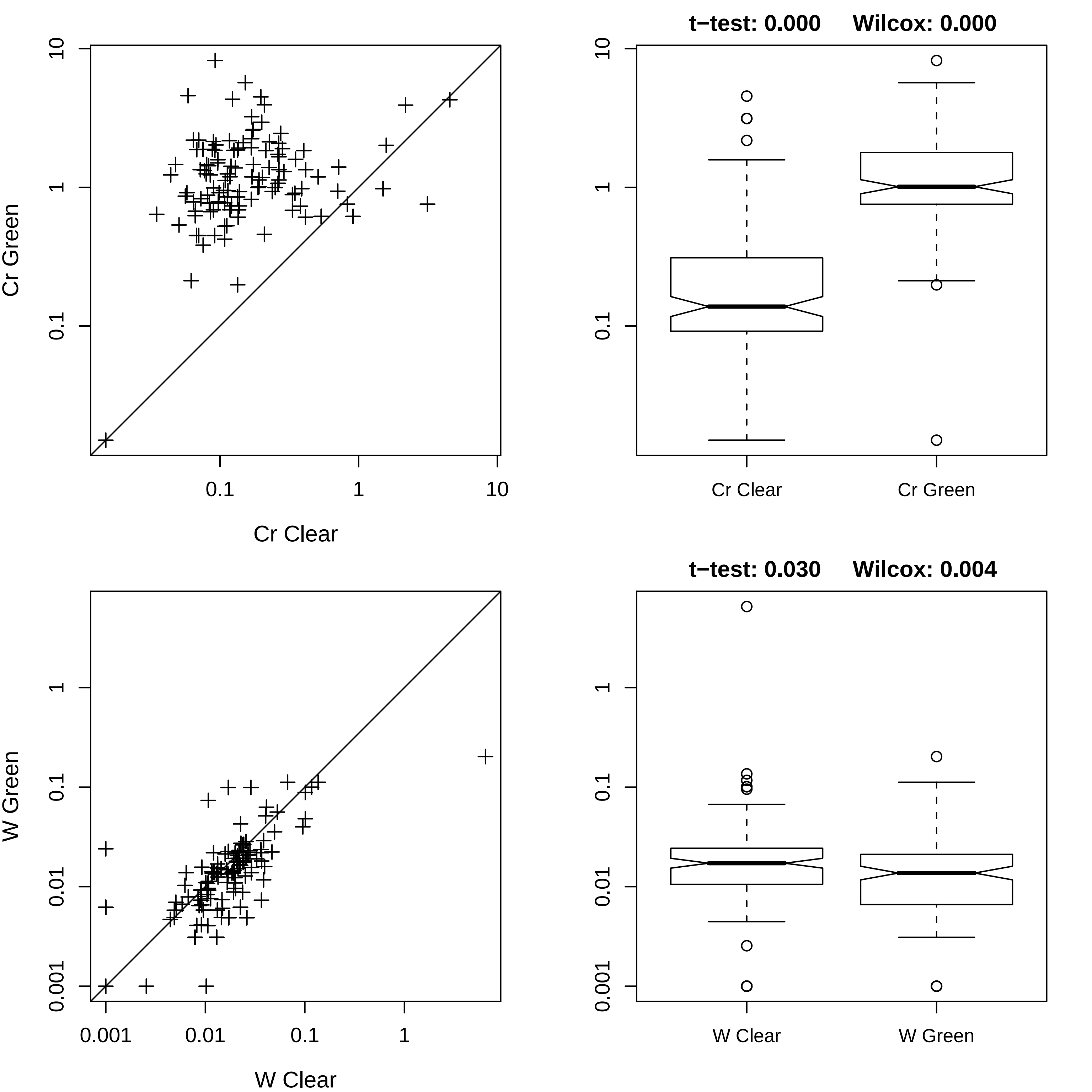


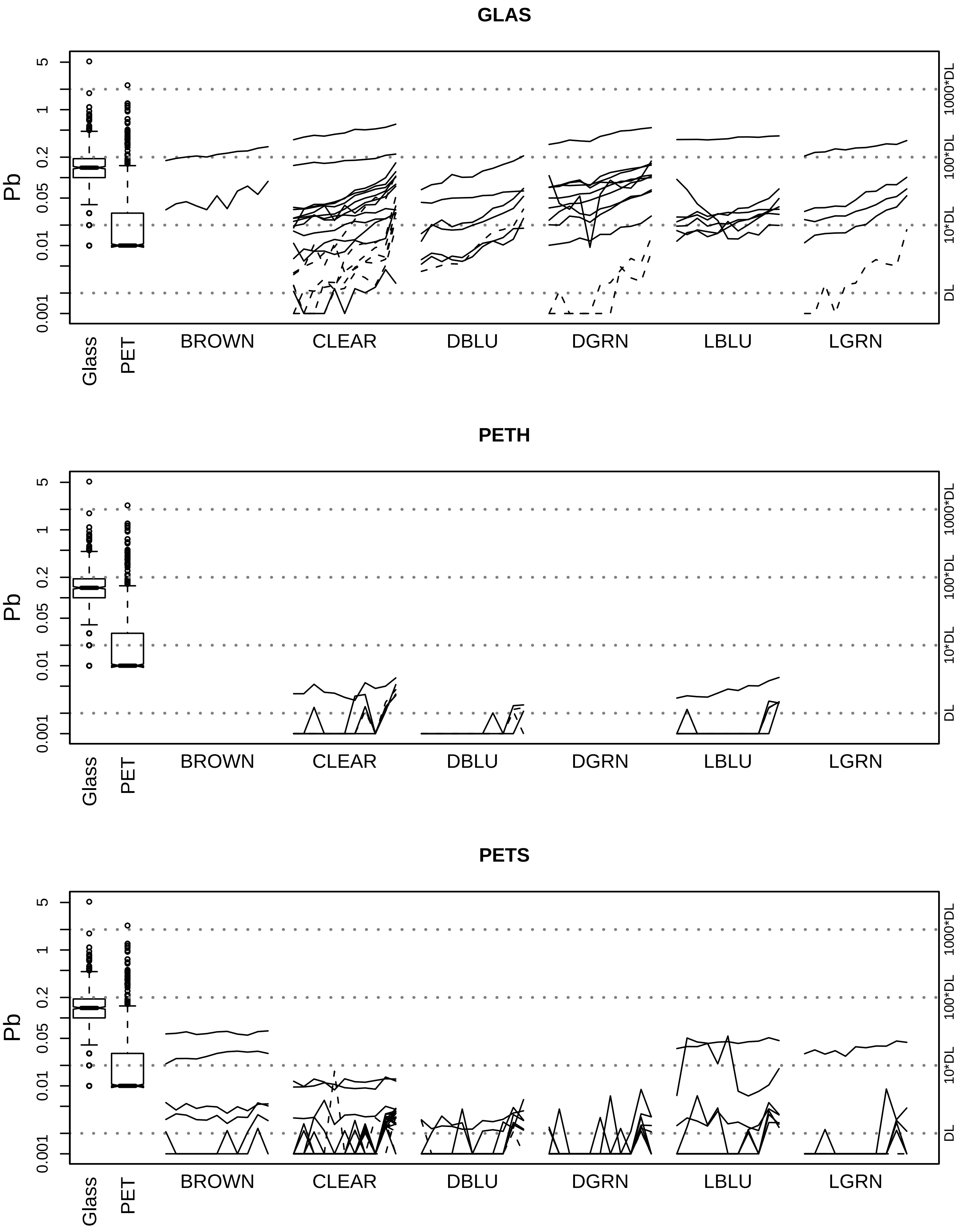
t−test: 0.000
Wilcox: 0.000
Sb PET
Sb Glass
1
0.1
0.01
Sb Glass
Sb PET

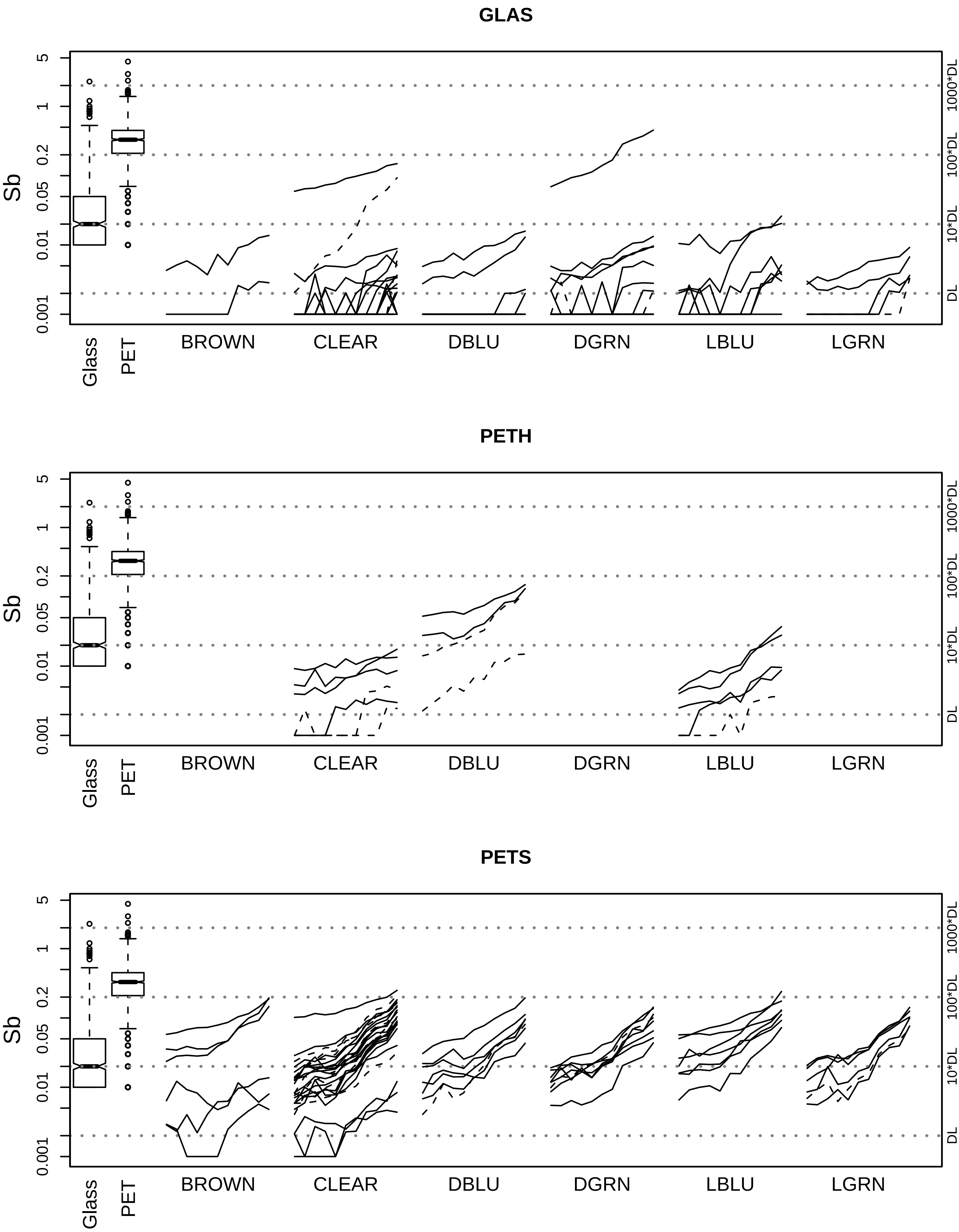


t−test: 0.000
Wilcox: 0.000
Pb PET
Pb Glass
0.001
0.01
0.1
1
Pb Glass
Pb PET
t−test: 0.000
Wilcox: 0.000
Th PET
Th Glass
1e−04
0.001
0.01
Th Glass
Th PET

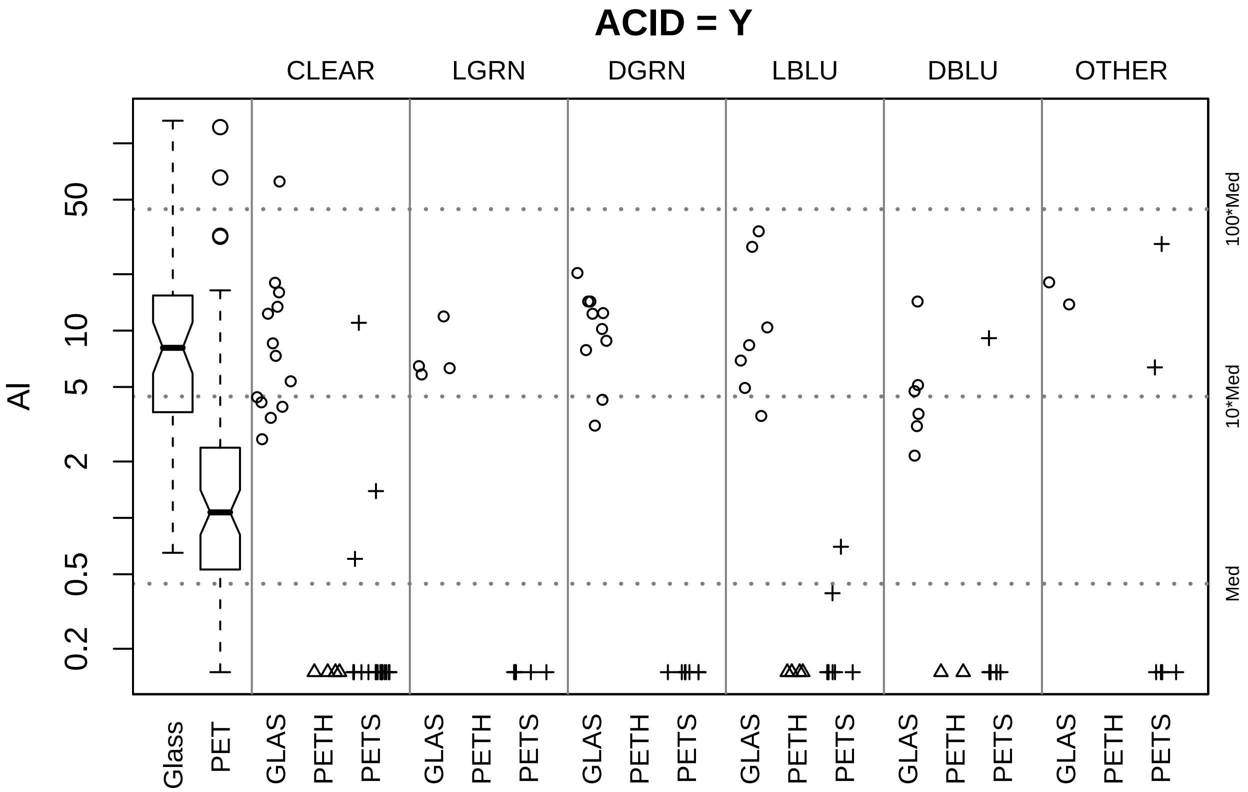

t−test: 0.983
Wilcox: 0.932
Li PET
Li Glass
0.1
1
10
100
1000
Li Glass
Li PET

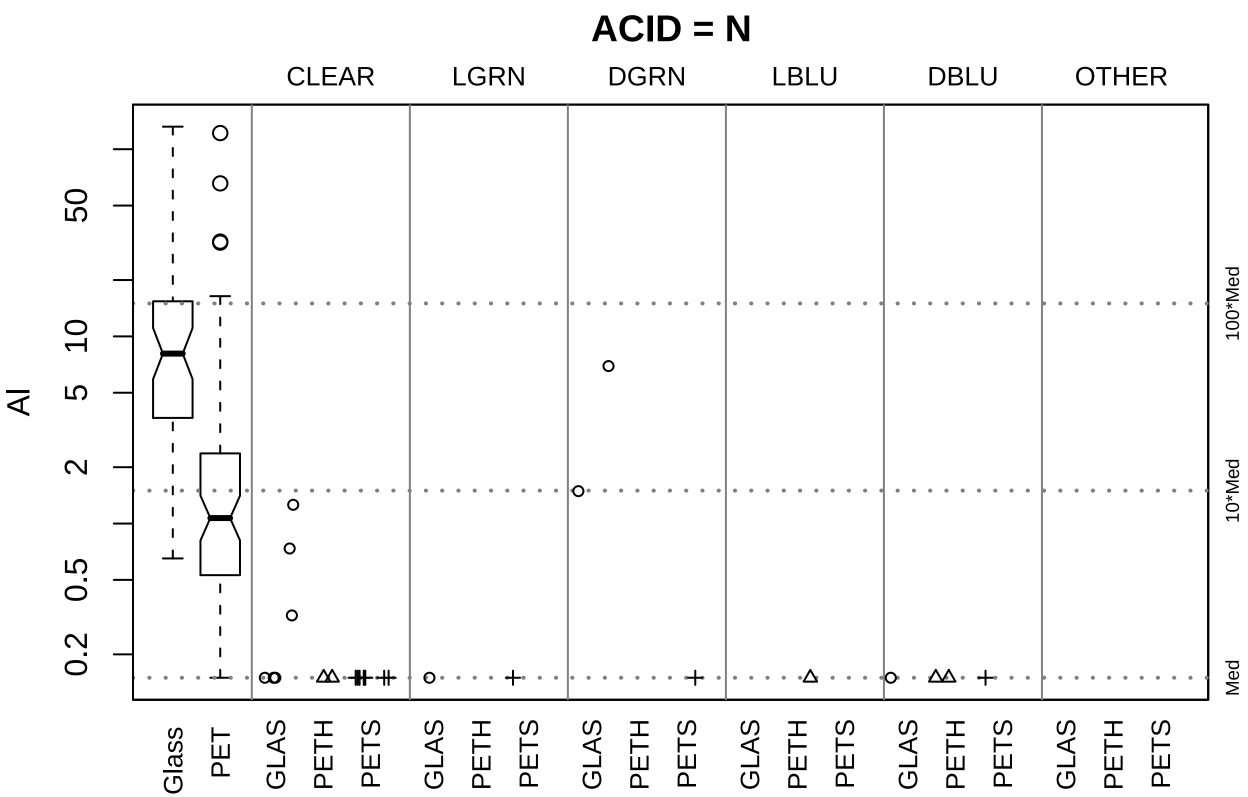

Cr Green
Cr Clear
10
1
0.1
t−test: 0.000   Wilcox: 0.000
Cr Clear
Cr Green
W Green
W Clear
0.001
0.01
0.1
1
t−test: 0.030   Wilcox: 0.004
W Clear
W Green

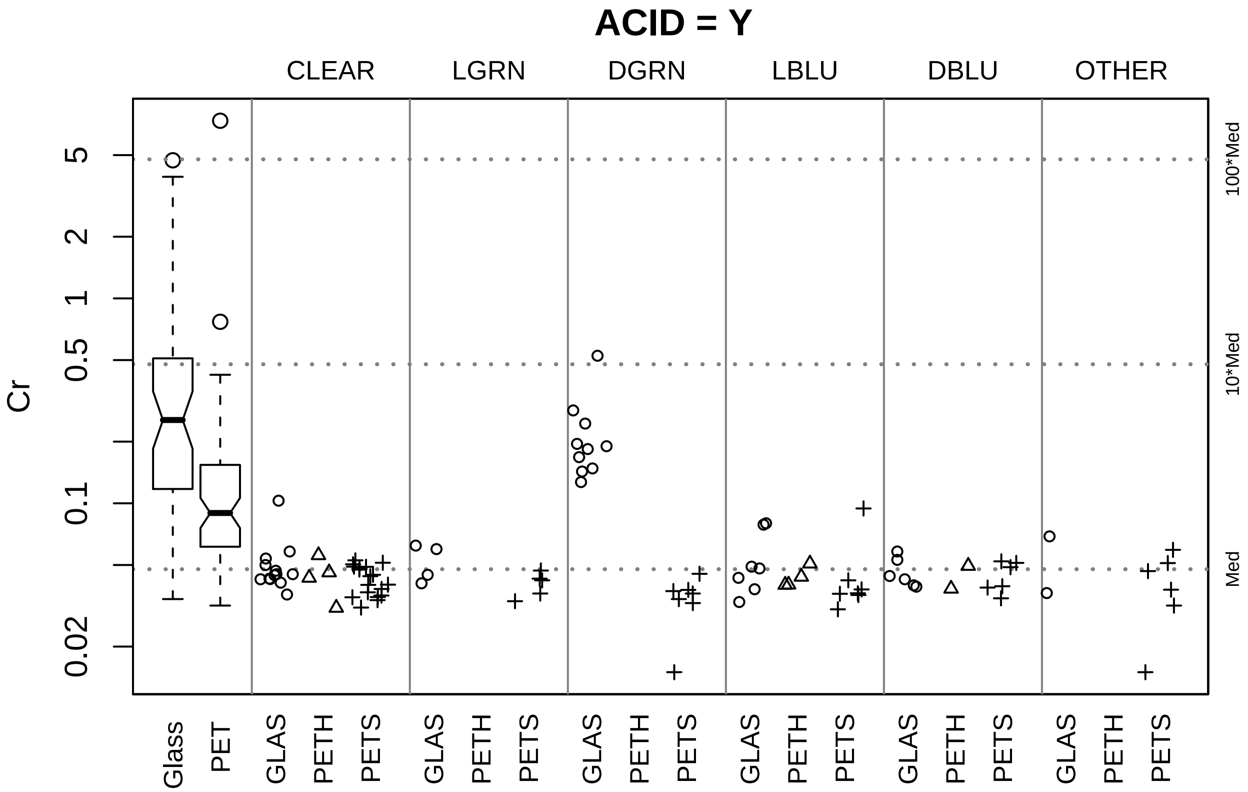

GLAS
PETH
PETS
Pb
5
1
0.2
0.05
0.01
0.001
1000*DL
100*DL
10*DL
DL
Glass
PET
BROWN
CLEAR
DBLU
DGRN
LBLU
LGRN



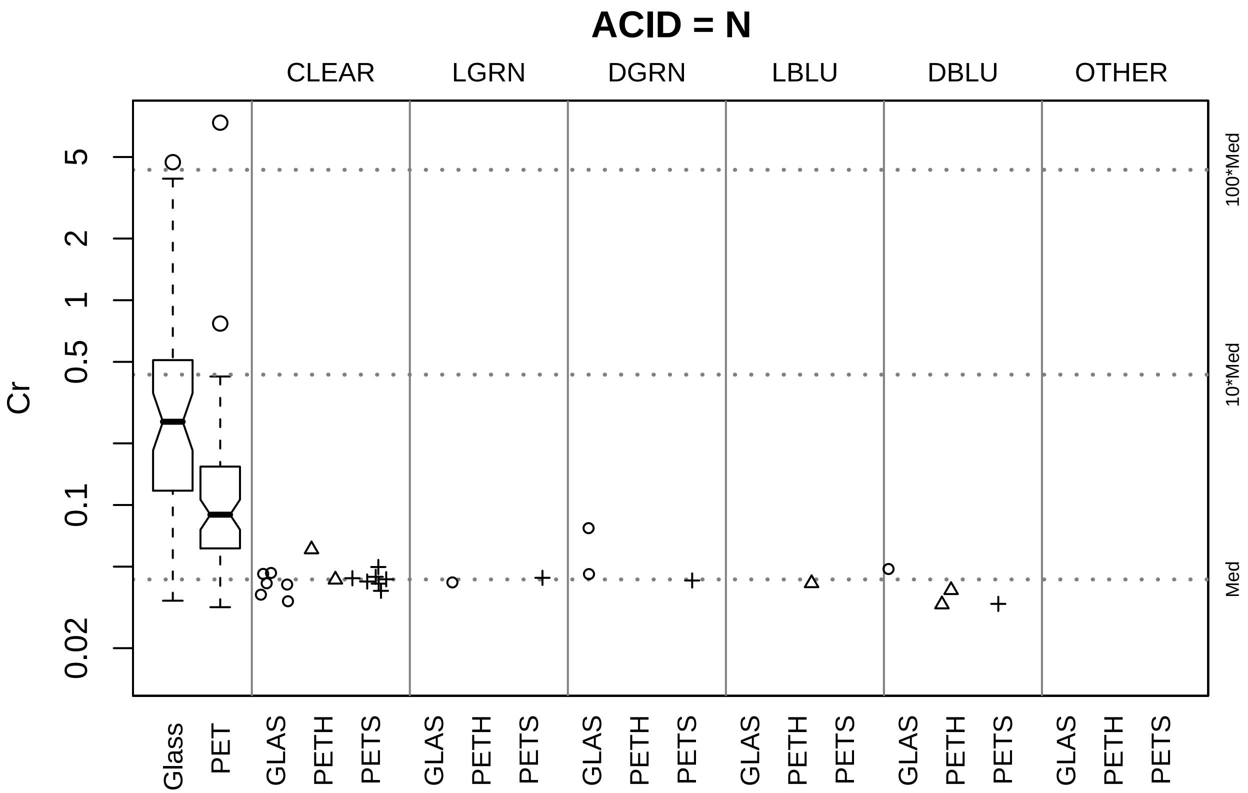
GLAS
PETH
PETS
Sb
5
1
0.2
0.05
0.01
0.001
1000*DL
100*DL
10*DL
DL
Glass
PET
BROWN
CLEAR
DBLU
DGRN
LBLU
LGRN


ACID = Y
CLEAR
LGRN
DGRN
LBLU
DBLU
OTHER
AI
50
10
5
2
0.5
0.2
100*Med
10*Med
Med
Glass
PET
GLAS
PETH
PETS

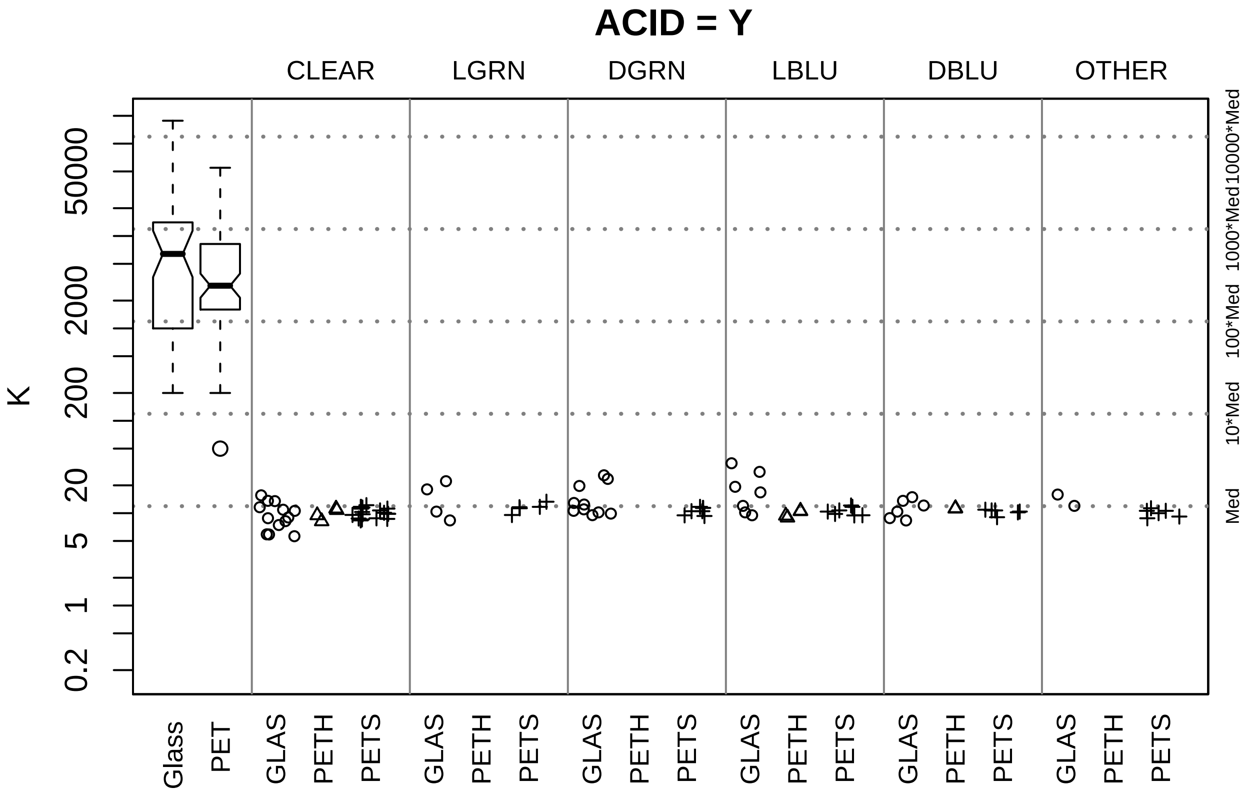

ACID = N
CLEAR
LGRN
DGRN
LBLU
DBLU
OTHER
AI
50
10
5
2
0.5
0.2
100*Med
10*Med
Med
Glass
PET
GLAS
PETH
PETS



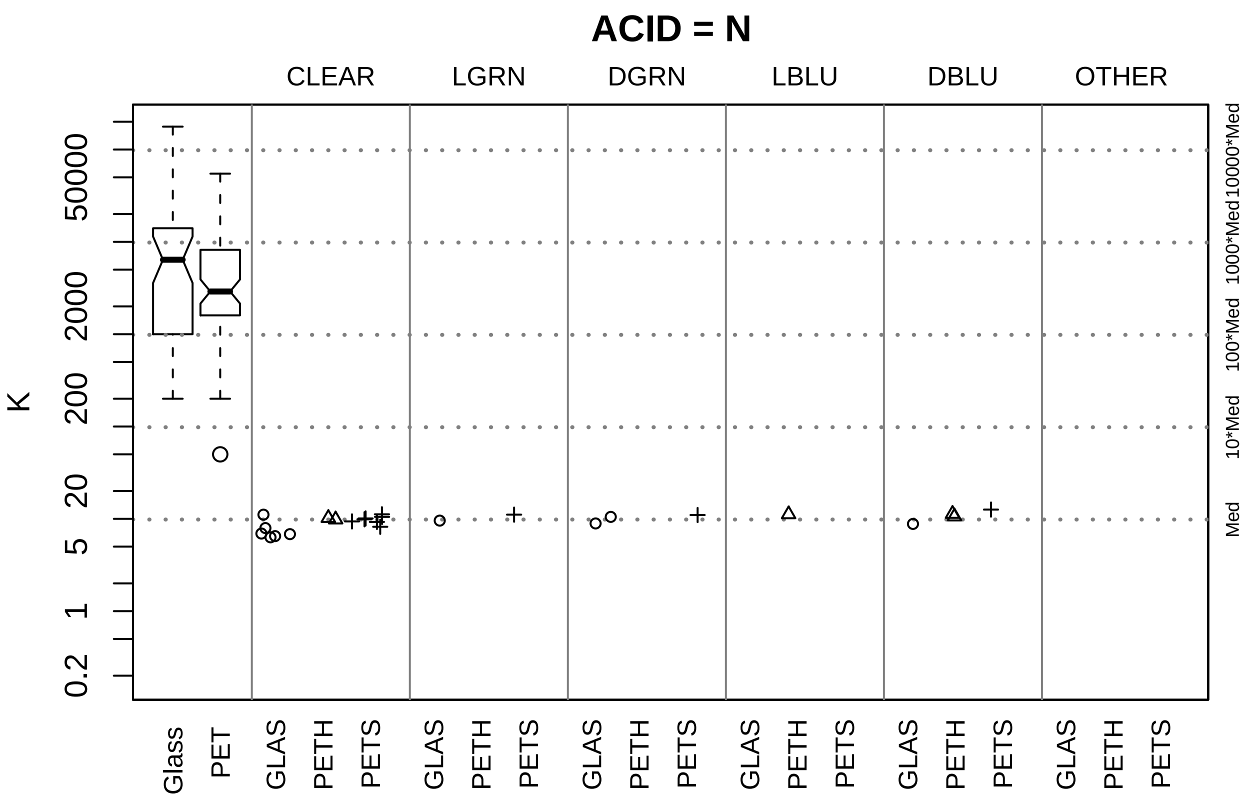
ACID = Y
CLEAR
LGRN
DGRN
LBLU
DBLU
OTHER
Cr
5
2
1
0.5
0.1
0.02
100*Med
10*Med
Med
Glass
PET
GLAS
PETH
PETS



ACID = N
CLEAR
LGRN
DGRN
LBLU
DBLU
OTHER
Cr
5
2
1
0.5
0.1
0.02
100*Med
10*Med
Med
Glass
PET
GLAS
PETH
PETS



ACID = Y
CLEAR
LGRN
DGRN
LBLU
DBLU
OTHER
K
0.2
1
5
20
200
2000
50000
Med
10*Med
100*Med
1000*Med
10000*Med
Glass
PET
GLAS
PETH
PETS



ACID = N
CLEAR
LGRN
DGRN
LBLU
DBLU
OTHER
K
0.2
1
5
20
200
2000
50000
Med
10*Med
100*Med
1000*Med
10000*Med
Glass
PET
GLAS
PETH
PETS