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Original Research Article

Comparison of the chemical composition of British and Continental European bottled waters by multivariate analysis

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ABSTRACT

The elemental composition of 37 bottled waters from the UK and continental Europe has been determined. Ca, K, Mg, Na, Al, As, Ba, Cd, Cr, Co, Cu, Fe, Mn, Mo, Ni, Pb, Sb, Sr, U, V and Zn were determined by ICP-OES and ICP-MS, in addition to inorganic and total organic carbon. The composition of all the waters analysed fell within the guideline values recommended by the World Health Organization. Na, Ca, Sr and Ba showed the widest variation in concentrations, ranging over two orders of magnitude. Levels of Fe were below the limit of detection (30 $\mu g\ L^{-1}$) in all samples analysed. Waters produced in the UK generally showed lower levels of most major elements and trace metals, with the exception of Ba (up to 455 $\mu g\ L^{-1}$). Italian waters showed the highest concentrations of Sr (3000–8000 $\mu g\ L^{-1}$) and U (8–13 $\mu g\ L^{-1}$), whereas waters produced in Slovakia and the Czech Republic showed the highest levels of Pb (0.7–4 $\mu g\ L^{-1}$). The use of multivariate analysis reveals an association between high alkaline metal content and high concentrations of As and Cr. There also appears to be a correlation between high Ca and Sr content and high levels of U. Analysis of variance (ANOVA) indicates that the composition of bottled water can be distinguished primarily by the country of origin, over other factors including the geological environment of the source. This would suggest that composition reflects, and is biased towards, consumer preferences.

1. Introduction

The consumption of bottled water is increasing worldwide, as a result of the following factors: affordability, availability, perceived better taste than tap water, consumer connotations of higher social status (especially some brands) and the widespread conception that it contains fewer impurities (Misund et al., 1999; Ikem et al., 2002; Naddeo et al., 2008). In some countries, consumption of bottled water reflects health concerns surrounding the safety of public water supplies. The origin of bottled waters can be quite diverse and includes aquifers, springs, reservoirs (Chiarenzelli and Pominville, 2008) or even tap water (Naddeo et al., 2008). In many cases, the source of the water is not clearly specified on the label and some brands may use different sources in the same product (Pip, 2000).

EEC directive 2009/54/EC (2009) provides the legislation for the exploitation and marketing of natural mineral waters in the European Union. This directive provides criteria for labelling high concentrations of mineral salt content (>1500 $mg\ L^{-1}$), HCO_3^- (>600 $mg\ L^{-1}$), SO_4^{2-} (>200 $mg\ L^{-1}$), Cl^- (>200 $mg\ L^{-1}$), Ca (>150 $mg\ L^{-1}$), Mg^- (>50 $mg\ L^{-1}$), Fe (>1 $mg\ L^{-1}$), F^- (>1 $mg\ L^{-1}$), CO_2 (>250 $mg\ L^{-1}$) and Na (>200 $mg\ L^{-1}$). However, these need only to appear on the labels with general statements, such as "rich in mineral salts" or "contains sulphate" and do not need to include the actual concentrations. Depending on national regulations, only the major elements are analysed and reported on the labels; trace constituents are not always evaluated. In the UK, four sets of parallel statutory instruments (for England, Wales, Scotland and Northern Ireland) establish the natural mineral water, spring water and bottled drinking water regulations (UK Statutory Instruments, 2007, No. 2785 and amendment; UK Statutory Instruments, 2009, No. 1598). These regulations indicate the list of elements and species that must be determined during the process of recognition as natural mineral water (namely, BO_3^-, Cl^-, F^-, HCO_3^-, NO_3^-, NO_2^-, PO_4^{3-}, SiO_2, SO_4^{2-}, S^{2-}, Al, NH_4^+, Ca, Mg, K, Na, Ba, Br, Co, Cu, I, Fe, Li, Mn, Mo, Sr and Zn, as well as gross alpha and beta activities). It also regulates the maximum limits for certain constituents: 0.005 $mg\ L^{-1}$ Sb, 0.010 $mg\ L^{-1}$ As (total = organic + inorganic), 1.0 $mg\ L^{-1}$ Ba, 0.003 $mg\ L^{-1}$ Cd, 0.05 $mg\ L^{-1}$ Cr, 1.0 $mg\ L^{-1}$ Cu, 0.070 $mg\ L^{-1}$ CN^-, 5.0 $mg\ L^{-1}$ F^-, 0.010 $mg\ L^{-1}$ Pb, 0.5 $mg\ L^{-1}$ Mn, 0.0010 $mg\ L^{-1}$ Hg, 0.02 $mg\ L^{-1}$ Ni, 50 $mg\ L^{-1}$ NO_3^-, 0.1 $mg\ L^{-1}$ NO_2^- and 0.01 $mg\ L^{-1}$ Se. However, UK regulations contain specific requirements only for

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fluoride; there must be an indication on the label where concentrations of F^{-} are higher than 1.5 mg L^{-1}, as this is deemed not suitable for regular consumption by infants and children under 7 years old. Despite their natural origin, some of these species may be harmful to public health; Misund et al. (1999) concluded that determination of As, Ba, Be, Br, F, Sr and Tl is fundamental in terms of possible health impact. The same authors suggested that other elements should be included in regular analyses, such as Hg, Mo, Ra, Rn, Se, Th and U. The chemical composition of bottled waters may also be affected by handling operations after extraction from the source, for example by carbonation whereas packaging may cause changes due to leaching. Leaching of lead has been reported from glass bottles whereas the use of plastic bottles can lead to leaching of organic compounds into the water, especially at high temperatures (Pip, 2000).

Owing to the increasing interest in bottled waters, from both economic and sanitary points of view, several studies have been published dealing with their composition and quality. Misund et al. (1999) studied the variation of 66 elements in European bottled mineral waters, observing a wide variation due to specific national regulations and also to geological environments, of up to several orders of magnitude for the same elements. The authors also indicated that the concentrations of Pb, Na, K, Li, U and Zr observed in glass bottled waters were significantly higher than those found in plastic bottles. Naddeo et al. (2008) published a thorough study of 371 Italian bottled waters, based on the reported chemical composition and compared the results to Italian and international legislations (European Union, Spain, United States Environmental Protection Agency, Canada, Codex Alimentarius and World Health Organization). They drew attention to the contradictory and incomplete recommendations of the different sources of legislation, e.g. whilst the concentration of some essential metals is limited, regulations do not provide guidance regarding other toxic elements, such as Mo or U. Other examples are the comparisons of tap and bottled waters that have been performed in Saudi Arabia (Al-Saleh and Al-Doush, 1998) and in Croatia (Fiket et al., 2007). Because of the wide variation in composition of natural waters, even in the absence of pollution sources, it is necessary to generate a reliable database including as many elements/species as possible to help create more meaningful standardised limits.

The main objectives of this work are to characterise the chemical composition of popular bottled waters available in the UK and continental Europe, and to carry out multivariate statistical analyses of the data to assess the variability of the composition across the brands tested with respect to any geographical differences found. This work extends and differs from previous studies on British and European bottled waters in its statistical and multivariate approach to the comparison and classification of waters. It also investigates the underlying factors affecting the variability in their composition.

2. Material and methods

Commercial or brand names are not stated here; each sample is identified by its country of origin in Table 1 (the location of the source(s) can be found in Fig. S1 of the Supplementary Data). Table 1 also describes the geological setting of sources; the waters

Table 1
Classification of bottled waters.

Brand code	Origin	Source Characteristics	Bottled water type	Container type
1	Czech Republic	Sedimentary: chalk with marl and limestone	Still	PET
2	Czech Republic	Metamorphic: phyllite and schists	Sparkling	PET
3	Czech Republic	Sedimentary: sandstone, quartzite and siltstone	Sparkling	PET
4	Czech Republic	Igneous: granite	Sparkling	PET
5	Czech Republic	Igneous: granite and granodiorite	Sparkling	PET
6	Czech Republic	Sedimentary: sandstone, quartzite and siltstone	Still	PET
7	Czech Republic	Metamorphic: phyllite and schists	Still	PET
8	Czech Republic	Igneous: granite and granodiorite	Sparkling	PET
9	Slovakia	Sedimentary: sand with subordinate mud and gravel	Sparkling	PET
10	Slovakia	Sedimentary: sand with subordinate mud and gravel	Still	PET
11	Finland	a- Sedimentary: diamicton b- Sedimentary: sand with subordinate gravel	Sparkling	PET
12	Finland	Sedimentary: clay with subordinate silt	Sparkling	PET
13	Finland	Sedimentary: sand with subordinate gravel	Sparkling	PET
14	France	Metamorphic: calc-schists	Still	PET
15	France	Igneous: basanite with subordinate trachytoid and tephrite	Still	PET
16	France	Sedimentary: sand with subordinate impure carbonate sedimentary rock and clay	Sparkling	PET
17	France	Sedimentary: sand with subordinate impure carbonate sedimentary rock and clay	Still	PET
18	France	Sedimentary: clay with subordinate sand and gravel	Still	PET
19	France	Sedimentary: impure carbonate sedimentary rock with subordinate clay, sandstone and dolomite	Still	PET
20	France	Sedimentary: chalk with subordinate impure limestone, carbonates and sand	Still	PET
21	France	Sedimentary: sandstone with subordinate conglomerate and impure limestone	Sparkling	Glass (Green)
22	France	Sedimentary: sand with subordinate clay and gravel	Sparkling	PET (Green)
23	Germany	Sedimentary: shale with subordinate wacke and sandstone	Still	PET
24	Germany	Sedimentary: limestone with subordinate impure carbonate sedimentary rock and claystone	Sparkling	PET
25	Germany	Sedimentary: limestone	Sparkling	PET
26	Italy	Metamorphic: gneiss and mica-schists	Still	PET
27	Italy	Metamorphic: calcarenites and calc-schists	Sparkling	PET (Green)
28	UK (England)	Sedimentary: mudstone, siltstone and sandstone	Still	Glass
29	UK (England)	Sedimentary: shale-mudstone	Still	PET
30	UK (England)	Sedimentary: mudstone, siltstone and sandstone	Still	PET
31	UK (England)	Sedimentary: limestone	Still	PET
32	UK (England)	Sedimentary: sandstone	Still	PET
33	UK (England)	Sedimentary: sandstone	Still	PET
34	UK (Scotland)	Sedimentary: sandstone	Still	PET
35	UK (Scotland)	Igneous: pyroxene andesite	Still	PET
36	UK (Scotland)	Sedimentary: sandstone with subordinate mudstone and siltstone	Still	Glass
37	UK (Scotland)	Igneous: pyroxene andesite	Sparkling	PET (Green)