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Antimony and PET bottles: Checking facts

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HIGHLIGHTS

- Most bottled water is sold in polyethylene terephthalate (PET) bottles.
- Antimony is present in bottled waters because used as a catalyst in PET production.
- Antimony concentrations are usually below regulated values.
- Faulty analytical practices and lack of well-designed studies in existing literature.
- Progress needs a better understanding of the structure of the bottle PET polymer.

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ABSTRACT

Over the last 30 years, bottled water has gained in popularity reaching high sales world-wide. Most of this water is sold in polyethylene terephthalate (PET) bottles. About 15 years ago, the presence of antimony in water in those PET bottles raised concerns and studies on the subject have been regularly published since then. This review aims to evaluate whether the use of good analytical practices and the correct design of these studies support the accepted facts (i.e., PET is the origin of antimony presence in bottled waters, antimony concentrations are usually below regulated values, temperature increasing favours antimony leaching). The detailed analysis of published data has confirmed these facts but has also revealed frequency of faulty analytical practices and a lack of well-designed studies. A better understanding of the structure of PET polymer in the bottles, coupled with statistically-robust antimony release experiments, is required to progress in the field.

Contents

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1. Introduction

Polyethylene terephthalate (PET) is a thermoplastic polymer widely used in synthetic fibres, films and bottles. PET is the most popular material for food and beverage packaging. Some 485 billion PET bottles were produced in 2016 and it is forecast that 583 billion will be produced in 2021 (www.statista.com). PET is the material of choice because it is transparent and shatterproof, lightweight and CO_2 impermeable. PET is used for carbonated soft drinks, mineral water, edible oils, juices and sauces, flasks, and transparent blister packaging. PET bottles are produced by injection stretch blow moulding. PET is also thermoformed to make trays and pots with higher thermal stability used for cooking and heating foods in conventional and microwave ovens (Gilbert, 2017).

Antimony has been associated in the public eye with bottled water following the publication of articles by Shotyk and co-workers (Shotyk et al., 2006; Shotyk; Krachler, 2007) where these authors reported significant concentrations of antimony in bottled waters and linked them to the use of antimony in the production of PET bottles. Antimony trioxide (Sb_2O_3) —together with antimony acetate and antimony glycolate— is the most frequently used catalyst in the production of PET resin and antimony remains in the objects produced. In fact, studies on the presence and release of antimony from PET had preceded the studies from Shotyk and co-workers but they remained in the technological and scientific literature with no significant public impact (Ashby, 1988; Fordham, 1995; Thompson et al., 1997).

Since the publication of Shotyk's papers, similar articles, essentially repeating previous studies and reaching similar conclusions, frequently appear in the scientific literature. Although some compilation tables have been published (Welle and Franz, 2011; Bach et al., 2012; Rungchang et al., 2013; Kiyataka et al., 2018) and, in general, authors cite previous papers in introductions and discussions, to our knowledge no critical evaluation exists where analytical quality and methodological issues have been carefully considered. The objective of this work is twofold: 1) the evaluation of published studies in order to check whether conclusions were sound (i.e., the evaluation of "what is known") and 2) the identification of aspects that still merit research (i.e., the identification of "what we need to know"). It is important to mention that the ensemble of studies published on this subject is one of the largest existing sets of experiments dealing with the release of a chemical element from a manufactured product. Although they concern only one chemical element, antimony, and one type of polymer, PET, they contribute to the general understanding of the potential (eco)toxicity of plastics while in use in consumer products, and when disposed of in a controlled way or when mismanaged.

2. Working method and reading guide

Although this study is neither a compilation nor a narrative review, a comprehensive compilation of published papers was the necessary initial step, with the collection of 192 articles. Secondary sources were avoided. The second step was the careful reading of all collected articles and the identification of those that contained pertinent information. These were then classified in four types depending on their objective (Fig. 1): studies where antimony concentrations were determined in bottled water with the objective of checking compliance with regulations (type A); studies where antimony concentrations were measured in bottled waters in order to study the parameters driving them (type B); experiments studying the release of antimony from PET polymers with extractants of known composition (type C); multielement studies describing global surveys of mineral waters often launched by different Geological Surveys (type D). Type A and type B studies often overlap. The third step consisted in the extraction of analytical and methodological information from the studies and the summation of it in table form (Tables 1–3). Finally, relevant information concerning the facts to check (section 4) was systematised in tables SI1 to SI5.

3. Methodological issues

3.1. Analytical questions

Published studies include measurements of antimony concentrations in two types of matrices: water and PET bottles. Information about chemical analysis, such as measuring technique, detection limits, use of certified reference materials (CRMs), etc., can be found in Table 1 for types A and B studies, Table 2 for type C and Table 3 for type D. Similar information for measurements of antimony concentrations in PET is given in Table SI7.

The two main aspects to consider concerning analytical quality are (i) whether the limit of detection (LOD) of the analytical technique used is adapted to the analyte concentration to be measured and (ii) whether the accuracy of the concentration values found has been assessed. Both detection limits and values obtained for CRMs need to be adequately reported.

Even if the definition used is not always given, detection limits are often reported in the studies considered. Unfortunately, in many cases they are too high. Even if antimony concentrations are expected to be higher in waters contained in PET bottles than in natural waters, detection limits need to be low enough to be able to measure antimony concentration levels in source waters. This means detection limits of a few ng per litre. This LOD is perfectly achievable by ICP-MS (Birke et al., 2010) or voltammetry (Quentel and Filella, 2002). With a few exceptions, it is striking to observe that often older studies used analytical methods with lower and, thus, more adequate LODs, than more recent ones. Studies with insufficient LODs appear in italics in Tables 1 and 3 It is important to emphasise that the use of inadequate LODs not only precludes the measurement of low antimony concentrations but also leads to wrong conclusions. For instance, Greifenstein et al. (2013) studied the effect of temperature and storage of military packaged water in PET containers with a method detection limit (MDL) of 1 $\mu g\ L^{-1}$ and

Categories	Objective	Studies
Type A Sb concentrations in bottled water	Regulatory	Table 1
Type B Sb concentrations in bottled water	Study effects (time, T, etc.)	Table 1
Type C Release experiments with controlled extractants	Study effects (time, T, etc.)	Table 2
Type D Multielement measurements in bottled water	Variable, often study effect of geology on composition	Table 3

Fig. 1. Categorisation of published studies. Many studies are simultaneously types A and B.

failed to detect antimony after 70 days in 70 °C heated waters in contradiction with many other studies. Similarly, Kiyataka et al. (2018), with a technique having a limit of quantification (LOQ) of 20 μg L^{-1}, did not manage to measure any concentration and concluded that antimony in PET does not migrate into soft drinks. Particular mention needs to be made of studies where ‘new’ analytical methods are developed with insufficient LODs for the intended application. This is the case for instance of de Jesus et al. (2016) who developed a method with a LOD of 1 μg L^{-1} and a LOQ of 3.7 μg L^{-1} and, when they did not manage to measure any antimony concentration in bottled waters or any release, concluded that “leaching is really very low or even does not occur”. These and other studies with all samples below detection limit (e.g., Baba et al., 2008) or where the high LOD made the studies too unreliable (e.g. Bach et al., 2013; de Andrade et al., 2017; Roje; Sutalo, 2019) have not been considered further.

Accuracy checking relies on the use of CRMs that contain the analyte in the concentration range of the samples. The situation is unsatisfactory concerning the use of CRMs, with most recent studies not mentioning their use. This is extremely worrisome and calls for reviewers and editors to take some responsibility. It is also necessary to remember that CRMs need to be adapted to the analyte concentrations expected. As shown in Tables 1 and 3, many of the CRMs used had concentrations orders of magnitude higher than those needed.

Table SI7 gathers published values for antimony in PET together with information on the analytical method (i.e., method of digestion of the solid sample, measuring technique, use of CRMs). Unless non-destructive techniques such as Neutron Activation Analysis (NAA) or X ray fluorescence (XRF) are used, prior digestion of the samples is required. Takahashi et al. (2008), after obtaining unsatisfactory results with the usual mixture used to digest PET (HNO_3, H_2O_2), tested different methods and concluded that the use of 98% H_2SO_4 was the best way to digest PET. Many authors, however, continued to use the $HNO_3{-}H_2O_2$ method (Table SI7). Only a few authors mention the use of a CRM.

Two further issues concerning antimony analysis need to be mentioned: the possible contamination of samples by scientific equipment (Birke et al., 2010; Shotyk et al., 2005) and instrumental memory effects in the case of ICP-MS (Hu et al., 2007), the most widely used technique. Most of the authors seem to be unaware of these potential sources of analytical problems.

3.2. *Experimental design*

An adequate study design is a prerequisite of any sound investigation. Some issues that need to be considered in the evaluation of the results published are discussed in this section.

3.2.1. *How much Sb was in the water before being bottled?*

In order to ascertain whether antimony concentrations measured in waters contained in PET bottles are due to release from the PET polymer, it is necessary to know how much antimony was present in the water before being bottled. However, almost none of the studies in Table 1 includes this information (see column 6 in Table 1). Although, it might be correctly argued that it is not needed if the aim of the study is just to check whether antimony concentrations fulfil regulations (Type A studies), it is indispensable when the objective is to evaluate the effect of storing water in PET containers and understand the processes involved (Types B and C studies) or extract geochemical information (type D studies). Natural antimony concentrations in waters are not negligible, quite variable, and dependent on geology. For instance, measured dissolved antimony concentrations in European stream water ($n = 807$) ranged over three orders of magnitude from <2 ng L^{-1} (LOD) to 1.21 μg L^{-1} with a median value of 70 ng L^{-1} (Salminen et al., 2005). Moreover, sometimes natural spring waters have much higher concentrations (e.g. up to 3 mg L^{-1} in spring water in the Eiblschrofen Massif in Austria (Millen, 2003)) than the 2.2 ng L^{-1} of the pristine groundwater used for comparisons by Shotyk et al. (2006). Some authors have tried to circumvent this lack of information by comparing values of water in PET bottles with values in glass bottles but it is surprising that so few attempts have been made with knowledge of the composition of the water source.

3.2.2. *Time zero?*

Linked to the previous point, the question of time zero in release

Table 1

Published studies on Sb in bottled water (Type A and B studies, see Fig. 1 for classification). Methodological and analytical information. Entries in ascending chronological order. Studies with insufficient detection limits in italics.

Origin	Number of samples	Volume of bottles	Days after manufacturing	Sb concn. in polymer	Sb conc. before bottling?	CRM?^{x}	CRM in the needed conc. range?[a]	Limit of detection[b]	Technique[c]	Reference
Canada and Europe	15 brands (Canada), 48 (Europe) in PET 1 brand from Canada in PP 3 brands from Germany both PET and glass	not given	information not given	yes, in 2 bottles	not given but see Table 2	SLRS-4	yes	0.03 ng L^{-1}	ICP-SF-MS	Shotyk et al. (2006)
28 countries	132 brands (includes samples in Shotyk et al., 2006)	not given	information not given	no	not given	SLRS-4	yes	0.35 ng L^{-1}	ICP-SF-MS	Shotyk and Krachler (2007)
Arizona, US	9 brands; summer 2006; all PET, some clear, some with a blue tint	not given	information not given	yes, in one bottle	not given	NIST 1640, 1643e, SRLS-4	yes (SRLS-4)	4 ng L^{-1} (LOD) 28 ng L^{-1} (MRL)	ICP-MS	Westerhoff et al. (2008)
Hungary	10 brands of still and sparkling water, “PET but in some cases glass”	0.33–2 L	yes	yes	no	SLRS-4	yes	0.7 ng L^{-1}	ICP-SF-MS	Kereskes et al. (2009)
US	14 brands (31 bottles), including carbonated and flavour added; not all PET bottles; 14 brands are PET	given but not considered in the discussion	information not given	no	not given	NIST 1643e	no	0.5 ng L^{-1} (LOD) 1.0 ng L^{-1} (LOQ)	ICP-MS	Andra et al. (2012)
Lebanon	*8 brands*	*0.5, 1.5, 2 L*	*information not given*	*yes*	*no*	*not mentioned*	-	*200 ng L^{-1}*	*AAS*	*Hureiki and Mouneimne (2012)*
UK	47 bottles (water and soft drinks)	bottle size effect discussed	information not given	yes	not given	TM-DWS.2	no	0.02 no units	ICP-MS	Tukur et al. (2012)
Saudi Arabia	*7 brands (6 PET, a glass)*	*not given*	*information not given*	*no*	*no*	*not mentioned*	-	*not given*	*ICP-MS*	*Mortula and Ahmad (2013)*
South Africa	*6 brands (6 sparkling and 6 still waters)*	500 mL	*information not given*	*yes*	*no*	*NIST 1640a*	*no*	*109.7 ng L^{-1} (IDL)*	*F-AAS*	*Brandão* et al. *(2014)*
Iran	*5 brands*	*not given*	*information not given*	*no*	*no*	*not mentioned*	-	*not given*	ICP-OES	Molaee Aghaee et al., 2014
Spain	*3 brands (1 clear, 1 light blue, 1 dark blue); non-carbonated*	*not given*	*information not given*	*yes*	*no*	*not mentioned*	-	*110 ng L^{-1} (LOD) 140 ng L^{-1} (LOQ)*	HG-AFS	*Carneado* et al. *(2015)*
Mexico	12 brands of bottled drinking water, not carbonated, clear PET	all 500 mL	information not given	yes	no	not mentioned	–	5.65 ng L^{-1} (LOD) 19.01 ng L^{-1} (LOQ)	Dowex 1X8-100 prec., HG-AFS	Chapa-Martínez et al. (2016)
Qatar	53 brands from 15 countries PET and glass, carbonated and non-carbonated	250 mL to 1.5 L	information not given	yes	no	NIST 1640a	no	0.1–1.0 ng L^{-1} (IDL)	ICP-MS	Rowell et al. (2016)
Qatar *but includes foreign brands*	*22 brands (noncarbonated)*	*330 to* 550 mL[d]	*18–226*	*no*	*no*	*not mentioned*	-	*not given*	*ICP-MS*	*Al-Otoum* et al. *(2017)*
Poland	*35 bottles from 19 brands* *18 PET, 16 glass,* 1 Al *PET: 6 bright blue, 2 dark blue, 3 colourless, 1 bright pink, 2 bright green,4 green*	*not given*	*information not given*	*no*	*not given*	*SLRS-5, TM-27.3, 1643d*	*none certified for Sb^{V} and Sb^{III}*	*Sb^{III}: 83 ng L^{-1} (LOD), 250 ng L^{-1} (LOQ)* *Sb^{V}: 38 ng L^{-1} (LOD), 111 ng L^{-1} (LOQ)*	*HPLC-ICP-DRC-MS*	*Marcinkowska* et al. *(2017)*
Spain but includes foreign brands	*16 brands (11 Spain)* *5 colourless, 10 blue, 1 green*	*given but not considered in the discussion*	*information not given*	*no*	*no*	*not mentioned*	-	*40-80 ng L^{-1} (LOD)* *130-260 ng L^{-1} (LOQ)*	*ICP-MS*	*Payán* et al. *(2017)*
China	10 brands	given	information not given	no	no	not mentioned	–	50 ng L^{-1}	ICP-MS	Qiao et al. (2018)
Turkey	1 brand	0.5, 1.5, 5.0 L	information not given	yes	no	not mentioned	–	3 ng L^{-1} (LOD) 9 ng L^{-1} (LOQ)	ICP-MS	Dogan and Cebi (2019)

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[a] Sb concentrations in certified reference materials: NIST SRM1640: 13.79 μg kg^{-1}; NIST SRM1640a: 5.105 μg L^{-1}; NIST SRM1643d: 54.1 μg L^{-1}; NIST SRM1643e: 56.88 μg L; NRC SLRS-3: 0.12 μg L^{-1}; NRC SLRS-4: 0.23 μg L^{-1}; NRC SLRS-5: 0.3 μg L^{-1}; NWTM-DWS.2: 3.21 μg L^{-1}; NWTM-27.3: 1.49 μg L^{-1}

[b] When reported, the type of detection limit is given. However, a given abbreviation does not necessarily imply the same way of calculating the parameter. Abbreviations: see corresponding table.

[c] Abbreviations: see corresponding table.

[d] “For the 500 and 550 mL, the Sb content were calculated into 330 mL to unify the volume”.

experiments (types B and C studies) arises. Time zero is usually defined as the time when the measurements start but authors rarely consider when the water was bottled (see column 4 in Tables 1 and 3). This means that initial conditions are not comparable among experiments and that, in case of an H type of kinetics (i.e., fast release followed by a plateau), what are considered to be as starting conditions might correspond to the plateau stage and no further increase be observed when, in fact, antimony had been released from PET before measurements started.

3.2.3. Bottle volume and surface area

The common practice in dissolution and releasing studies is to refer the amount released to the surface from which the substance has been delivered (Stumm, 1992). This requires knowing the specific surface area of the solid, often measured by the BET (Brunauer—Emmett—Teller) technique. However, in the studies considered here, results are nearly exclusively given as antimony concentrations in the bottled water and not as antimony released per surface unit of the polymer. The immediate consequence is that the results obtained in different studies, or even within a given study, cannot be directly compared unless all bottles had the same surface and volume. Moreover, this information is often lacking (see column 3 in Tables 1 and 3).

The effect of the ratio surface area:volume of water exposed was considered in some initial studies: “antimony release is probably proportional to contact area to liquid volume ratio” (Westerhoff et al., 2008), “the Sb concentration of the stored water depends on the PET surface/water volume ratio, therefore the storage in smaller bottles results in higher Sb concentration” (Keresztes et al., 2009) and similar statements can be found in a few later studies (Welle and Franz, 2011; Hureiki; Mouneimne, 2012; Tukur et al., 2012; Dogan and Cebi, 2019). In spite of them, however, the common procedure is the direct comparison of antimony concentrations without considering the volume and shape of the bottles.

The few authors that consider this factor give different estimations of the surface:volume ratio (Table SI6) and none explains how the surface was estimated. Surface estimation is not a straightforward question, particularly when the bottles are not perfect surface of revolution objects. Correct estimations are even more difficult nowadays because most bottles have a self-standing petaloid-shaped base (Hanley et al., 2006).

3.2.4. On choices and statistics

The number of bottles in many studies is often low and repeatability is rarely assessed. Inadequate reporting makes it sometimes difficult to distinguish between number of bottles and number of brands. It is not unusual to extract conclusions from only one brand of water and one experiment. This is the case, for instance, of temperature dependence in Westerhoff et al. (2008) with the choice of the brand studied being driven by being the one that “contained the highest initial antimony and showed a propensity to release antimony over time” and thus ignoring the potential response of seven other brands that showed no statistically-significant leaching at ambient temperature. The effect observed has been widely cited as being the “general behaviour” of release of antimony from PET bottles in subsequent studies. Similarly, only one brand was chosen by Chapa-Martínez et al. (2016) to perform leaching experiments on the basis that “it contained the highest initial antimony concentration in PET and showed a propensity to release antimony over time” (yes, exactly the same text as in Westerhoff et al. (2018)). In practice, it looks as if a lot of data were available but, when examined more closer, frequently accepted facts are based on very few, though repeated, cited observations.

4. Facts?

4.1. Sb in PET is the source of Sb in bottled waters

Several authors have measured the antimony content in PET bottles and a relatively narrow range of concentrations has been reported (Table SI7). The measured concentrations mostly correspond to the amount of antimony reported to be used as catalyst (Duh, 2002).

That the origin of the antimony concentrations measured in bottled waters is linked to the presence of antimony in the bottle material has been repeatedly observed by comparing antimony contents in water contained in PET bottles with water in bottles made of other materials, usually glass (Table SI1). Even if glass might contain low amounts of antimony (Shotyk et al., 2006), presumably due to its use as opacifier (Turner and Filella, 2020), and that some glass bottles can leach a lot of antimony (e.g., Reimann et al. (2010) observed the highest leaching value from a glass bottle), existing data clearly points to a link between antimony concentration in water bottled in PET and the antimony in the polymer. Comparisons with water stored in other type of plastics such as polypropylene (PP) and low density polyethylene (LDPE) (Shotyk et al., 2006; Shotyk and Krachler, 2007), high density polyethylene (HDPE), polycarbonate (PC) and polystyrene (PS) (Andra et al., 2012) confirm it.

Authors have failed to find correlation between antimony concentrations in water and in the PET of the corresponding bottles. Moreover, the response variability of antimony released from PET has also been mentioned by different authors (Shotyk et al., 2006; Takahashi et al., 2008; Keresztes et al., 2008; Fan et al., 2014). Some variability can be explained by the many confounding, non-controlled factors in play such as not considering the initial concentration in the source water or the time elapsed between bottling and measurement, etc. (see section 3.2) but, probably, not all variability is accounted for by these factors. While Takahashi et al. (2008) suggested that it had to do with the alteration of the bottles and was not related to the initial PET in the bottles, Fan et al. (2014) attributed it to “different quality of PET materials, which may vary depending on raw materials as well as technology used for PET production”.

Possible differences in the structure and properties of the PET of the bottles have not received much attention. Some authors have considered bottle colour with no conclusive results (Table SI2). A few authors have mentioned the existence of different types of PET but not linking them to any structural polymer difference. For instance, Reimann et al. (2010) made the distinction between hard and soft PET observing more leaching from soft but without giving any exact definition of the categories. Takahashi et al. (2008) categorised the bottles depending on their use and found that the antimony concentration in PET depended on the type of use with bottles for noncarbonated drinks, pressure-tight bottles for carbonated drinks and heat-resistant and pressure-tight bottles for semi-sparkling drinks having statistically-significant ($p = 0.002$, χ^2 test) more antimony than heat-resistant bottles and bottles for freezing. Later, Rungchang et al. (2013) observed different leaching behaviour in different types of Takahashi’s classification.

Different behaviour might be related to the degree of crystallinity in combination with polymer orientation. PET can be in a semi-crystalline or in an amorphous state with a glass transition temperature (T_g) between 67 and 80 °C, value that depends on information source but also on the dependence of T_g on polymer crystallinity, with higher crystallinity having higher T_g. Crystallinity in PET is induced by thermal and by stress or strain induced crystallization. While quenching the melt quickly results in completely amorphous PET, thermally induced crystallization occurs when the

Table 2

Published migration experiments from PET to water or other extractants of initial known composition (Type C studies, see Fig. 1 for classification). Entries in ascending chronological order.

Origin	Samples	Extractant	T/^{o}C	pH	Exposure time	Analytical parameters	Observations	Reference
2 L PET bottles	2 dm^2 of 350 μm thick PET bottle wall	100 mL of water, 3% acetic acid, 15% and 50% ethanol, olive oil	40	See column 3	10 days	No information	Water: 0.33 mg dm^{-2} Acetic acid: 0.53 mg dm^{-2} 15% ethanol: 0.38 mg dm^{-2} 450% ethanol: 0.57 mg dm^{-2} Olive oil: 0.7 mg dm^{-2}	Ashby (1988)
PET bottles	3 bottles of unknown volume	Deionised water	probably room	–	unknown	See Table 1	Sb concentrations: 134–195 ng L^{-1}	Shotyk et al. (2006)
PP and PET bottles	3 bottles of unknown volume: one PP, one Canada PET, one Germany PET	Pristine groundwater	probably room	–	6 months	See Table 1	Initial Sb concentrations: 1.8 ng L^{-1}, kept in Canada PET: 26.6, in Germany PET: 281	Shotyk and Krachler (2007)
A. 5 bottles from China/Japan B. 3 of them	A. 2 × 2 mm^2 pieces	A. 2.0 g MQ water B. 4% acetic acid, 20% ethanol, humic acid	40	A. 3.5 –4.5 B. See column 3	30, 45 days	See Table 1	- Sb leached in water: 0.874–4.40 μg kg^{-1} - Behaviourvariable depending on sample	Takahashi et al. (2008)
PET bottles bought in Arizona, US Only 2 bottles out of 9 studied	A. Bottle “that contained the highest initial Sb concentration in PET and showed a propensity to release Sb over time” B. 9.4 × 9.4 cm (88 cm^2) pieces from 2 PET and 1 HDPE bottles	A. 1 L glass bottle containing nanopure water B. 1 L nanopure water	A. 22, −20, 80, UV B. 80	A. 6.3, 7.3, 8.3	A. 48 h B. 10 days	See Table 1	A. No effect: pH, freezing; increase: 80 °C, UV B. 4x more Sb released by clear PET than blue PET; no release from HDPE	Westerhoff et al. (2008)
PET (and other polymers) bottles from recycled materials, bought US	7 new bottles, washed and unwashed	20 mL ultrapure water added	boiling water, ice-cold added, microwavein-car storage	4	24 h at 25C after each treatment	ICP-MS 20 mgL^{-1} (LOD), CRM used	- Sb increased leaching: heating, microwave - Smaller increase: low-pH water, outdoor UV radiation, in-car storage - Cooling: no effect - No statistics	Cheng et al. (2010)
hard PET, soft PET, glass bottles	126 PET and glass bottles, rinsed	unknow volume bottles filled with demineralised water and HNO$_3$	not given	3.5, 6.5	2, 3, 4, 5, 15, 30, 56, 80 days	See Table 3	- Sb leaching from PET - Effect “almost” pH independent - Concentrations still increasing after 150 days - Less leaching from hard than soft PET bottles	Reimann et al. (2010)
1.5 L PET soft drink bottle	5 1 cm × 5 cm strips extracted simultaneously	3% acetic acid	30–150	See column 3	1 h repeated extractions at 100 bar	ICP-MS 1 μg L^{-1} (LOD)	- Results in graphic form, calculation of diffusion coefficients and Arrhenius parameters - No results <90 °C because concentrations BDL	Welle and Franz (2010)
PET and glass bottles	3 PET, 37 glass bottles, rinsed	1.5 L bottles filled with demineralised water and HNO$_3$	2, 22, 45, 60, 80	3.5	1 week	ICP-MS In Reimann et al. (2010)	Median values: 2 °C, 0.00276; 22 °C, 0.0117; 45 °C, 1.24; 60 °C, 2.75; 80 °C, 15.8. All μg L^{-1}	Reimann et al. (2012)
PET and bottles	A. 10 500 mL PET bottles different colours and brands B. 8 PET bottles C. 2 green, colourless glass 750 mL bottles	A. 300 mL water B. Unknown C. 500 mL deionised water	A. 40, 60, 80 B. 60 C. 60	–	A. 6, 24, 48 h B. 6 h C. 6, 24, 48 h	See Table 1	- Confusing experiment design and reporting - Much higher increase at 80 °C - Very different behaviour depending on bottle	Tukur et al. (2012)
6 brands representative of different PET types categorised by the authors as in Takahashi et al., (2008)	2 cm × 3 cm pieces	15 mL simulant (MQ water, 4% acetic acid, 50% ethanol)	25, 40, 55, 70	See column 3	440 days	GF-AAS 8 ng L^{-1} (LOD), CRM not mentioned	- Results in graphic form, calculation of diffusion coefficients and Arrhenius parameters - No apparent release in MQ water at 25 °C; significant effect of acidity, alcohol and temperature	Rungchang et al. (2013)
PET bottles, number and origin unknown	12 cm^2 pieces	20 mL simulant (distilled water, 3% acetic acid, 10% 20% ethanol, olive oil, sunflower oil)	40	See column 3	10 days	ICP-MS 3 ng L^{-1} (LOD) 10 ng L^{-1} (LOQ)	Final concentrations.: water and acetic acid “not statistically different” (test not given); oils, no measurable concentration increase; ethanol higher release	Sánchez-Martínez et al. (2013)
PET bottles (16 brands)	PET bottles	MilliQ water	A. 4, 25, 70 B. 70	6.0	A. 7 days B. 1, 2, 4 weeks	ICP-MS <0.10 ng L^{-1}	A. Median 4^{o}C: 3.18 ng L^{-1}, median 25 °C: 6.88 ng L^{-1} but ANOVA significant ($p < 0.05$) in only 6 brands of the 16. At 70 °C, “substantial	Fan et al. (2014)

						(LOD), CRM: SLSR-5	higher concentrations.” B. Increase significantly in 13 brands. Releasing rate decreased with storage time	
PET bottles that contained salty sauces[a]	Already used PET bottles	NaCl solutions (0.1–6.0%)	room	–	5 days	ICP-MS 1.96 ng L^{-1} (LOD), 12 ng L^{-1} (MRL) CRM not mentioned	The concentration of Sb leached decreased when NaCl concentration increased; graphical result, no stats	Leechart et al. (2015)
500 mL colourless PET bottles bought in Mexico containing non-carbonated water[b]	5 cm × 4 cm pieces of one bottle (2.1 g); choice: sample “that contained the highest initial Sb concentration in PET and showed a propensity to release Sb over time”	100 mL water in glass-caped vials bath shaker: 50 oscillations/min	25, 75	3, 7	5, 15 days	See Table 1	- Factorial experiment; no detailed results given - Release of PET increased at 75 °C but release was variable depending on bottle	Chapa-Martínez et al. (2016)
green PET bottles	0.5 dm^2 pieces	50 mL of each food simulant: 3% acetic acid, 10–95% ethanol	40, 60	See column 3	usually 10 days	ICP-MS 0.1 mg dm^2 (LOD)	Acetic acid, 40 °C: BDL Acetic acid, 60 °C: 0.23 μg L^{-1} Arrhenius calculation; parameters from Welle and Franz (2010): end of shelf-life: 0.01 μg dm^{-2}	Gehring and Welle. (2018)
PET and glass bottles	Used bottles	A. Commercial water (62), tap water (20), synthetics water mixture (7) B. Synthetic water mixture	A. Outdoor UV exposure B. Oven heating (9 PET, 2 glass)	–	A. 12 days B. 4 h	See Table 1	-PET concentrations higher than in glass but magnitude of change difference not statistically significant ($p > 0.05$) - In PET, significant differences among water types ($p < 0.01$)	Rowell et al. (2018)

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[a] Fish sauce, distilled vinegar, tomato ketchup, chili sauce. Not clear whether all used or only “the bottle that had the highest antimony concentration leaching”.

[b] Initial Sb range: 0.07–2.30 μg L^{-1}; median: 1.035 μg L^{-1}; weak negative correlation Sb in water:Sb in PET ($R^2 = 0.55$).

polymer is heated above T_g and not quenched rapidly enough. In this case, the polymer turns opaque due to a spherulitic structure generated by thermal crystallization aggregates of non-oriented polymers (www.sciencedirect.com/topics/chemistry/glass-transition-temperature).

The manufacturing process of bottles includes two steps. In the first step, the PET bottle preforms are injection moulded and cooled to room temperature so that the preform is virtually amorphous. In the second step, the preforms are re-heated to a temperature above T_g and stretch blow moulded into bottles. Blowing injected preform moulds may lead to molecular orientation by deformation (Göschel et al., 1996). A detailed description of these processes is outside the scope of this review but the complexity of the process suggests that PET bottles may have different properties which could potentially affect their behaviour vis a vis water contact, temperature, etc. The extent of crystallinity in bottles depends on the injection procedure and is not uniform in a bottle. Harvey et al. (2006) found that the extent of crystallinity depends upon the geometry of the bottle base and that there is an abrupt change from the amorphous state to the crystalline regions. The orientation in the middle of the foot is more circular and crystallization is less than the valley and the transition zone to the foot. The effect of polymer orientation on antimony release was investigated by Ashby (1988). He found a significantly higher level of migration from amorphous cast PET compared to a blown bottle.

The form in which antimony is present in PET can also play a role in the diffusion of the element inside the polymer and, consequently, its possible release in the water. Even when added as Sb_2O_3, antimony is present as the glycolate complex in the polymerisation step because Sb_2O_3 is soluble in the ethylene glycol medium (Biros et al., 2002). Extended X-ray absorption fine structure spectroscopy (EXAFS) showed antimony to be present in PET bottles, not as Sb_2O_3 (at least at the microscale, nanosized not excluded) but as either free antimony glycolate or antimony glycolate bound to the end group of the PET polymer (Takahashi et al., 2008). In some samples, Sb^{III} was partially oxidised to Sb^{V} but with no coordination change. Antimony diffusivity in the polymer will be different depending on whether is present as free Sb^{III}, micro- or nano-size free antimony glycolate or attached to the polymer. Moreover, Haldimann et al. (2013) mentioned that these antimony complexes are sensitive to hydrolysis. Martin et al. (2010) reported different results from Takahashi et al. (2008), the presence of clusters of Sb^{III} having the dimensions of 10 μm but they concern one PET bottle only.

Since antimony diffusivity will strongly depend on how antimony is present in the polymer and on the polymer characteristics, any progress clearly requires more studies in these areas.

4.2. Sb concentration increases in the bottled water with time

The effect of storage time has been the most studied variable. Although it is generally accepted that a general increase of antimony concentrations with time at ambient temperature is observed, a detailed consideration of the observations in Tables 2 and SI3 gives a more nuanced view, with no statistically significant increase observed in many cases. The robustness of this conclusion is, of course limited by the variability discussed in the previous section and, in spite of the high number of studies, by the limited number of bottles and brands really studied.

Does the release, if any, go “for ever”? This does not seem to be the case if we consider Fig. 2. This figure shows concentrations published in studies type A, B (initial concentrations) and D with reasonable LODs. It is remarkable that antimony concentrations fall within a relatively limited range of concentrations and this in spite of variable antimony source concentrations, variable bottle sizes,

Table 3

Published multi-elemental studies on bottled waters containing specific information concerning Sb (Type D studies, see Fig. 1 for classification). Methodological and analytical and information. Entries in ascending chronological order.

Origin	Number of samples	Volume of bottles	Days after bottling	Sb conc. before bottling?	Sb concn. in polymer	CRM?[a]	LOD[b]	Technique[c]	Reference
Europe	56 brands	no	information not given	no	no	not mentioned	2 ng L^{-1}	ICP-MS	Misund et al. (1999)
Sweden	33 brands but only 16 with Sb results, no mention of material	no	information not given	no	no	not mentioned	5 ng L^{-1}	ICP-MS	Rosborg et al. (2005)
Croatia	18 brands	yes	information not given	no	no	SLRS-4	1 ng L^{-1}	HR ICP-MS	Fiket et al. (2007)
28 countries	132 brands, mostly PET, no glass but some metal bottles	no	information not given	no	no	SLRS-4	not given	SF ICP-MS	Krachler and Shotyk (2009)
Italy	186 bottles of 158 different brands 157 not carbonated, 11 natural CO_2 content, 18 artificially carbonated 167 PET with PE caps, 19 glass 119 clear, 67 coloured	no	information not given	no	no	probably, like in Birke et al., (2010)	2 ng L^{-1}	ICP-MS	Cicchella et al. (2010); Dinelli et al. (2010)
Greece	61 still waters from 41 locations (57 PET, 4 clear glass)	no	information not given	no	no	See Birke et al., 2010	1 (IDL), 10 (RDL)	ICP-MS	Demetriades (2010)
Nordic countries	Norway (9), Sweden (9), Finland (2), Iceland (2) All but one, PET	no	information not given	no	no	See Birke et al., 2010	See Birke et al., 2010	ICP-MS	Frengstad et al. (2010)
Hungary	36 brands (not all PET)	most 0.5 L	information not given	no	no	See Birke et al., 2010	See Birke et al., 2010	ICP-MS	Fugedi et al. (2010)
Croatia	14 brands	0.5 L	information not given	no	no	See Birke et al., 2010	2 ng L^{-1}	ICP-MS	Peh et al. (2010)
Europe	294 pairs of water samples sold in glass and in PET	no	information not given	no	no	SLRS-4	1 ng L^{-1} (IDL), 10 ng L^{-1} (RDL)	ICP-MS	Reimann et al. (2010)
British Islands	85 bottles all non-carbonated 71 PET, 14 glass	no	information not given	no	no	not mentioned	not given	ICP-MS	Smedley (2010)
Kuwait	73 brands of non-carbonated (6 local); soft PET with a PE screw cap 20 carbonated (not from Kuwait); glass with a metal cap (13) or hard PET with a PE screw cap (7)	no	information not given	no	no	not mentioned	2 ng L^{-1}	ICP-MS	Al-Mudhaf and Abu-Shady (2012)
Serbia	16 brands in PET (not all Serbian) 3 brands in glass (all Serbian)	0.5 L 0.25, 0.33, 0.75 L	information not given	no	no	SLRS-4	30 ng L^{-1} (MDL)	ICP-MS	Ristić et al. (2012)
Poland	47 brands (46 PET, 1 glass) 32 still, others sparkling	given	information not given	no	no	NIST 1643e	50 ng L^{-1}	ICP-MS	Astel et al. (2014)
Europe	Continental Europe (27 brands), UK (10) All PET except 3 glass Still (21), sparkling (16)	no	information not given	no	no	SLRS-5	20 ng L^{-1} (LOD) 70 ng L^{-1} (LOQ)	ICP-MS	Felipe-Sotelo et al. (2015)

[a] Sb concentrations in certified reference materials: NIST SRM1643e: 56.88 μg L; NRC SLRS-4: 0.23 μg L^{-1}; NRC SLRS-5: 0.3 μg L^{-1}.

[b] When reported, the type of detection limit is given. However, a given abbreviation does not necessarily imply the same way of calculating the parameter. Abbreviations: see corresponding table.

[c] Abbreviations: see corresponding table.

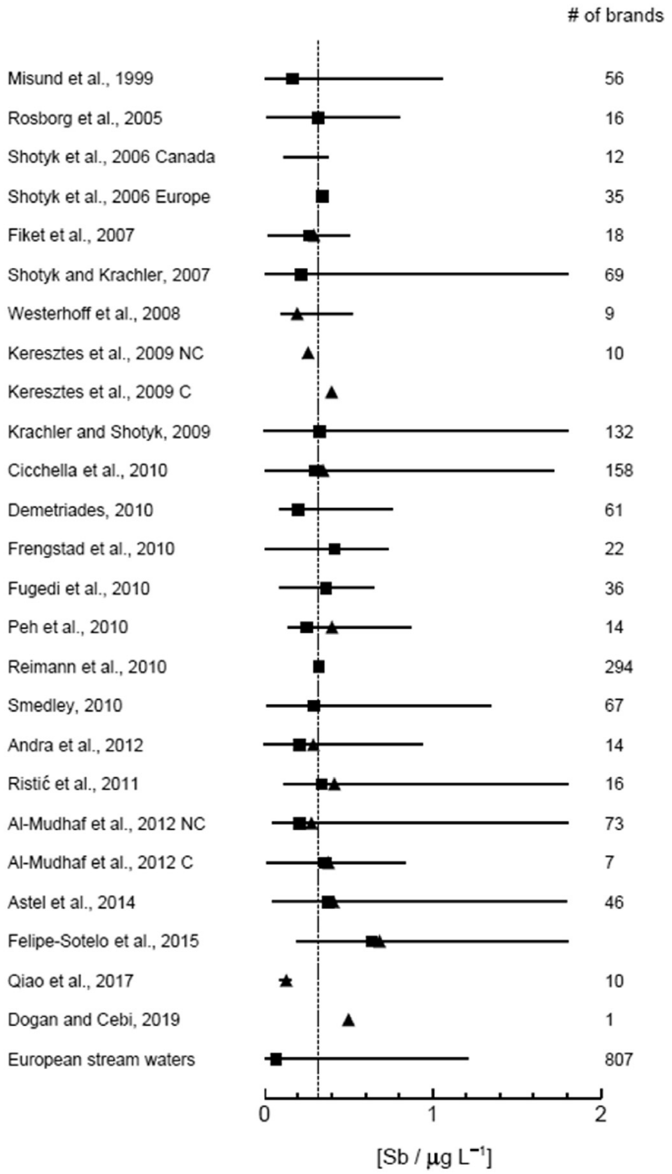

Fig. 2. Reported antimony concentrations in bottled waters. Studies are shown in chronological order. Details about the studies can be found in Tables 1 and 3 Values correspond to initial antimony concentrations in type B studies. Only studies with reasonable detection limits have been considered. Squares: median values, triangles: average values. The vertical dash line is at 320 ng L^{-1} (approximate median value). Studies where the range of values exceeded 2 µg L^{-1}: Shotyk and Krachler (2007), 2.57 µg L^{-1}, Krachler and Shotyk (2009), 2.57 µg L^{-1}, Al-Mudhaf et al. (2012), 2.27 µg L^{-1}.

unknown and variable times elapsed between bottling and measurement and unknown and uncontrolled storage conditions, etc. Median values (squares in Fig. 2) are often in the 300–400 ng L^{-1} range. This regularity, even if it has a limited statistical meaning, qualitatively points to some relatively fast initial release and a much slower leaching, if any, later. This hypothesis would fit with Keresztes et al. (2008) observations of antimony leaching from PET into water increasing rapidly during the first storage period and then migration reaching a "steady state". These authors described the increase of antimony concentration as a function of the storage time with a saturation curve reaching 0.7–0.8 µg L^{-1} in a year time span, but not exceeding 1 µg L^{-1} even after a three-year-long storage time. Fan et al. (2014) also observed that antimony release from PET bottles may become stable under long term storage. The fact that measured concentrations rarely exceed some few µg L^{-1} and that such concentrations represent a very tiny percentage of the total antimony present in the PET of a bottle suggests a very small diffusivity of the element in the polymer (if the mass flow is governed by Fick's law) at ambient temperature in the long term. Nothing excludes, however, an enhanced diffusion by alteration of the polymer with time by the contact of water or other factors such a temperature increase.

Leaching experiments into extractants of well-known composition (Type C studies) show a similar pattern, but interpretation of these experiments is not simple because the two-sides leaching can overestimate results since they may simply reflect a difference in contamination levels between inside and outside walls of blow moulded bottles.

If confirmed, the reason for the initial pulse of antimony release remains fully to be understood. It could easily be leachable antimony present at the surface of the bottles. Cheng et al. (2010) detected that more antimony was leached from unwashed bottles compared with washed ones. This, according to the authors, pointed to some antimony leaching coming from contamination during the production process. These authors recommended washing the new bottles before first use as a way of reducing the element contents in bottled waters.

4.3. *Temperature increase promotes Sb release from PET*

Temperature combined with storage time has been a widely studied factor. The accepted fact is that high temperatures favour antimony release from PET with a change in behaviour above 60–70 °C. As is often the case, this observation was already made in one of the first studies. Westerhoff et al. (2008), in a one brand one experiment exercise, adjusted a power function to describe the effect of temperature and its time dependent relationship on the migration of antimony from the PET bottles to the water. A higher rate of release was observed above 60 °C. These initial observations have been confirmed by many posterior studies (Table SI4). Following these results, storing PET bottles under high temperature conditions (e.g. car boots) has been not recommended.

The effect of temperature on the release of antimony from PET into extractant solutions has been modelled by applying the Arrhenius equation (Welle and Franz, 2011; Haldimann et al., 2013; Rungchang et al., 2013). The procedure starts with the calculation, at different temperatures, of the diffusion coefficient of antimony in the polymer from the regression of antimony concentrations at certain times against the square root of time. Then, the activation energy is calculated by:

$$D = D_0 e^{-E_0/RT} \qquad [1]$$

where D is the diffusion coefficient ($cm^2\ s^{-1}$), D_0 is the pre-exponential factor ($cm^2\ s^{-1}$), E_0 is the activation energy for the diffusion process ($kJ\ mol^{-1}$), R is the gas constant ($kJ\ mol^{-1}\ K^{-1}$) and T is the temperature (K).

Published results are represented in Fig. 3. In two cases, migration has been studied in specimens from PET bottles (Welle and Franz, 2011; Rungchang et al., 2013) while results by Haldimann et al. (2013) are for a different type of polymer, ovenable PET trays. Conditions of extraction were rather different from the usual release in bottle waters because they implied the use of extractors at high pressure, short extraction times and, in two studies (Welle and Franz, 2011; Haldimann et al., 2013), 3% acetic acid extractant solutions. The exact conditions of each study can be found in the legend of Fig. 3. The use of 3% acetic acid corresponds to the composition of one of the food simulants in EU regulation October 2011 (European Commission, 2011). The use of analytical

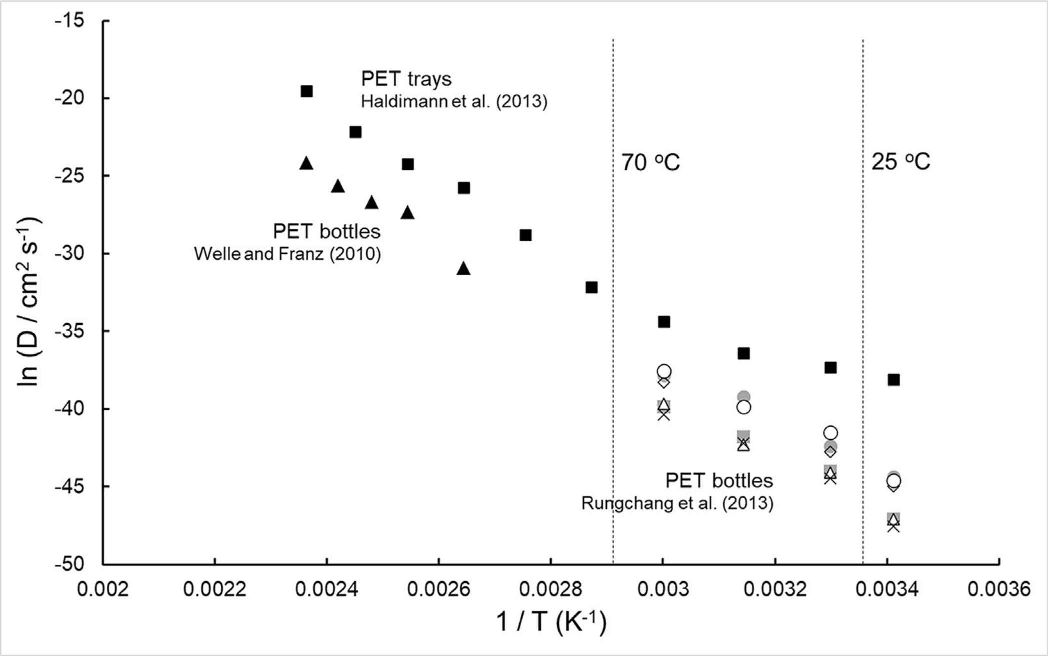

Fig. 3. Temperature dependence (Arrhenius plot) of the apparent diffusion coefficient of Sb in PET. Solid squares: data from Haldimann et al. (2013) for ovenable PET trays; extraction into 3% acetic acid in a pressurized chamber (50–90 bar). Each isothermal test lasted for 1, 5 and 24 h, or in some cases 46 h. Values often cited as Alt (2008), a conference communication, are included in this publication. Solid triangles: data from Welle and Franz (2011), migration into 3% acetic acid from a commercially available 1.5 L PET soft drink bottle (not used) in a Bücki extractor at 100 bar for 4 h renewing the solution every hour (or 1, 2, 3 h intervals for a total of 6 h, depending on sample). Below 90°C, the diffusion coefficients could not be detected because the Sb concentrations in the migration solutions were below de detection limit (2 µg L^{-1}). Non solid symbols: data from Rungchang et al. (2013) for PET bottles in contact with three types of simulants: MQ water, 4% acetic acid, 50% ethanol at 25, 40, 55 and 70 °C for 440 days.

techniques with high LODs precluded to obtain data at low temperatures where the release is small, if any (Welle and Franz, 2011).

Deviation of the linearity of the Arrhenius plot was observed at temperatures below the T_g of PET. In spite of the lack of linearity, pre-exponential factors, D_0, and activation energies, E_0, were calculated. Pre-exponential factors were very different; possible reasons are discussed in Welle and Franz (2010) and Haldimann et al. (2013). Activation energies were 188 kJ mol^{-1} (for temperatures above 45 °C only; Haldimann et al. (2013)), 189 kJ mol^{-1} (Welle and Franz, 2003) and in the range 129–136 kJ mol^{-1} (Rungchang et al., 2013). Activation energy variation with temperature, detectable when Arrhenius plots are non-lineal, are common in the kinetics of thermally stimulated reactions in the condensed phase (Vyazovkin, 2016). Thus, it is not astonishing to find it here when PET polymer property changes upon temperature (i.e., glassy ($T < T_g$) and rubbery ($T > T_g$) states) have a large influence on the migrant diffusion coefficients (Piringer and Baner, 2008). Moreover, PET undergoes significant hydrolysis above T_g (Allen et al., 1994).

The modelling approach used in the calculation of activation energies is based on a number of hypotheses, not the lightest hypothesis that the process is purely diffusive, with the water composition playing no role in the migration. Variable results from Rungchang et al. (2013) depending on extractant composition and observations in Section 4.6 suggest that this might not be the case.

When the values above are used to predict antimony concentrations in bottled waters, the type of curves obtained reach no plateau because the model does not consider any possible limit in the antimony supply other than the depletion of all antimony present in the polymer. This shape does not entirely correspond to experimentally-measured kinetics (Section 4.2).

According to regulation EU October 2011 (European Commission, 2011), time and temperature conditions for accelerated migration tests simulating long-term applications should be calculated by using the Arrhenius equation and a default activation energy of 80 kJ mol^{-1}. This means that, for real contact times exceeding 30 days at room temperature, the material needs to be subjected to an accelerated test for a maximum of 10 days at 60 °C (Gehring and Welle, 2018). These authors found that these conditions overestimate migration after storage for 365 days at 23 °C by a factor of 23 and that, therefore, they are not applicable to migration from PET.

4.4. Sb concentrations in PET bottled waters are below regulated values

Antimony concentrations in waters contained in PET bottles rarely exceeds regulated values for drinking waters (Fig. 3). However, although in many type A studies, authors compare measured concentrations with values regulated for drinking water, they forget that, in many countries, drinking water values no longer apply once the water is packaged. This is the case for the European Union where the maximum admissible concentration for drinking water is 5 µg L^{-1} but 40 µg kg^{-1} (which translates into 40 µg L^{-1}) applies for food in plastic containers. In the U.S., while tap water is regulated by the U.S. Environmental Protection Agency (EPA) with a maximum contaminant level (MCL) for drinking water at 6 µg L^{-1}, bottled water is considered a food product and thus falls under the scope of the U.S. Food and Drug Administration (FDA) that has not specified a migration limit for antimony from PET packaging materials. This surprising difference of admitted levels between drinking and bottled water was already spotted and criticised by Reimann et al. (2012).

4.5. Effect of exposure to sunlight or UV radiation

Keresztes et al. (2009) observed that antimony concentrations in mineral water stored in the dark increased, even at room temperature. This implies that light is not necessary for the process to take place but not that UV or sun exposure enhances the release. Since most observations combine exposure to sunlight and temperature increase −a factor that does have a positive effect on leaching− it is impossible in practice to disentangle both effects. This methodological artefact was already mentioned in the initial study by Westerhoff et al. (2008) where the authors concluded that sunlight irradiation had only a "small effect" on antimony leaching. Later studies seem to confirm that exposure to sunlight or UV exposure by itself is less important than other factors (Cheng et al., 2010; Hureiki and Mouneimne, 2012; Tukur et al., 2012).

Assessing the potential release of antimony when exposed to radiation has a particular interest because of the use of empty PET bottles in solar water disinfection in developing countries (SODIS treatment). PET has the advantage over glass of being transparent to ultraviolet radiation while window glass is not. Andra et al. (2011) studied the leaching from drinking-water containers subject to SODIS and concluded that, regardless of UV exposure duration, frequency of reuse (up to 27 times) was the major factor that linearly increased antimony leaching from PET bottles at all temperatures tested.

4.6. *Effect of water properties*

The effect of pH on antimony release from PET has been studied with two type of approaches: comparison of carbonated waters with still waters (Table SI5) and release experiments with controlled extractants (Table 2). In the first case, the effect of pH cannot be separated from any possible effect of the presence of CO_2 itself. If we compare results in Table SI5, particularly those based on adequate analytical chemistry, we will observe that carbonated waters usually have higher antimony concentrations but also that observations are based on a small number of samples and often lack statistical treatment. Some studies did not find any difference (e.g. Hureiki and Mouneimne (2012), no stats; Tukur et al. (2012), with stats).

Conclusions from migration experiments (Table 2) are widely variable: no effect but only one probably non-representative bottle (Westerhoff et al., 2008), "not statistically different" but test not given (Sánchez-Martínez et al., 2013), "smaller•• increase than other factors such as heating (Cheng et al., 2010), release "almost" pH independent (Reimann et al., 2010), "significant effect of acidity" (Rungchang et al., 2013). Apart from a general lack of adequate statistical treatment, these experiments have one methodological problem in common: they usually use acetic acid to acidify the samples and acetate is a potential complexant of antimony.

Based on existing evidence, we can conclude that there is from low to no effect of pH on antimony leaching from PET at the pH range studied.

A very few authors have paid attention to the potential effect of major ion composition of water on the antimony release from PET bottles (Westerhoff et al., 2008; Hureiki and Mouneimne, 2012; Tukur et al., 2012). It is difficult to extract any clear conclusion, partly because data are limited but, mainly, because of the possible relationship of water chemistry with antimony concentrations in the source water itself. For instance, Westerhoff and et al. (2008) showed that waters with higher Ca + Mg concentrations had higher antimony concentrations but they never considered the possibility that this effect could originate in the original water source and, thus, could kept no relationship with antimony releasing from PET. Hureiki and Mouneimne (2012) observed higher release in brands richer in calcium but the number of samples was low and initial concentrations unknown (and impossible to know correctly with a LOD of 200 ng L^{-1}).

The effect of calcium on antimony leaching provides examples of bad information transmission. Greifenstein et al. (2013) based their discussion on the fact that "antimony leaching increases with calcium concentration" and gave Westerhoff et al. (2008) and Hureiki and Mouneimne (2012) as only references when the results from these studies were inconclusive. Since milk is a Ca-rich product, Koyuncu and Alwazeer (2019) assumed a mediator effect of milk calcium in the antimony-releasing process from PET bottles with, again, Westerhoff et al. (2008) and Hureiki and Mouneimne (2012) as only references.

Leechart et al. (2015) reported that the concentration of antimony leached into the solution decreased with increasing NaCl concentrations under the same conditions but this effect has not been further studied. Tukur et al. (2012) found no correlation of antimony release with conductivity.

The effect of the presence of organic compounds in water has an enhancing effect on antimony release. This is the case with ethanol at different percentages (Takahashi et al., 2008; Rungchang et al., 2013; Sánchez-Martínez et al., 2013) and oil (Sánchez-Martínez et al., 2013). Antimony migration in Greek spirit samples has been shown to be fast and higher than in water samples (Carneado et al., 2017). The formation of a complex with ligands existing in the raki matrix was evidenced using liquid chromatography with high-resolution tandem spectrometry but this complex disappeared when samples heated at 60 °C. Simulants containing more than 10% ethanol cause some PET to swell and might contribute to an increase of migration (Gehring and Welle, 2018) but, probably, this effect depends on the type of PET.

Some of the highest antimony concentrations measured in commercially available drinks are in fruit juices (Hansen et al., 2006). The observed differences in the extraction ability of the individual drinks was related to the different efficiency of organic extractants such as citrate. However, high concentrations in soft drinks might come from the drinks themselves and not from the PET bottles. For instance, Hansen et al. (2010) when studying commercial juices observed that antimony elevated concentrations in a brand of juices were also found when sold in Tetra Pak cartons concluding that "the antimony might originate from some contaminated ingredient or from the production equipment".

Existing evidence is thus inconclusive concerning the presence of inorganic cations in the water but points to an enhancing effect of the presence of antimony complexing organic ligands.

4.7. *Geochemical studies*

As it has been already commented by Krachler and Shotyk (2006), among other authors, the use of data on trace elements in bottled water in geochemical studies need to consider the possible contribution of the bottles themselves. This means that antimony concentrations cannot be considered in such studies.

5. Conclusions

This review study has confirmed some "known" facts: 1) the presence of antimony in PET bottles is due to the presence of antimony in PET; 2) temperatures above ~70 °C (the glass transition temperature of PET) increases antimony leaching; 3) reported antimony concentrations are below regulated values. However, at the same time, this study brings out shortcomings in many studies with poor analytical practices and faulty study designs. In spite of the publication of many studies these last 15 years, the main "known" facts were already unveiled in the initial studies and not much new knowledge, particularly at the mechanistic level, has been contributed later. Many studies have been performed on a limited number of samples and statistical treatments are often absent. This makes conclusions less robust and needs to be corrected in future studies. There is a tendency, in the transmission of information through citing, to stress the results showing higher antimony release even if based on a few samples.

It is at present not possible to explain the variability in the responses, observed already in the first studies. However, and in spite of the many confounding factors at play, the relative similarity of antimony concentration values reported in water in PET bottles, together with a low ceiling of the higher concentrations measured after time or when heating, suggests an initial fast release step followed by a lack of release or very slow release after. This hypothesis is supported by antimony concentrations in water, even in the most unfavourable case, to be just a tiny percentage of the total antimony in PET bottles but it remains to be proved and better studied.

Future studies should focus on the characterisation of PET in the bottles intended to be used for different types of water and beverages and on the form in which antimony is present in PET. The expression of results per surface unit of PET in release studies is strongly recommended and will allow comparisons to be made on a more solid basis.

Declaration of competing interest

The author declares that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2020.127732.

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Glossary

AAS: atomic absorption spectroscopy
CRM: certified reference material
EXAFS: extended X-ray absorption fine structure
F-AAS: flame atomic absorption spectrometry
HDPE: high density polyethylene
HPLC-ICP-DRC-MS: high-performance liquid chromatography hyphenated with inductively coupled plasma mass spectrometry with dynamic reaction cell
HG-AFS: hydride generation atomic fluorescence spectrometry
ICP-OES: inductively coupled plasma optical emission spectroscopy
ICP-MS: inductively coupled plasma mass spectrometry
ICP-SF-MS: inductively coupled plasma sector field mass spectrometry
IDL:: instrumental detection limit
LOD: limit of detection
LOQ: limit of quantification
MDL:: method detection limit
MRL:: method reporting level
NAA: neutron activation analysis
PET: polyethylene terephthalate
PC: polycarbonate
PP: polypropylene
PS: polystyrene
RDL:: reported detection limit
XRF: X-ray fluorescence