Original research article

Comparison of the nutritional composition and the concentrations of various contaminants in branded and private label yogurts

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Abstract

This study was aimed at comparing the nutritional and contaminant profiles among several of the most consumed trademarks of yogurts in Catalonia (Spain). The nutritional composition, the levels of aflatoxin M_1 (AFM_1), as well as the concentrations of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs), polychlorinated biphenyls (PCBs), and various heavy metals were determined in six of the main trademarks of yogurts in Catalonia. To assess potential quantitative differences, a critical comparison between “private label” yogurts and branded yogurts was performed. AFM_1 was detected in six samples (33%), while the most detected heavy metals were As, Cd and Pb. Slight differences were found between yogurt samples in some minerals and trace elements including Ba, Ca, Cu, Cr, Fe, K and Mg. The WHO-TEQ values ranged between 0.006-0.008 and 0.003-0.012 ng/kg, for PCDD/Fs and PCB, respectively. To the best of our knowledge, this is the first study providing a transversal approach on the occurrence and co-occurrence of the major chemical contaminants in yogurt with a critical comparison between trademarks. The results do not show relevant differences on the nutritional composition, or on the levels of the assessed contaminants (chemicals and AFM_1) between “private label” and branded plain yogurts.

Keywords: Yogurt; Chemical contaminants; Persistent organic pollutants; Biological contaminants; Essential mineral; Heavy metal; Aflatoxin; $AFM_{1;}$ Nutrients; Food analysis; Food composition; Food safety

1 Introduction

Fermented milks and yogurts are typical constituents of the Mediterranean diet. Their beneficial effects and nutritional value have been thoroughly studied, and the results have been widely reported in the scientific literature (Astrup, 2014; Kimoto-Nira et al., 2014). The content of live lactic acid bacteria (LAB), *Lactobacillus bulgaricus* and *Streptococcus thermophilus* species, has been correlated with a wide range of positive health effects in clinical trials (Kimoto-Nira et al., 2014; Wang et al., 2013). Furthermore, yogurt is a rich source of dietary minerals including calcium (Ca), magnesium (Mg), potassium (K), phosphorous (P) and zinc (Zn), among others. Compared with milk, the concentrations of these minerals are higher in yogurt by nearly 50%. Yogurt is also a good source of riboflavin, niacin, vitamin B_6, and vitamin B_{12}, as well as an excellent source of essential amino acids of high biological quality, generally containing higher protein levels than milk (Germani et al., 2014). The proteolytic activity of LAB increases the digestibility of the proteins through a pre-digestion, which efficiently actives the aminoacids (El-Abbadi et al., 2014).

In spite of the beneficial effects of the frequent yogurt consumption, as for most other food items, the potential presence of undesirable chemical substances in foodstuffs is an issue of important concern which has been increasing in recent years. A considerable number of studies have reported the presence of environmental pollutants (i.e. heavy metals, polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs), polychlorinated biphenyls (PCBs), etc.) and contaminants of biological origin (i.e. aflatoxin M_1) (Arnich et al., 2009; Cano-Sancho et al., 2010; Martí-Cid et al., 2009; Martorell et al., 2011; Perelló et al., 2009, 2012, 2014).

Regarding environmental pollutants, a prolonged exposure to toxic elements such as arsenic (As), cadmium (Cd), mercury (Hg) or lead (Pb), which are frequently found in foodstuffs, can cause adverse effects to human health even at relatively low levels (Domingo, 1994; Sharma & Agrawal, 2005). In turn, due to their relevant potential bioaccumulation and toxicity, organochlorinated contaminants such as PCDD/Fs and PCBs are among the most known and investigated persistent organic pollutants (POPs). Both PCDD/Fs and PCBs were included at the 1998 UN-EC POP protocol (UNEP, 2008). Although human exposure to PCDD/Fs and PCBs occurs by various routes, food is the primary source (Llobet et al., 2008; Perelló et al., 2012). On the other hand, aflatoxin M_1 (AFM_1) is the main monohydroxylated derivative of aflatoxinB_1 (AFB_1), formed in the liver by means of cytochrome p450-associated enzymes. Aflatoxins are highly toxic, mutagenic, teratogenic and carcinogenic (Cano-Sancho et al., 2010).

In 2013, yogurt was the most consumed dairy product in Spain, with 15.53 L/person the annual consumption of this product (MAGRAMA, 2013). Between 2009 and 2013, the consumption of dairy products in Spanish homes has increased by an 8.6% (MAGRAMA, 2013; MARM, 2009). Despite the growing variety of dairy products, including flavored recipes and formulations with added prebiotics and/or probiotic bacteria, the most consumed dairy product in Catalonia (Spain) is plain yogurt (MAGRAMA, 2013). In Spain, the market of dairy products in general, and that of yogurts in particular, is characterized by an oligopolistic situation, where the branded yogurts (BY) and the "private label" yogurts (PLY, also known as *white-label* products) hold equal market shares. As a result, the BY seek to differentiate themselves from PLY by means of high investments in nutritional and health research strategies that may raise the price of the final products (Baena & Rodríguez, 2013). As consumers face tighter economic constraints with respect to household food budget spending, determining the cost/benefit ratio of highly consumed food items becomes an issue of socioeconomic, as well as nutritional, interest.

The main objective of this study was to compare the nutritional and contaminant profiles between branded and private-label yogurts purchased in Catalonia. For this study, we determined the nutritional composition and the levels of AFM_1, PCDD/Fs, PCBs, heavy metals and minerals, in the six main yogurt brands sold in Catalonia.

2 Materials and methods

2.1 Sampling

In April 2014, plain yogurt samples from the six major producers (four PLY: "M1, M2, M3 and M4" and two BY: "M5 and M6") were purchased from supermarkets and large markets in Catalonia. The selected brands cover more 80% of the yogurt market in Catalonia, a rate that can be also applied to Spain, where these brands are also similarly distributed. For each brand/company, three composite samples were prepared by pooling twenty individual yogurts (125 g each) from three different production batches. Samples were stored until their subsequent analyses at -20ºC.

2.2 Analysis of nutrients

The ash determination was conducted by subjecting the samples at 525ºC in a muffle furnace (JP Selecta, Abrera, Barcelona, Spain) until a constant weight was obtained.

Moisture content was determined by drying the samples at 105ºC in an air oven (JP Selecta, Abrera, Barcelona, Spain). Protein contents were estimated from the crude nitrogen content of the samples determined by the Dumas method (Jakob et al., 1995). Total fat contents were determined by the Soxhlet method (Manganiello et al., 2000), with the extract gravimetrically determined. Carbohydrates were calculated by difference with the sum of the ash, humidity, protein and fat content combined.

2.3 Analysis of aflatoxin M_1

AFM_1 was determined in each composite sample by competitive ELISA method RIDASCREENs Aflatoxin M_1 30/15 nºR1111 (Ridascreens, R-BiopharmAG, Darmstadt, Germany), according to the procedure described by R-Biopharm GmbH (Kanungo et al., 2014) with minor modifications. First 10 g of triturated and homogenized composite samples of yogurt were weighed and extracted with 40 mL of dichloromethane by shaking for 15 min on a magnetic stirrer and subsequently filtered. The determination was made photometrically at 450 nm on a microplate reader (LT-4000MS, Labtech, Uckfield, East Sussex, UK). The limit of detection was 25 ng/kg.

2.4 Analysis of PCDD/Fs and PCBs

The concentrations of the 17 most toxic PCDD/F congeners and 18 PCBs (including also 12 dioxin-like PCBs (DL-PCBs)) were determined according to the US EPA method 8290 for PCDD/Fs and the US EPA Method 1668 and JIS K 0311 for PCBs. Appropriate C_{13}-labelled extraction standards were added to the homogenized samples in order to control the sample preparation process. Samples were extracted using hexane/acetone (3:1 v/v, pesticide grade, Sigma-Aldrich, Steinheim, Germany) as solvent. The extracts were then concentrated to determine the concentrations of PCDD/Fs and PCBs. A multi-step sample clean-up was performed to remove the matrix, as well as the potential interfering components. The first stage was fat destruction by treatment of the sample solution with acid silica to breakdown the fat. The obtained extract was then subjected to a multilayer silica clean-up column in order to further remove the matrix. After the clean-up, the extract was eluted on a basic alumina column to separate the PCDD/Fs from the PCBs, and from interfering components, by applying different eluent solutions on the column. The PCDD/F and PCB fractions were separately collected and concentrated until near dryness. After adding 25 µL of the C13-labeled injection standards, the extracts were ready for the analysis. The final obtained PCDD/F and PCB extracts were injected and analyzed separately by high-resolution gas chromatography/ high-resolution mass spectrometry (HRGC/HRMS) on an Agilent 6890 Capillary Gas Chromatograph equipped with a DB5-MS capillary column and coupled to a Waters Autospec Ultima High Resolution Mass Spectrometer.

Following the chromatographic separation, the mass spectrometric parameters allowed to separate PCDDs, PCDFs and PCBs between the different chlorination degrees, and between the C^{13}-labeled congeners and the native C^{12}-congeners. The mass spectrometer measured two selected ions per congener group for the native, as well as the labelled components (via “selected ion recording” at a resolution of 10,000). Quantification was carried out using the corresponding isotope-labelled compounds as internal standards (Llobet et al., 2008; Perelló et al., 2012). Toxic equivalents (TEQ) of the analyzed PCDD/Fs and DL-PCBs were calculated using the WHO-toxic equivalency factors (WHO-TEF) for dioxins and dioxin-like compounds (Van den Berg et al., 2006). The limits of detection (LOD, fresh weight) were 0.001-0.023 and 0.038-1.20 ng/kg, for PCDD/Fs and PCBs, respectively.

2.5 Analysis of trace elements

About 0.10 g of each composite sample were pre-digested with 3 mL of 65% nitric acid (Suprapur, Merck, Darmstadt, Germany), 3 mL of 30% hydrogen peroxide (Suprapur, E. Merck), and 2 mL of ultrapure water in Teflon vessels with a Milestone Start D Microwave digestion system (Milestone Srl, Sorisole, Italy). The characteristics of the selected program consisted of ten intervals of 5 to 10 min each for a total of 1 h 35 min, heating up to a maximum temperature of 210ºC (Martorell et al., 2011). The accuracy of the instrumental methods and analytical procedures was checked by duplication of the samples. Analytical grade reagents (Merck, Darmstadt, Germany) were used for blanks and calibration curves. Quality control of the methodology was assured by analyzing a certified reference material (*Lobster hepatopancreas,* NRC Canada, *TORT-2,* Ottawa, ON, Canada). Every 7 samples, a blank sample involving all reagents, was run to check for interference and cross-contamination.

The concentrations of silver (Ag), aluminum (Al), arsenic (As), barium (Ba), calcium (Ca), cadmium (Cd), cobalt (Co), chromium (Cr), copper (Cu), iron (Fe), mercury (Hg), potassium (K), magnesium (Mg), manganese (Mn), nickel (Ni), sodium (Na), phosphorus (P), lead (Pb), selenium (Se), antimony (Sb), tin (Sn), strontium (Sr), vanadium (V) and zinc (Zn) were determined by inductively coupled plasma-mass spectrometry (ICP-MS, Perkin-Elmer Elan 6000, Woodbridge, ON, Canada), (Bocio et al., 2005; Ferre-Huguet et al., 2008,2009). Rhodium was used as internal standard. Replicate measurements were performed. The limits of detection (LOD, fresh weight) were the following: Ag and Zn 0.025 μg/g; Al, Fe and Se 0.50 μg/g; As, Cr and Cu 0.0005 μg/g; Ba 0.025 μg/g; Ca 12.50 μg/g; Cd 0.0001 μg/g; Co, Mn, Sr 0.05 μg/g; Hg 0.01 μg/g; K and P 125 μg/g; Mg 25 μg/g; Pb 0.015 μg/g; Ni and Sn 0.005 μg/g; Na 1250 μg/g; Sb 0.10 μg/g, and V 0.25 μg/g. For calculations, when the concentration of an element was under the respective LOD, that value was assumed to be equal to one-half of the LOD (ND = ½ LOD). Recovery rates for the elements analyzed under the different experimental conditions ranged between 98.4 and 113.5%.

2.6 Statistics

Results were evaluated using the statistical software SPSS.v19.0. The statistical significance of the differences was assessed by applying the Kruskal-Wallis test, considering as significant a level of 0.05. Associations between chemicals were explored through a Pearson's correlation matrix, assuming significant correlations for *P*-values below 0.05.

3 Results and discussion

While information regarding the health benefits of yogurts is abundant (Donovan and Shamir, 2014; Güler and Sanal, 2009; Navarro-Alarcón et al., 2011; Tunick and van Hekken, 2015), data concerning health risks due to the intake of pollutants through consumption of yogurts is rather scarce (Boada et al., 2014; Jensen and Bolger, 2001; Srivastava et al., 2001). In this study, we performed a transversal approach on the occurrence and co-occurrence of the main nutrients and the major chemical contaminants in yogurt, with a critical comparison between trademarks.

3.1 Nutrients

The contents of the major nutrients (total fat, carbohydrates and proteins), as well as the percentages of ash and humidity, which were determined for each composite sample, are summarized in Table 1. The main differences between brands were observed in fat and carbohydrate compositions. However, they were not statistically significant (*P*>0.05). Nutritional compositions were also compared with those provided by the respective manufacturers; the differences were only minor. The lack of nutritional differences found here was also noted by Cleanthous et al. (2008) in a study performed with a wide range of foodstuffs purchased from the Australian market. It was concluded that Australian food products, manufactured on behalf of supermarket retailers and sold as “private label products”, were not nutritionally different from the respective branded products.

3.2 Contaminants

3.2.1 Levels of aflatoxin M_1

AFM_1 was only detected in six composite samples (33%). The measured levels were close to the LOD (25 ng/kg) and below the EU limit of 50 ng/kg, with 33.8 ng/kg the maximum concentration found. Positive samples and mean concentrations were slightly higher in the samples of branded yogurts than in those of the private labels. The mean percentages of positive samples were 25 and 50%, while the mean concentrations were 16.6 and 21.6 ng/kg, for private labels and branded yogurts, respectively. In a previous study, also carried out in Catalonia, only two positive samples of yogurt (among a total of 72) were found. However, one composite sample reached a level of 51.6 ng/kg, which exceeded the EU limit of 50 ng/kg (Cano-Sancho et al., 2010).

3.2.2 Levels of PCDD/Fs and PCBs

The mean concentrations of PCDD/Fs for the 17 most toxic congeners are shown in Table 2. Based on these concentrations and the WHO-TEF of each congener, the mean WHO-TEQ (ng/kg) for each composite sample was calculated (Table 2). Differences between brands were significant for congeners such as 2,3,7,8-TCDD, OCDD, 1,2,3,6,7,8-HxCDF, 1,2,3,4,7,8,9-HpCDF and OCDF. The brands M2 and M3 showed the highest concentrations of these congeners ($P<0.05$), belonging both to the group of private labels. Recently, we found similar levels of most of the PCDD/Fs congeners in yogurts samples purchased in 2008. However, the global estimation of WHO-TEQ (0.010 ng/kg) was slightly higher than our previous estimation, which ranged between 0.006 and 0.008 ng/kg (Perelló et al., 2012).

Table 3 summarizes the mean concentrations of 18 PCB congeners in plain yogurts purchased from the Catalonian market. While no differences among brands were noted for total PCBs, significant differences ($P<0.05$) were found for the congeners PCB 77, 81, 114, 123, 126, 156, 157, 167, 169 and 189. Most PCBs were detected at the highest concentrations in the brands M2 and M3 (Private labels), but occasionally also in the branded yogurts M5 and M6. The WHO-TEQs for Total PCBs ranged between 0.003 and 0.012 ng/kg, with no significant differences between brands. These values are in accordance with those recently reported by Perelló et al. (2012) in yogurt samples from the Catalonian market, which were purchased in 2008. Notwithstanding, a notable difference between the total PCBs results of Perelló et al. (2012) in comparison to those of the current study was noted, being higher in the previous study (57.5 ng/kg) than in the currently one (17.5-34.65 ng/kg).

3.2.3 Levels of toxic metals and other trace elements

The concentrations of 24 toxic and other trace elements and minerals are shown in Table 4. Silver (Ag), Co, Hg, Na, Se and V were not detected in any of the nine composite samples analyzed. Some toxic and non-essential elements were found at concentrations ranging between 0.53 and 4.19 μg/g (Al), 0.05 and 0.10 μg/g (Ba), and 0.01 and 0.09 μg/g (Pb), or at constant concentrations of 0.03 and 0.01 μg/g, for As and Cd, respectively.

As expected, the highest levels of trace elements corresponded to Ca, K, Mg and P. For Ca, its concentration agrees with the nutritional data provided by the yogurt manufacturers. In general terms, the concentrations of most analyzed elements were quite uniform and without relevant differences between brands. Significant differences were noted for Ba, Ca and Sn. The lowest concentrations of Ba and Ca were found in brands M2 and M3, while the highest level of Sn was detected in brand M1. The comparison of the results of the current study with those of a previous survey assessing toxic metals (As, Cd, Hg and Pb) in yogurts also purchased in Catalonia, shows similar results for the toxic metals Cd, Hg and Pb (Martorell et al., 2011). Moreover, we should highlight the current finding of As vs. non-detected levels reported by Martorell et al. (2011), where the LOD was significantly higher (0.10 μg/g).

3.3 Co-occurrence of contaminants/nutrients and correlation analysis

The simultaneous presence of contaminants was studied. The Pearson correlation analysis was performed to detect the pairs and cocktails that more often occur in plain yogurt. Table 5 summarizes the correlation matrix with the Pearson's correlation coefficients and *p*-values, for a selection of chemical pairs with significant correlations. Significant correlations ($P<0.05$) were found between Ca and the major nutrients (fat, carbohydrates and protein), as well as with As, K, Mg, P, Sr and Zn. In turn, Sr was also correlated with carbohydrates and protein, As, Ca and P. On the other hand, total PCBs were correlated with Ba, Cu and Pb.

To the best of our knowledge, this is the first study providing a transversal approach with useful information on the occurrence and co-occurrence of some important potential chemical contaminants in yogurts, with a critical comparison between major brands. On one hand, we have considered the growing market share of private labels during the last two decades; while on the other hand, we also considered the best quality facts proposed by the branded products. Consequently, according to a perception survey performed by the Spanish Government (MARM, 2009), 14% of consumers responded that the lower quality of private labels could explain their lower prices.

4 Conclusion

In summary, potential cocktails of contaminants involving AFM_1, PCDD/Fs, PCBs and toxic metals should be expected in yogurts, whose combined adverse effects are still quite unknown. However, yogurts should not be considered as a relevant source of human health risks, because the pollutant concentrations are usually low. On the other hand, our comparison between private label and branded yogurts did not show notable differences in their nutritional compositions, or in their contamination levels. The most relevant differences were found in some PCDD/Fs and PCBs congeners, pollutants that in two private label yogurts were found at higher concentrations than in the remaining brands. As conclusion, in the context of the serious current economic crisis, and based on the results of the present survey, from the point of view of the nutrients and contaminants here analyzed, the significant differences among the prices of the BY and PLY would not be justified.

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Table 1. Nutritional composition of the main yogurt brands marketed in Catalonia, Spain (%). Comparison between private label (PLY) and branded yogurts (BY).

Yogurt sample		Ash (%)	Fat (%)	Carbohydrates (%)	Humidity (%)	Protein (%)
PLY	**M1**	0.4	2.8	4.6	88.6	3.7
	M2	0.6	2.2	5.1	88.7	3.4
	M3	0.4	0.8	6.2	89.3	3.5
	M4	0.3	5.0	2.6	88.8	3.6
BY	**M5**	0.5	2.7	5.0	88.6	3.4
	M6	0.4	3.6	3.4	89.3	3.4

Values are means of 3 composite samples pooled, in turn, from 20 individual yogurts from 3 different series.

Table 2. Mean concentrations of PCDD/Fs in some of the yoghurt brands mainly consumed in Catalonia, Spain (ng/kg). Comparison between private label (PLY) and branded yogurts (BY).

Yogurt sample		2,3,7,8-TCDD	1,2,3,7,8-PeCDD	1,2,3,4,7,8-HxCDD	1,2,3,6,7,8-HxCDD	1,2,3,7,8,9-HxCDD	1,2,3,4,6,7,8-HpCDD	OCDD	2,3,7,8-TCDF	1,2,3,7,8-PeCDF	2,3,4,7,8-PeCDF	1,2,3,4,7,8-HxCDF	1,2,3,6,7,8-HxCDF	2,3,4,6,7,8-HxCDF	1,2,3,7,8,9-HxCDF	1,2,3,4,6,7,8-HpCDF	1,2,3,4,7,8,9-HpCDF	OCDF	WHO-TEQ
PLY	**M1**	0.0004^{a}	0.002	0.001	0.004	0.002	0.011	0.029^{b}	0.005	0.002	0.002	0.003	0.002^{a}	0.001	0.001	0.027	0.002^{a}	0.024^{a}	0.006
	M2	0.001^{b}	0.002	0.002	0.003	0.002	0.013	0.044^{c}	0.002	0.001	0.003	0.003	0.002^{a}	0.002	0.001	0.084	0.003^{b}	0.071^{b}	0.007
	M3	0.001^{b}	0.001	0.002	0.001	0.001	0.013	0.045^{c}	0.002	0.003	0.005	0.005	0.004^{b}	0.004	0.001	0.104	0.003^{b}	0.076^{b}	0.008
	M4	0.002^{b}	0.003	0.002	0.003	0.004	0.007	0.020^{a}	0.001	0.001	0.003	0.003	0.001^{c}	0.002	0.001	0.026	0.002^{a}	0.020^{a}	0.008
BY	**M5**	0.001^{b}	0.002	0.002	0.004	0.003	0.010	0.021^{ab}	0.002	0.001	0.003	0.003	0.002^{a}	0.003	0.0003	0.030	0.002^{ab}	0.018^{a}	0.007
	M6	0.001^{b}	0.002	0.002	0.003	0.002	0.008	0.009^{a}	0.002	0.002	0.004	0.002	0.002^{a}	0.002	0.001	0.024	0.002^{a}	0.017^{a}	0.007

^{a,b,c} Superscripts mean statistically significant differences between brands according to the Kruskal-Wallis test ($p<0.05$).

Table 3. Concentrations of PCBs in some of the yogurt brands mainly consumed in Catalonia, Spain (ng/kg). Comparison between private label (PLY) and branded yogurts (BY).

Yogurt sample		PCB 28	PCB 52	PCB 77	PCB 81	PCB 101	PCB 105	PCB 114	PCB 118	PCB 123	PCB 126	PCB 138	PCB 153	PCB 156	PCB 157	PCB 167	PCB 169	PCB 180	PCB 189	Total PCBs	WHO-TEQ
PLY	**M1**	6.583	3.650	0.538^{a}	0.039^{a}	1.300	0.830	0.080^{a}	2.983	0.080^{a}	0.050^{a}	3.883	5.050	0.390^{a}	0.393^{a}	0.020^{a}	0.080^{a}	2.217	0.080^{a}	17.48	0.006
	M2	2.133	3.100	1.200^{b}	0.055^{b}	1.417	0.950	0.108^{c}	4.133	0.108^{b}	0.092^{a}	7.933	11.53	0.548^{b}	0.548^{b}	0.027^{b}	0.108^{b}	6.900	0.108^{b}	34.57	0.010
	M3	1.750	3.267	1.433^{b}	0.062^{b}	1.250	1.267	0.125^{c}	4.167	0.125^{b}	0.107^{a}	5.900	8.367	0.617^{b}	0.617^{b}	0.031^{b}	0.125^{b}	3.833	0.270^{bc}	26.86	0.012
	M4	2.300	2.733	0.833^{a}	0.045^{ab}	0.900	0.903	0.090^{ab}	2.400	0.090^{ab}	0.052^{a}	3.633	5.400	0.452^{ab}	0.452^{a}	0.023^{a}	0.090^{a}	2.733	0.452^{c}	17.72	0.006
BY	**M5**	1.817	2.017	0.425^{a}	0.048^{b}	0.967	1.470	0.135^{bc}	5.733	0.097^{ab}	0.055^{a}	7.100	8.700	0.702^{b}	0.475^{ab}	0.024^{ab}	0.155^{ab}	4.167	0.518^{c}	30.35	0.007
	M6	0.867	0.867	0.087^{a}	0.043^{ab}	0.867	1.150	0.087^{b}	2.300	0.087^{ab}	0.022^{b}	3.800	5.433	0.432^{ab}	0.087^{b}	0.022^{a}	0.432^{c}	2.833	0.432^{c}	18.03	0.003

^{a,b,c} Superscripts mean statistically significant differences between brands according to Kruskal-Wallis test ($p < 0.05$).

Table 4. Mean concentrations of metals and minerals in some of the yogurt brands mainly consumed in Catalonia, Spain (µg/g). Comparison between private label (PLY) and branded yogurts (BY).

		PLY				BY	
Metals and minerals	**Limit of detection**	**M1**	**M2**	**M3**	**M4**	**M5**	**M6**
Ag	0.025	<0.025	<0.025	<0.025	<0.025	<0.025	<0.03
Al	0.50	1.07	0.67	4.19	1.30	0.87	0.53
As	0.0005	0.008	0.01	0.010	0.010	0.010	0.008
Ba	0.025	0.09^{a}	0.05^{b}	0.05^{ab}	0.10^{a}	0.06^{ab}	0.09^{a}
Ca	12.50	1134^{a}	930^{b}	1020^{b}	1103^{a}	1023^{a}	1095^{a}
Cd	0.0001	0.002	0.001	0.001	0.001	0.001	0.001
Co	0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
Cr	0.0005	0.13	0.03	0.04	0.08	0.15	0.15
Cu	0.0005	1.68	0.12	0.20	0.11	0.09	0.09
Fe	0.50	1.69	1.34	1.36	1.41	1.34	1.13
Hg	0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
K	125	1566	1363	1490	1523	1450	1513
Mg	25	105.88	90.95	98.08	103.32	97.15	105.67
Mn	0.05	0.04	0.04	0.03	0.07	0.04	<0.05
Ni	0.005	0.08	0.03	0.16	0.04	0.03	0.04
Na	1250	<1250	<1250	<1250	<1250	<1250	<1250
P	125	858	627	666	739	731	766
Pb	0.0002	0.09	0.02	0.02	0.01	0.01	0.02
Se	0.50	<0.50	<0.50	<0.50	<0.50	<0.50	<0.50
Sb	0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10
Sn	0.005	0.03a	0.01^{b}	0.02^{ab}	<0.005	<0.005	<0.005
Sr	0.05	0.53	0.80	0.65	0.59	0.66	0.60
V	0.25	<0.25	<0.25	<0.25	<0.25	<0.25	<0.25
Zn	0.025	4.31	2.76	2.56	3.87	4.31	3.21

^{a,b} Superscripts mean significant differences between brands according to Kruskal-Wallis test ($p < 0.05$)

Table 5. Pearson correlation's matrix with a selection of the most relevant pairs of chemicals.

		Fat	**CH**	**Protein**	**PCDD/Fs (WHO-TEQ)**	**Total PCBs**	**PCBs (WHO-TEQ)**	**As**	**Ba**	**Ca**	**Cr**	**Cu**	**K**	**Mg**	**P**	**Pb**	**Sn**	**Sr**	**Zn**
Fat	PCC	1.00	**-0.96**	0.43	0.15	-0.14	-0.30	-0.35	0.36	**0.50**	0.18	-0.38	0.38	0.45	0.37	-0.35	0.19	-0.43	0.19
	p-value		**<0.01**	0.08	0.54	0.57	0.23	0.16	0.14	**0.04**	0.46	0.12	0.12	0.06	0.13	0.15	0.45	0.08	0.45
CH	PCC		1.00	**-0.49**	-0.14	0.03	0.25	0.46	-0.24	**-0.510**	-0.18	0.45	-0.40	-0.44	-0.37	0.43	0.12	**0.56**	-0.13
	p-value			**0.04**	0.58	0.91	0.33	0.06	0.34	**0.03**	0.48	0.06	0.10	0.07	0.13	0.08	0.63	**0.02**	0.62
Protein	PCC			1.00	-0.03	0.04	-0.01	-0.37	0.04	**0.59**	0.18	-0.01	**0.54**	0.45	**0.57**	0.01	0.22	**-0.48**	**0.47**
	p-value				0.90	0.87	0.98	0.13	0.88	**0.01**	0.48	0.97	**0.02**	0.06	**0.01**	0.98	0.38	**0.04**	**0.05**
PCDD/Fs (WHO-TEQ)	*PCC*				1.00	0.16	0.27	0.19	-0.24	-0.03	-0.30	-0.16	0.09	0.02	-0.05	-0.17	-0.32	0.06	-0.03
	p-value					0.51	0.28	0.46	0.34	0.92	0.22	0.51	0.71	0.93	0.85	0.51	0.19	0.81	0.9
Total PCBs	PCC					1.00	**0.67**	0.07	**-0.81**	-0.36	-0.28	**-0.48**	-0.25	-0.41	-0.40	**-0.50**	-0.05	0.25	-0.24
	p-value						**<0.01**	0.79	**<0.01**	0.14	0.26	**0.04**	0.32	0.09	0.10	**0.04**	0.86	0.33	0.34
PCBs (WHO-TEQ)	PCC						1.00	0.19	**-0.59**	-0.28	**-0.57**	-0.25	-0.08	-0.32	**-0.48**	-0.30	0.20	0.36	-0.40
	p-value							0.46	**0.01**	0.25	**0.01**	0.32	0.74	0.20	**0.04**	0.23	0.43	0.14	0.10
As	PCC							1.00	-0.16	**-0.57**	**-0.56**	-0.21	**-0.51**	-0.46	**-0.58**	-0.23	-0.46	**0.68**	-0.37
	p-value								0.52	**0.01**	**0.01**	0.39	**0.03**	0.05	**0.01**	0.37	0.05	**<0.01**	0.13
Ba	PCC								1.00	0.46	0.42	0.46	0.26	**0.48**	0.44	**0.47**	0.07	-0.21	0.33
	p-value									0.05	0.08	0.05	0.29	**0.04**	0.07	**0.05**	0.79	0.40	0.17
Ca	PCC									1.00	0.49	0.12	**0.95**	**0.95**	**0.76**	0.13	0.35	**-0.58**	**0.55**
	p-value										0.04	0.63	**<0.01**	**<0.01**	**<0.01**	0.60	0.16	**0.01**	**0.02**
Cr	PCC										1.00	0.35	0.27	0.42	**0.65**	0.37	0.09	-0.44	**0.63**
	p-value											0.16	0.27	0.09	**<0.01**	0.13	0.71	0.07	**0.01**
Cu	PCC											1.00	0.07	0.12	0.35	**0.99**	0.44	0.00	0.44
	p-value												0.78	0.64	0.15	**<0.01**	0.07	0.99	0.07
K	PCC												1.00	**0.93**	**0.62**	0.07	0.38	-0.46	0.44
	p-value													**<0.01**	**0.01**	0.78	0.11	0.05	0.07
Mg	PCC													1.00	**0.70**	0.13	0.25	-0.41	0.42
	p-value														**<0.01**	0.61	0.31	0.09	0.08
P	PCC														1.00	0.40	0.36	**-0.66**	**0.66**
	p-value															0.10	0.14	**<0.01**	**<0.01**
Pb	PCC															1.00	0.44	-0.03	0.46
	p-value																0.07	0.91	0.05
Sn	PCC																1.00	-0.38	0.20
	p-value																	0.12	0.42
Sr	PCC																	1.00	-0.41
	p-value																		0.09

PCC. Pearson's correlation coefficient. Correlations with *p-value* below 0.05 were considered to be statistically significant (bold numbers).