

TRIHALOMETHANE CONCENTRATIONS IN TAP WATER AS DETERMINANT
OF BOTTLED WATER USE IN THE CITY OF BARCELONA

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Abstract

Background. Bottled water consumption is increasing worldwide, despite its huge economic and environmental cost. We aim to describe personal and tap water quality determinants of bottled water use in the city of Barcelona.

Methods. This cross-sectional study used data from the Health Survey of Barcelona in 2006 (N=5,417 adults). Use of bottled water to drink and to cook was evaluated in relation to age, gender, educational level, district and levels of trihalomethanes (THMs), free chlorine, conductivity, chloride, sodium, pH, nitrate and aluminium in municipal tap water using Robust Poisson Regression.

Results. The prevalence of bottled water use to drink and cook was 53.9% and 6.7%, respectively. Chemical parameters in water had a large variability (interquartile range of THMs concentrations: 83.2-200.8 $\mu\text{g/L}$) and were correlated between them, except aluminium. Drinking bottled water increased with educational level, while cooking with bottled water was higher among men than among women and decreased with age. After adjusting by these personal determinants, a dose-response relationship was found between concentrations of all chemicals except aluminium in tap water and bottled water use. The highest association was found for THMs, with a Prevalence Ratio (PR) of 2.00 (95%CI 1.86, 2.15) for drinking bottled water and 2.80 (95%CI=1.72, 4.58) for cooking with bottled water, among those with $>150 \mu\text{g/L}$ vs. $<100 \mu\text{g/L}$ THMs in tap water.

Conclusion. More than half of Barcelona residents regularly drank bottled water, and the main determinant was the chemical composition of tap water, particularly THMs level.

Keywords: tap water, drinking water, trihalomethanes, organoleptics, determinants, prevalence

Introduction

Bottled water consumption is increasing worldwide (Rodwan 2010). However, bottled water is 240 to 10,000 times more expensive than tap water (Olson 1999) and requires 1,000 to 2,000 times more energy, especially for bottle production and transportation (Gleick and Cooley 2009). One of the main causes of the increasing bottled water consumption is the belief that it is healthier than tap water, which has been promoted by heavy marketing campaigns, but also that it has a better taste (Doria 2006; Levallois et al. 1999; Saylor et al. 2011; Ward et al. 2009). These beliefs and willingness to buy bottled water vary between communities but also within them, depending on personal characteristics and socioeconomic factors (Castaño-Vinyals et al. 2011; Dupond et al. 2010; Font-Ribera et al. 2010; Hu Z et al. 2011; Huerta-Saenz et al. 2012).

Chemical properties and organoleptic characteristics of bottled and tap water vary across different locations and types of bottled water (Marcussen et al. 2013; Platikanov et al. 2013; Whelton et al. 2007). Although expert panels can blindly distinguish between tap and bottled water (Platikanov et al. 2013), there is little scientific evidence to support that bottled water has better sensory qualities compared to tap water (Marcussen et al. 2013) and that general population can appreciate these differences. Taste problems in tap water occur more frequently when surface water is used as source water to produce potable water (Levallois et al. 1999) and chlorination is applied (Marcussen et al. 2013). Mineral bottled water is not disinfected, and consequently, it does not contain free chlorine and chlorination by-products (Font-Ribera et al. 2010).

Spain is among the top ten countries with the highest per capita consumption of bottled water (Rodwan 2010). Barcelona, located on the Mediterranean coast, is the second largest city of the country. Level of trihalomethanes (THMs), the most common disinfection by-product in drinking water, were reported to be high in tap water (Gómez-Gutierrez et al. 2012). Since the city is supplied by surface water from two rivers with very different organic matter and bromide content (López-Roldán et al. 2016), chemical characteristics of tap water including THMs level have a large geographical variability within the city (Gómez-Gutierrez et al. 2012).

Previous studies on bottled water consumption determinants described personal determinants and health beliefs in small and/or selected samples (Saylor et al. 2011;

Huerta-Saenz et al. 2011; Merkel et al. 2012; Ward et al. 2009; van Erp et al. 2014). Although previous work has shown a high prevalence of bottled water use in areas of high THMs concentrations in tap water (Villanueva et al. 2007; Villanueva et al. 2011), there are no studies describing the relationship between tap water quality and the prevalence of bottled water use. The aim of this study is to identify personal and tap water quality determinants of the prevalence of bottled water use to drink and to cook in a population-based sample in the city of Barcelona.

1. Material and methods

This is a cross-sectional study using data from the Health Survey of Barcelona in 2006 and the concentrations of chemical parameters in municipal tap water during the three years prior to the survey (2003-2006).

1.1. Health Survey

The Barcelona Health Survey collected data through home-based personal interviews on a representative sample of all non-institutionalized subjects aged ≥ 15 years living in Barcelona in 2006. The sampling design involved stratification by the 10 districts of the city and participants were randomly selected through proportional assignments of gender and age. Non-respondents, resulting from refusal to participate or absence, were replaced by other residents with the same characteristics (20% in 2006) (Rodríguez-Sanz et al. 2008).

Prevalence of bottled water consumption was calculated with the following questions “How often do you use bottled water to drink?” and “How often do you use bottled water to cook?”. The three possible answers “Usually”, “Occasionally” or “Never”, were categorized into two: “Yes” for usually and “No” for occasionally or never. Gender, age and educational level were collected as potential socioeconomic determinants of bottled water use. The district of residence was used to link the data with the water parameters.

1.2. Water quality parameters

We used routine monitoring data that was part of the surveillance carried out by the Public Health Agency of Barcelona between 2003-2006 in compliance with the Directive 98/83/EC, on the quality of water intended for human consumption. Data for

the period 2003-2006 was analysed, including measurements from 109 water samples (1 L) collected in 87 public fountains selected randomly and covering all the city water distribution system. During 2003-2005, at least one water sample was collected per year and per district. Water samples were collected after flushing the fountain for about two minutes and stored refrigerated until chemical analysis. Free chlorine was determined *in situ* by means of a Lovibond Water Testing Kit and the rest of the water parameters were determined in the Laboratory of the Public Health Agency of Barcelona according to accredited standard analytical methodologies.

The following parameters were analyzed in all the samples collected: free chlorine, chloroform, dibromochloromethane, bromodichloromethane, bromoform, conductivity and nitrite. A more complete analysis was conducted in a random selection of 30 water samples (including at least one sample per district in two different years). The complete analysis included nitrate, pH, lead, aluminium, chloride, copper, iron and sodium. Total THMs (TTHMs) was calculated summing up the levels of chloroform, dibromochloromethane, bromodichloromethane and bromoform and was expressed as µg/L. Levels below the limit of detection (LOD) were substituted by half of the LOD value.

1.3. Statistical analysis

The Barcelona Health Survey 2006 was available for 5,419 subjects older than 14 years. Two subjects were excluded for having missing data in the water use question and the final analyzed sample was 5,417.

The normal distribution of the water chemical parameters was checked with a Skewness and Kurtosis based test for Normality and the Spearman correlation between the water parameters was calculated. The median level of the parameters during the period of study was calculated for each of the 10 districts of the city and linked to the study participants. Each water parameter was then categorized into three categories balancing the number of subjects across the categories.

A bivariate analysis was performed to study the potential role of the water quality parameters and socioeconomic characteristics as determinants of bottled water use using the χ^2 -squared test. The Prevalence Ratios (PR) of bottled water use to drink and to cook for each determinant was calculated using Robust Poisson Regression and multivariate

models were fitted to adjust for confounders. Statistical analyses were conducted with STATA 11.2.

2. Results

Table 1 shows the water chemical composition in the city during the period of study. The median TTHMs was 119.6 $\mu\text{g/L}$, with an Interquartile range (IQR) of 83.2-200.8. A strong variability within the city was also observed for other parameters, such as conductivity (IQR=473.0-1191.0 $\mu\text{S/cm}$) or chloride (IQR=40.7-279.8 mg Cl/L). The vast majority of samples were below the limit of detection for nitrites, lead, copper and iron, and therefore these parameters were excluded from the statistical analysis.

The overall prevalence of bottled water use in the city is summarized in Figure 1. Bottled water use to drink was 53.9% on average and it varied greatly between districts, from 27.3% to 74.1%. The prevalence of bottled water use to cook was 6.7% on average, varied from 2.0% to 12.5% between districts and was strongly correlated with the bottled water use to drink ($p\text{-value}<0.001$). The study population is described in Table 2. Among the study participants, 46.8% were men and 27% had university education. Men used more bottled water to cook but not to drink than women. Bottled water use decreased with age and increased with education.

All the water chemical parameters analyzed were strongly associated with both bottled water use to drink and to cook ($p\text{-values}<0.001$) (Table 3). For example, the prevalence of bottled water use to drink was 37.1% in the area with lower TTHMs level ($<100 \mu\text{g/L}$), 56.1% in the area with intermediate level and 73.5% in the area with highest TTHMs level ($>150 \mu\text{g/L}$).

The multivariate models indicate that drinking bottled water increased with educational level, while cooking with bottled water was higher among men and decreased with age (Table 4). After adjusting by educational level, the level of several chemical parameters in tap water was significantly related to an increased PR of using bottled water to drink. The highest PR was found for TTHMs with a PR of 2.00 (95%CI 1.86, 2.15) for $>150 \mu\text{g/L}$ vs. $<100 \mu\text{g/L}$. A dose-response relationship was observed for TTHMs, conductivity, free chlorine, pH, chloride, sodium and nitrate (which are all correlated, supporting material Table S1), while this was not the case for aluminium. The PR for bottled water use to cook was even more affected by some water parameters, although

the statistical significance was lost in some category. The adjusted PR for bottled water use to cook was 2.80 (95% CI=1.72, 4.58) for >150 µg/L vs. <100 µg/L of TTHMs.

3. Discussion

More than half (54%) of Barcelona residents regularly drank bottled water and 7% used it regularly to cook. These percentages showed a large geographic variability within the city, depending on socioeconomic determinants but mainly on the chemical composition of tap water, including TTHMs level.

Although it is difficult to compare the prevalence of regularly drinking bottled water to other studies due to methodological differences, the prevalence of drinking bottled water found in Barcelona is the highest reported among the general population. Prevalence of regularly drinking bottled water was 43% in Québec (Canada) in 1995 (Levallois et al. 1999), 34% in Ontario (Canada) in the mid 2000' (Pintar et al. 2009), 42% in France in 2000 (IFEN 2000), 32% in Santa Clara County (US) in 2011 (van Erp et al. 2014), 30% in Italy in 2011 (Giglio et al. 2015) and 26% in Shanghai (China) in 2011 (Chen et al. 2012). In Barcelona, drinking bottled water increased with education, while no differences were detected by gender or age in multivariate models. Different results have been reported in the literature (Jones et al. 2006; Saylor et al. 2011; Chen et al. 2012) indicating that the personal determinants of drinking bottled water vary by community and cultural background (Huerta-Saenz et al. 2011). The positive correlation between drinking bottled water and socioeconomic status has previously been found in Spain (Castaño-Vinyals et al. 2011), in Canada (Dupond et al. 2010) and in China (Chen et al. 2012), while the opposite association has been repeatedly described in the US (Huerta-Saenz et al. 2012; Hu et al. 2011; van Erp et al. 2014). We have not found other studies to compare the prevalence and determinants of using bottled water to cook. In Barcelona, the prevalence of bottled water use for cooking was much lower than for drinking, but still it can have a considerable impact since the amount of water consumed might be higher for cooking than for drinking.

Although personal characteristics were associated with bottled water consumption, the geographic component had a higher effect in the prevalence, which ranged across districts from 23% to 74% and from 2% to 12% for drinking and cooking, respectively. After accounting for socioeconomic and personal determinants, a dose-response

relationship between the levels of TTHMs, free chlorine, pH, chloride, sodium and nitrate in tap water and the consumption of bottled water for drinking and cooking was observed. Although it is not possible with these data to disentangle the chemical that most influenced the acceptability of tap water in the city, TTHMs level was the parameter showing a highest association with bottled water use. The areas with the highest TTHMs level in tap water doubled and almost tripled the prevalence of bottled water use to drink and to cook, respectively, compared to the areas with the lowest TTHMs level. By the time of the study, THMs concentration in tap water was high in some areas of the city and 70% of the population received water with TTHMs level above the current maximum regulated level in the EU of 100 µg/L. THMs have not been described to modify water taste *per se*, but as shown in the present study, THMs level in tap water is highly correlated with other parameters that are known to impair water taste at high levels, such as chloride, chlorine, sodium or salt content (WHO 2011; Platikanov et al. 2013). In fact, some areas of the city had sodium or chloride in tap water above the level resulting in unpalatable taste (WHO 2011). Therefore, TTHMs level in tap water could be a marker of chemical quality of tap water related to organoleptic characteristics and acceptability. The dose-response relationship between the level of chemicals in tap water and bottled water use was not observed for aluminium, which was not correlated to the other chemicals and has no organoleptic properties at the levels found in Barcelona municipal water (<0.1 mg/L) (WHO 2011).

This study indicates that in a given community, organoleptic characteristics of tap water can be a main determinant of tap water acceptability and bottled water consumption, more than personal characteristics. This data is in accordance with studies in Canada (Levallois et al. 1999), France (IFEN 2000) or UK (Ward et al. 2009) reporting taste as the main reason for drinking bottled water. It also indicates that the general population appreciate differences in taste between tap and bottled water, at least when tap water has a high salt content. Globally, a large proportion of bottled water (40–60%) consists of packaged tap water (Canadean 2004) with THMs in it (Marcussen et al. 2013), but this is not common in Spain and THMs were extremely low in a sample of popular Spanish bottled water brands (Font-Ribera et al. 2010). The situation described in Barcelona in 2006 may not be extrapolated to other settings where bottled and tap water may be more similar or even to the current situation in the city where THMs level has decreased

considerably and homogenized across the city (Gómez-Gonzalez et al. 2012) due to improvements in water treatment (López-Roldán et al. 2016)

This is the study on bottled water determinants with largest sample size (>5,000), with a population based sample of citizens in a large south-European city. This is the first study on the topic to combine personal and water chemical composition. We don't have data on actual reasons or believes to choose bottled water and we have assumed that the differences observed by tap water parameters after adjusting by personal characteristics correspond to differences in taste. However, we don't know if people reject drinking tap water because simply they don't like the taste or if they relate an unpleasant taste with potentially more unsafe water. It is unlikely that people living in areas with high THMs level were aware of this and avoided drinking tap water to reduce the exposure to these compounds. This study did not consider the use of tap water filters. The prevalence of the use of these filters in the city is unknown, but two studies carried out in Spain during the same period reported that 9% of children at 9-12 years (Font-Ribera et al. 2010) and 6% of pregnant women (Villanueva et al. 2011) usually drank filtered tap water. The use of tap water filters is probably also related to the organoleptic quality of tap water (Levallois et al. 1999) and therefore should be considered in future studies.

4. Conclusions

More than half of Barcelona residents regularly drank bottled water, and the main determinant was the chemical composition, particularly TTHMs level, followed by socioeconomic factors. This is in agreement with studies on drinking preferences reporting taste as the main reason for drinking bottled water. The study illustrates that objective quality of drinking water is more relevant than subjective reasons in explaining the preferences of type of water consumed in the population.

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Figure 1. Prevalence of bottled water consumption to drink and cook in the city of Barcelona, 2006 (N=5,417).

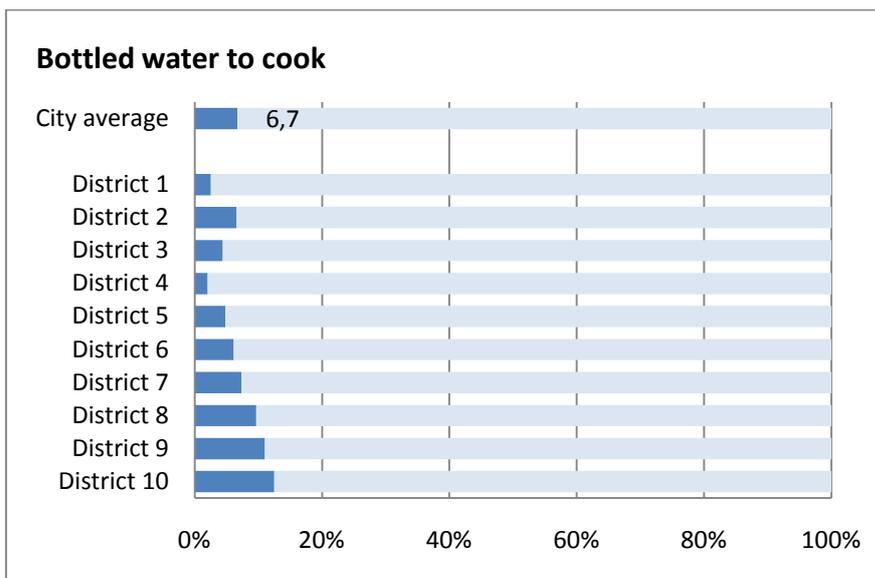
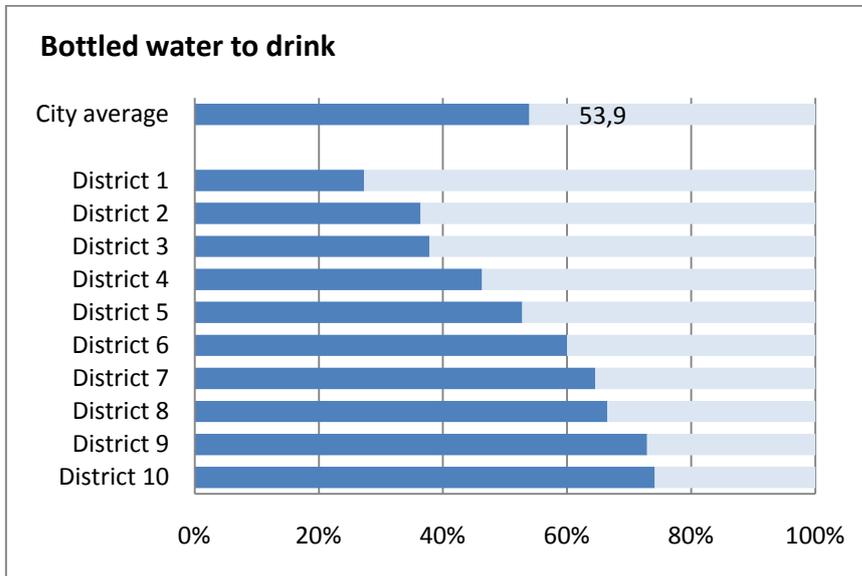


Table 1. Chemical composition of tap water. Barcelona, 2003-2006.

	N	mean	median	IQR	LOD	%<LOD
Chloroform (µg/L)	109	33.5	34.9	19.0-45.0	5.0	0
Bromoform (µg/L)	109	18.8	5.0	2.5-31.0	5.0	32.7
DBCM (µg/L)	109	17.1	9.3	2.5-31.4	5.0	31.8
BDCM (µg/L)	109	19.4	18.6	14.0-23.0	5.0	0
TTHMs (µg/L)	109	144.2	119.6	83.2-200.8	20.0	0
Free chlorine (mg/L)	103	0.5	0.5	0.3-0.7	0.01	0
Conductivity (µS/cm)	109	840.6	532.0	473.0-1191.0	<10	0
Nitrite (µg NO ₂ /L)	109	19.4	5.0	5.0-50.0	0.01/0.1	87.3
Nitrate (mg NO ₃ /L)	30	11.1	9.3	8.9-13.6	4.0	0
pH	30	7.7	7.7	7.5-7.8	4.0	0
Chloride (mg Cl/L)	30	153.3	101.7	40.7-279.8	10.0	0
Sodium (mg/L)	30	96.0	41.5	30.2-167.1	10.0	0
Aluminium (µg/L)	30	42.7	46.0	20.0-62.0	40.0	46.7
Lead (µg/L)	30	2.6	2.5	2.5-2.5	5.0	93.3
Copper (µg/L)	30	20.0	20.0	20.0-20.0	0.04	100
Iron (µg/L)	30	20.0	20.0	20.0-20.0	40.0	100

IQR: interquartile range. LOD: limit of detection. DBCM: dibromochloromethane. BDCM: bromodichloromethane. TTHMs: total trihalomethanes.

Table 2. Description of the study population and prevalence of bottled water use by each personal determinant category. N=5,417.

	%	Bottled water to drink (%)	Bottled water to cook (%)
Gender			
Men	46.8	54.4	7.8
Women	53.2	53.5	5.8
p-value		0.549	0.003
Age (years)			
15-30	21.5	55.0	7.9
31-50	34.3	58.2	7.7
51-70	25.8	51.1	6.3
>70	18.4	48.6	4.2
p-value		<0.001	0.001
Educational level			
< Primary	11.9	46.6	3.7
Primary	36.9	51.6	7.2
Secondary	24.3	55.0	6.5
University	27.0	59.2	7.8
p-value		<0.001	0.013

p-value for χ^2 -squared test.

Table 3. Distribution of the water quality parameters in the population and prevalence of bottled water use by each water quality category. N=5,417.

		%	Bottled water to drink (%)	Bottled water to cook (%)
TTHMs ($\mu\text{g/L}$)	<100	30.1	37.1	3.0
	100-150	49.4	56.1	6.9
	>150	20.5	73.5	11.8
	p-value		<0.001	<0.001
Conductivity ($\mu\text{S/cm}$)	<500	49.6	41.4	4.3
	500-1000	29.9	61.3	7.3
	>1000	20.5	73.5	11.8
	p-value		<0.001	<0.001
Free chlorine (mg/L)	<0.5	30.1	69.2	10.0
	0.5	20.0	51.7	8.1
	>0.5	49.9	45.6	4.2
	p-value		<0.001	<0.001
pH	<7.7	40.6	66.7	9.6
	7.7	29.7	46.2	6.1
	>7.7	29.7	44.2	3.5
	p-value		<0.001	<0.001
Chloride	<55	59.4	45.2	4.8
	55-200	10.0	52.8	4.8
	>200	30.6	71.2	11.1
	p-value		<0.001	<0.001
Sodium (mg/L)	<50	59.6	43.3	4.4
	50-100	9.8	64.5	7.4
	>100	30.6	71.2	11.1
	p-value		<0.001	<0.001
Nitrate ($\text{mg NO}_3/\text{L}$)	<9	19.9	49.5	3.4
	9-12	49.5	45.0	5.4
	>12	30.6	71.2	11.1
	p-value		<0.001	<0.001
Aluminium ($\mu\text{g/L}$)	<40	25.1	51.7	8.1
	40-50	37.3	54.5	4.7
	>50	37.5	41.4	4.3
	p-value		<0.001	<0.001

p-value for χ -squared test. TTHMs: total trihalomethanes.

Table 4. Prevalence ratio (PR) of using bottled water to drink and to cook by personal and water quality determinants. Barcelona, 2006 (N=5.417).

		Bottled water to drink		Bottled water to cook	
		PR ^a	95%CI	PR ^b	95%CI
Gender	Men	1.00	0.95-1.05	1.75	1.32-2.32
	Women	1		1	
Age (years)	15-30	1		1	
	31-50	1.05	0.98-1.12	0.63	0.44-0.90
	51-70	0.95	0.88-1.02	0.58	0.38-0.87
	>70	0.93	0.85-1.02	0.60	0.34-1.08
Educational level	<Primary	1		1	
	Primary	1.11	1.01-1.22	1.48	0.68-3.21
	Secondary	1.18	1.07-1.30	0.97	0.42-2.27
	University	1.27	1.16-1.39	1.73	0.71-4.21
TTHMs (µg/L)	<100	1		1	
	100-150	1.48	1.38-1.59	1.85	1.14-3.00
	>150	2.00	1.86-2.15	2.80	1.72-4.58
Conductivity (µS/cm)	<500	1		1	
	500-1000	1.47	1.38-1.56	1.17	0.82-1.67
	>1000	1.81	1.71-1.92	1.96	1.37-2.80
Free chlorine (mg/L)	<0.5	1.51	1.43-1.59	2.36	1.66-3.37
	0.5	1.13	1.05-1.21	1.48	1.06-2.07
	>0.5	1		1	
pH	<7.7	1.54	1.45-1.64	1.67	0.69-1.85
	7.7	1.05	0.98-1.14	1.13	1.07-2.60
	>7.7	1		1	
Chloride (mg Cl/L)	<55	1		1	
	55-200	1.19	1.09-1.30	0.95	0.51-1.76
	>200	1.60	1.52-1.68	1.77	1.32-2.38
Sodium (mg/L)	<50	1		1	
	50-100	1.46	1.35-1.57	1.10	0.74-1.64
	>100	1.66	1.57-1.74	1.81	1.33-2.46
Nitrate (mg NO ₃ /L)	<9	1		1	
	9-12	0.90	0.84-0.97	1.43	0.82-2.51
	>12	1.45	1.35-1.55	2.34	1.38-3.98
Aluminium (µg/L)	<40	1		1	
	40-50	1.21	1.13-1.29	1.08	0.82-1.41
	>50	0.80	0.74-0.87	0.93	0.61-1.42

TTHMs: total trihalomethanes. a. Models for bottled water to drink are adjusted by educational level. b. Models for bottled water to cook are adjusted by gender and age.

Table S1. Spearman correlation between the water quality parameters. Barcelona, 2003-2006.

	TTHMs	Conductivity	Chloride	Free chlorine	Sodium	pH	Nitrate	Aluminium	N samples
TTHMs	1								109
Conductivity	0.73**	1							109
Chloride	0.88**	0.85**	1						30
Free chlorine	-0.40**	-0.34**	-0.30	1					103
Sodium	0.64**	0.67**	0.75**	-0.37*	1				30
pH	-0.56*	-0.69**	-0.66**	0.29	-0.62**	1			30
Nitrate	0.78**	0.67**	0.76**	-0.38*	0.70**	-0.43*	1		30
Aluminium	-0.15	-0.14	-0.13	-0.32	-0.01	-0.03	-0.40*	1	30

TTHMs: total trihalomethanes. *pvalue<0.05 **p-value<0.001