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Significant variation in the concentration of carcinogenic polycyclic aromatic hydrocarbons in *yerba maté* samples by brand, batch and processing method

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Abstract

Drinking *maté*, common in southern South America, may increase the risk of esophageal squamous cell carcinoma (ESCC). In 2006, we found high but variable polycyclic aromatic hydrocarbon (PAH) content in commercial *yerba maté* samples from eight Brazilian brands. The PAH content of new samples from the same brands, purchased in 2008, and four brands from a single manufacturer processed in different ways, obtained in 2010, were quantified to determine whether PAH concentration was still high, PAH content variation was brand specific, and whether processing method affects PAH content of commercial *yerba maté*. Concentrations of individual PAHs were quantified using gas chromatography/mass spectrometry with deuterated PAHs as internal standards. Median total PAH concentration was 1500 ng/g (range: 625 to 3710 ng/g) and 1090 ng/g (621 to 1990 ng/g) in 2008 and 2010 samples, respectively. Comparing 2006 and 2008 samples, some brands had high PAH concentrations in both years, while PAH concentration changed considerably in others. Benzo[*a*]pyrene concentrations ranged from 11.9 to 99.3 ng/g and 5.11 to 21.0 ng/g in 2008 and 2010 samples, respectively. The 2010 sample processed without touching smoke had the lowest benzo[*a*]pyrene content. These results support previous findings of very high total and carcinogenic PAH concentrations in *yerba maté*, perhaps contributing to the high incidence of ESCC in southern South America. The large PAH content variation by brand, batch and processing method suggests it may be possible to reduce the content of carcinogenic PAHs in commercial *yerba maté*, making it a healthier beverage.

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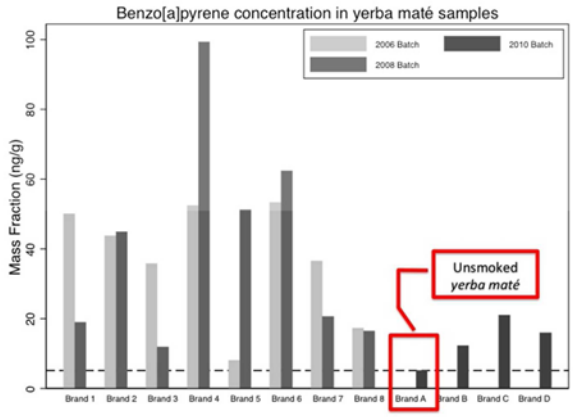

Keywords

Polycyclic Aromatic Hydrocarbons; *Yerba Maté*; Carcinogens; Esophageal Cancer; Benzo(α)pyrene; Processing Method; Lifestyle

Introduction

Esophageal cancer (EC), the sixth most common cause of cancer death in the world, has a striking geographical variation in both incidence and mortality [1]. Southern South America, including Uruguay, Paraguay, southern Brazil, and northern Argentina, is one of the areas with high incidence rates of EC [1, 2]. This geographic clustering suggests that some environmental factors and/or habits specific to this area are risk factors for esophageal squamous cell carcinoma (ESCC), the dominant histological type of EC in South America.

Previous research has shown that in addition to alcohol consumption and tobacco smoking, which are known to cause ESCC in many parts of the world, drinking *maté*, a habit common to southern South America, may also increase risk of ESCC [3–5]. *Maté* is a drink prepared from dried leaves of the plant *yerba maté* (*Ilex paraguariensis*), and it is consumed in high quantities by many people in this region [3]. The average per capita annual consumption of processed *yerba maté* leaves in this region has been estimated to be between 5 and 8 kg [6]. While a causal link between *maté* drinking and ESCC is not yet certain, it is highly probable. A number of studies have shown a consistent association between amount, duration, and temperature of *maté* consumption and risk of ESCC [3, 5, 7]. Two plausible mechanisms proposed for the carcinogenecity of *maté* are chronic mucosal irritation due to repeated thermal injury after drinking hot *maté*, and exposure to carcinogenic components of the drink, such as polycyclic aromatic hydrocarbons (PAHs) [3, 5, 7]. We focus on the latter mechanism in this paper.

PAHs are compounds produced during incomplete combustion of organic materials, including tobacco and coal. PAH exposure has been shown to be high in a number of areas with very high rates of ESCC [8–11]. Humans may be exposed to PAHs in many ways, including inhalation of tobacco smoke, exposure to air pollution, eating foods cooked at high temperature, jobs such as coal mining, and therapeutic use of coal tar [12]. Of these, smoking tobacco is the most common major source of PAH exposure. Recent evidence shows that *maté* drinking may also be a major source of PAH exposure, possibly causing as much exposure as smoking tobacco [8, 13]. *Yerba maté* leaves undergo several processing steps before they are sold commercially, including slow drying using wood smoke. The drying

process, which takes approximately 8 hours to 24 hours, can result in high concentrations of PAHs in commercial *yerba maté* [14, 15].

A study by our group in Brazil showed that consuming *maté* led to a strong dose-dependent increase in the urine concentration of 1-hydroxypyrene glucuronide (1-OHPG), a stable metabolite of PAHs [8]. In a second study, we measured the concentrations of 21 PAHs in the leaves of 8 commercial *yerba maté* brands and in hot and cold *maté* extracted from two of these brands, and we found very high PAH concentrations in all of the leaf and beverage samples. Of special interest, we found up to a 4-fold variation among brands in the PAH content of the *yerba maté* leaves [13]. This variation may be of public health importance, because if PAHs are responsible for the increased risk of ESCC associated with *maté* drinking, then it may be possible to reduce the cancer risk by processing *yerba maté* in ways that result in a lower PAH content. As a follow-up to this second study, we purchased new samples of the same brands of *yerba maté* and repeated the PAH measurements, to substantiate our previous findings of high concentrations and variation of PAHs in *yerba maté* and to learn whether the differences in PAH content were brand-specific or they simply reflected batch-to-batch variation within each brand. In addition, we obtained four different brands of *yerba maté* that were made by the same company but processed in different ways, including one brand that was never exposed to smoke during the processing of the *yerba maté* leaves, to see if such different processing methods might affect the PAH content of the commercial product.

Methods

In our previous study in 2006, we purchased eight packaged commercial brands of *yerba maté* in Santa Maria, Rio Grande do Sul (RS), Brazil, and measured 21 individual PAHs in the dry *yerba maté* leaves [13]. In 2008 we purchased new loose (unpackaged) samples of the same 8 *yerba maté* brands from a market in Porto Alegre, RS, and quantified the concentrations of 26 individual PAHs. In 2010 we obtained four additional packaged commercial brands of *yerba maté* leaves from one of the major *yerba maté* manufacturers, with a request to test these brands for PAH content. Three of these brands were processed in the traditional way (including smoking the leaves) and one was processed in an alternative way in which the leaves were never exposed to smoke. In these four brands, we quantified the concentrations of 22 individual PAHs. Of the individual PAHs measured in the 2006, 2008 and 2010 samples, 20 were the same among the three studies, and the rest were study specific. All measurements were performed in the Analytical Chemistry Division of the National Institute of Standards and Technology (NIST) using the same validated methods used for the 2006 samples [13]. Standard Reference Material (SRM) 2260a Aromatic Hydrocarbons in Toluene, SRM 2269 Perdeuterated PAH-I, SRM 2270 Perdeuterated PAH-II, and SRM 1649a Urban Dust were obtained from the Standard Reference Materials Group, NIST. One subsample (approximately 2 grams (g), exact mass known) from each brand of *yerba maté* from the 2008 samples or 3 subsamples (between 2 g and 4 g, exact mass known) from the 2010 *yerba maté* samples and one subsample (approximately 0.3 g, exact mass known) from one bottle of SRM 1649a were put into pressurized fluid extraction (PFE) cells containing hydromatrix (Isco, Lincoln, NE) and mixed with the hydromatrix. Hydromatrix was then used to fill the void space of the cells. A solution prepared from diluting SRM 2269 and SRM 2270 was added to each extraction vessel for use as the internal standard solution. The fact that only one subsample was tested from each of the 2008 samples was a limitation of the study, but this was unlikely to have affected our overall findings.

Since some fine *yerba maté* particles came through with the extract, the entire extract (approximately 15 milliliter (mL)) was eluted through a silica plus Sep Pak column (Waters

Corporation, Milford, MA) that was preconditioned with 20% dichloromethane in hexane (volume fraction) for the 2008 samples or 10% (volume fraction) for the 2010 samples. The *yerba maté* extract was then eluted from the Sep Pak using 20mL of the same solvent mixture. The eluents from the Sep Pak were concentrated to approximately 0.5 mL using an automated evaporation system prior to gas chromatography/mass spectrometry (GC/MS) analysis.

The column used for the GC/MS analysis was a 0.25 millimeter (mm) × 60 meter (m) fused silica capillary column containing a 50% phenyl methyl-substituted polysiloxane phase, 0.25 μm film thickness. All injections were done on-column (1 μL) with helium as a carrier gas at a constant flow rate of 1.2 mL/min.

A calibration curve bracketing the concentrations in the *yerba maté* samples (not forced through the origin) was determined for each analyte of interest by linear regression of response data from gravimetrically-diluted solutions of SRM 2260a mixed with the internal standard solution (prepared from SRM 2269 and 2270). These calibration solutions, as well as the blank extract (hydromatrix with internal standards spiked), were taken through the entire procedure (extraction, clean-up, and analysis).

Statistical Analysis

All PAH concentrations were rounded to three significant figures. For each of the measured PAHs, median concentrations were calculated separately for the samples purchased in 2008 and for the samples obtained in 2010. Pairwise Spearman correlation coefficients were calculated to compare the distribution of PAH concentrations in different brands. This analysis was done to examine whether there was a consistent pattern in the PAH concentrations of processed *yerba maté*. All analyses were done using Stata, version 11 (StataCorp. 2009. *Stata Statistical Software: Release 11*. College Station, TX: StataCorp LP.). Two-sided p-values less than 0.05 were considered as statistically significant.

Results and Discussion

The concentrations of the 20 PAHs analyzed in both the 2008 and 2010 samples are shown in Table 1. The median concentration of these 20 PAHs in the *yerba maté* samples purchased in 2008 was 1500 nanograms/grams (ng/g), with a range from 625 ng/g to 3710 ng/g. In the 2010 samples, the median concentration was 1090 ng/g, and the range was from 621 ng/g to 1990 ng/g. The concentration of the carcinogenic PAH benzo[*a*]pyrene ranged from 11.9 ng/g to 99.3 ng/g in the 2008 samples and from 5.11 ng/g to 21.0 ng/g in the 2010 samples. The lowest concentration of benzo[*a*]pyrene was found in the “Brand A” brand of the 2010 samples. The concentrations of total PAHs in the Brand C and Brand D brands of the 2010 samples were in the same range as most of the brands in 2006 and 2008, while the Brand A and Brand B brands appeared to be similar to the low-PAH brands in the previous series (Figure 1).

There was a strong and statistically significant positive pairwise correlation in the distribution of individual PAH concentrations between each two of the twelve brands purchased in 2008 and 2010 (Table 2). This indicates that the relative content of the individual PAHs in different *maté* brands was similar. For example, in the 2008 samples, benzo[*a*]pyrene made up approximately 2% (1.9% to 2.7%), fluoranthene 14% (10% to 17%) and phenanthrene 27% (22% to 33%) of the overall PAH content in all brands. These results suggest that: 1) we can measure a limited number of PAHs, e.g., 2 or 3, rather than 20, to obtain a good measure of the total PAH content of *maté*; and 2) the PAH content of mate may have one main source, which produces a similar distribution of PAHs.

In order to evaluate batch-to-batch variation of the same eight yerba *maté* brands in 2006 and 2008, we compared the absolute values of the total PAH concentrations of the 20 PAHs that were measured in both years (Figure 1). Some brands had high or low total PAH concentrations in both years. For example, Brand 6 had the second highest concentration of PAHs in both 2006 and 2008. Brand 8 had relatively low concentrations and ranked 7th in both years. However, some brands changed considerably in their rank. For example, Brand 1, which had the highest concentration of PAHs in 2006, ranked 6th in 2008. Conversely, Brand 5, which had the lowest concentration of PAHs in 2006, had one of the higher concentrations of PAHs in the 2008 samples. Examining individual PAHs, we also found high batch-to-batch variation in six of the eight brands, with some having higher and some lower concentrations in the new samples. For example, benzo[*a*]pyrene concentrations in the 2006 and 2008 samples were 50.0 and 18.9 ng/g for Brand 1, and 52.5 and 99.3 ng/g for Brand 4 (Figure 2).

The lowest concentration of benzo[*a*]pyrene in any of the *yerba maté* samples, 5.11 ng/g, was found in the 2010 brand which was processed without ever touching smoke. Figure 2 shows the concentrations of benzo[*a*]pyrene in the 2006, 2008 and 2010 *yerba maté* samples, with the horizontal line denoting the benzo[*a*]pyrene concentration of the brand prepared without touching smoke.

Our analysis of the commercial *yerba maté* samples purchased in 2008 and 2010 supports our previous findings of very high total and carcinogenic PAH concentrations in most commercial *yerba maté*, and shows again that there is a consistent pattern to the distribution of the PAHs present in the leaves. Comparing the 2006 and 2008 samples, we observed relatively large batch-to-batch differences in most brands. And in the 2010 samples, we saw differences in PAH concentrations made by the same company but processed in different ways.

PAH contamination of *yerba maté* may come from several sources, including environmental pollution, but probably the most important source is the traditional way of drying the leaves, during which the leaves are smoked [14, 15]. PAH contamination of other foods can happen during preparation [16–18], and it has been shown that *yerba maté* leaves acquire PAHs during the drying process [15]. Of all of the *yerba maté* samples tested in all three years, the concentration of benzo[*a*]pyrene was the lowest in the 2010 brand that had never touched smoke during the drying process, suggesting that this method of processing may be a promising one for reducing the PAH content of commercial *yerba maté*.

The high PAH content found in *yerba maté* leaves in this study and in our previous study supports a role for PAHs in the etiology of ESCC. The IARC working group on the evaluation of carcinogenic risks to humans has classified benzo[*a*]pyrene as “carcinogenic to humans” [12], and exposure to this carcinogenic PAH has been shown to be high in several high-risk areas for ESCC [8, 10, 11, 19]. Furthermore, we have previously shown that the usual practice of repeated infusions of the same *yerba maté* leaves can elute more than 50% of the benzo[*a*]pyrene content of the leaves into either hot or cold *maté* drinks [13].

In southern Brazil, *maté* is traditionally prepared and drunk in gourds called “cuias”. These cuias usually hold 100–200 mL of liquid, and are filled with *yerba maté* leaves up to two-thirds of their volume, about 50–100 g of leaves [20]. The 16 traditional *yerba maté* brands that we analyzed in 2006 and 2008 had a median benzo[*a*]pyrene concentration of 40.1 ng/g of leaves. If we round this to 40 ng/g and assume an average use of 50 g of leaves per cuia and a 50% elution of the benzo[*a*]pyrene from the leaves into the fluid, then drinking an average cuia of *maté* in the traditional way would expose the consumer to 40 ng/g × 50 g × 0.5 elution = 1000 ng of benzo[*a*]pyrene, equivalent to the benzo[*a*]pyrene content of the

smoke from 100 cigarettes (five packs)[21]. In contrast, if the benzo[*a*]pyrene concentration of the *yerba maté* leaves was equal to that of the 2010 brand that never touched smoke, approximately 5 ng/g of leaves, then drinking the same cuia in the same way would expose the consumer to 5 ng/g × 50 g × 0.5 elution = 125 ng of benzo[*a*]pyrene, equivalent to the benzo[*a*]pyrene content of the smoke from 12.5 cigarettes.

One must be cautious, however, in implying similar biological effects from similar amounts of external PAH exposure from *maté* drinking and cigarette smoking, since the internal exposure of this intake may differ by the route of administration (gastric vs. pulmonary absorption) [12]. It is possible that gastric absorption may be less efficient, and thus the biological effect per ng of gastric exposure may be less. This possibility is supported by the fact that in the same population, urinary 1-OHPG, a valid short-term marker of total internal PAH exposure, has been shown to be almost equal between cigarette smokers (smoking an average of 15 cigarettes/day) who do not drink *maté* and *maté* drinkers (drinking an average of 840 mL/day) who do not smoke cigarettes [8]. But whatever the exact equivalency, a substantial reduction in external PAH exposure from any source will most likely be beneficial to individual and public health.

Given the high prevalence of *maté* consumption in certain areas of South America [6, 22, 23], the population-attributable fraction of ESCC due to *maté* drinking has been estimated to range from 10% to 79% [3, 5, 24], which implies that reducing consumption or reducing the carcinogens in it could potentially lead to a substantial decrease in the burden of ESCC. While implementing strategies to eliminate *maté* drinking in this region may not be feasible (or desirable), diminishing the carcinogenic content of this common exposure would be a plausible preventive approach. Diminishing the PAH content of maté could also reduce other adverse effects of PAH exposure, including its effects on cardiovascular diseases [25].

We observed large differences in PAH concentrations between brands and within each brand of *yerba maté* over time. This is not surprising, since it is difficult to control the exact amount of smoke that the *yerba maté* leaves are exposed to during the traditional drying process. But these differences by brand and batch suggest that very high PAH content is probably not essential for marketing *yerba maté*, so it may be possible to reduce the PAH content of processed *yerba maté* without significantly affecting its quality or marketability. The lower levels of total PAH and carcinogenic benzo[*a*]pyrene in the commercial *yerba maté* brand that had not been exposed to smoke are consistent with this hypothesis.

It is probable that most commercial *maté* producers, along with the general public, have not been aware that *maté* drinking currently has health risks as well as health benefits, and that at least some of these risks might be avoided if the drying methods were changed. It is also possible that there may be a difference in taste after different processing methods, which may make long-time *maté* drinkers not want to switch to a “smokeless” brand, but this has not yet been evaluated. The best solution would be to find processing methods that could significantly reduce PAH exposure and still keep optimal taste.

In summary, the results of this study confirm high concentrations of total and carcinogenic PAHs in most commercial *yerba maté* leaves, and suggest that these concentrations may possibly be reduced by altering the processing of the leaves. Reducing the PAH content of commercial *yerba maté* is a sensible precaution which will probably make *maté* a healthier beverage. Given the popular use of *maté,* the public health impact of such a reduction could be significant.

Acknowledgments

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Abbreviations

EC	Esophageal cancer
ESCC	Esophageal squamous cell carcinoma
PAHs	polycyclic aromatic hydrocarbons
1-OHPG	1-hydroxypyrene glucuronide
NIST	National Institute of Standards and Technology
SRM	Standard Reference Material
g	grams
PFE	pressurized fluid extraction
GC/MS	gas chromatography/mass spectrometry
mL	milliliter
mm	millimeter
m	meter
ng/g	nanograms/grams

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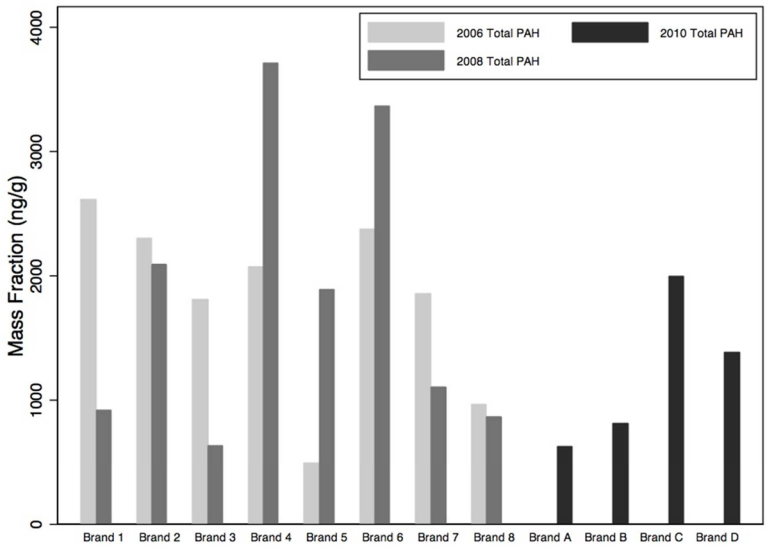

Figure 1.
Total PAH concentrations in the 2006, 2008 and 2010 *yerba maté* samples.

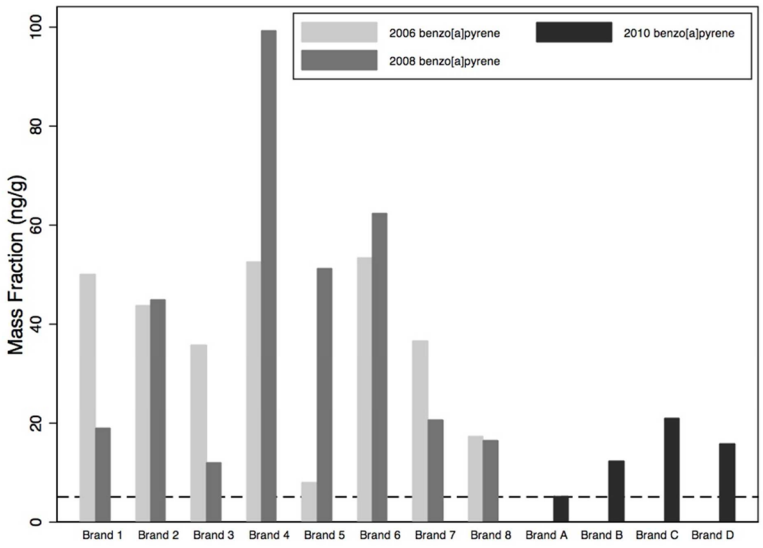

Figure 2.
Benzo[*a*]pyrene concentration in the 2006, 2008 and 2010 *yerba maté* samples. The dashed line shows the benzo[*a*]pyrene content of the *yerba maté* brand that never touched smoke (Brand A).

Table 1

Concentrations (ng/g) of the 20 PAHs in all samples of the commercial brands of *yerba maté* leaves purchased in 2008 and 2010

	2008									2010				
	Brand 1	**Brand 2**	**Brand 3**	**Brand 4**	**Brand 5**	**Brand 6**	**Brand 7**	**Brand 8**	**Median**	**Brand A**	**Brand B**	**Brand C**	**Brand D**	**Median**
Naphthalene	105	230	156	233	227	341	302	187	229	48.0	50.1	84.9	73.4	61.8
Fluorene	ND	71.7	23.4	67.0	31.4	114	49.4	34.4	49.4	4.50	10.2	17.7	9.63	9.92
Phenanthrene & Anthracene	264	626	153	1110	447	1010	284	296	372	182	268	624	528	398
Fluoranthene	156	315	72.2	634	307	564	132	85.5	232	82.2	114	309	214	164
Pyrene	145	295	65.6	516	316	446	113	79.4	220	70.1	105	318	197	151
Benzo[*ghi*]fluoranthene	24.2	45.5	11.3	97.2	59.6	86.2	19.3	ND	45.5	9.15	9.73	56.8	6.91	9.44
Benzo[*c*]phenanthrene	6.62	26.5	ND	51.5	12.8	46.0	9.21	ND	19.7	ND	5.52	16.1	6.08	6.08
Benz[*a*]anthracene	63.7	62.0	46.3	206	92.4	204	63.3	53.0	63.5	111	84.8	109	122	110
Triphenylene & Chrysene	39.5	139	30.5	271	102	234	52.0	42.0	77.0	33.1	36.6	112	11.3	34.9
Benzo[*b*+*j*+*k*]fluoranthene	32.5	86.6	18.5	190	68.8	120	38.6	27.9	53.7	23.6	45.4	113	55.5	50.5
Benzo[*a*]fluoranthene	4.20	10.6	ND	22.6	7.73	22.3	4.89	ND	9.17	12.2	14.9	7.28	31.0	13.6
Benzo[*e*]pyrene	14.1	33.4	7.84	74.1	33.7	41.4	13.6	10.6	23.8	23.4	27.8	97.1	48.7	38.3
Benzo[*a*]pyrene	18.9	44.9	11.9	99.3	51.2	62.3	20.6	16.4	32.8	5.11	12.3	21.0	15.9	14.1
Perylene	4.33	9.38	ND	15.0	9.42	8.48	3.44	ND	8.93	3.94	3.95	10.9	8.16	6.06
Indeno[1,2,3-*cd*]pyrene	15.2	44.0	10.2	60.7	43.7	30.8	ND	9.23	30.8	ND	ND	35.5	21.0	28.3
Benzo[*ghi*]perylene	22.8	49.8	18.0	67.2	77.1	32.6	ND	18.8	32.6	13.0	19.7	60.3	33.4	26.6
Total	916	2090	625	3710	1890	3360	1110	860	1500	621	808	1993	1382	1090

The concentrations of the 20 PAHs are given in 16 rows because the concentrations of phenanthrene and anthracene were reported together, the concentrations of triphenylene and chrysene were reported together, and the concentrations of benzo[*b*]fluoranthene, benzo[*j*]fluoranthene and benzo[*k*]fluoranthene were reported together as benzo[*b+j+k*]fluoranthene.

ND, not detected

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Table 2

Pairwise correlations of the distribution of the PAHs in the 2008 and 2010 *yerba maté* samples

		2008								2010			
		Brand 1	**Brand 2**	**Brand 3**	**Brand 4**	**Brand 5**	**Brand 6**	**Brand 7**	**Brand 8**	**Brand A**	**Brand B**	**Brand C**	**Brand D**
2008	**Brand 1**	1											
	Brand 2	0.87	1										
	Brand 3	0.87	0.97	1									
	Brand 4	0.95	0.93	0.91	1								
	Brand 5	0.97	0.93	0.92	0.96	1							
	Brand 6	0.88	0.95	0.94	0.95	0.90	1						
	Brand 7	0.79	0.88	0.91	0.89	0.80	0.96	1					
	Brand 8	0.85	0.95	0.98	0.91	0.91	0.93	0.90	1				
2010	**Brand A**	0.90	0.82	0.83	0.90	0.88	0.82	0.79	0.85	1			
	Brand B	0.87	0.86	0.85	0.91	0.87	0.87	0.84	0.89	0.97	1		
	Brand C	0.91	0.88	0.82	0.93	0.92	0.86	0.74	0.84	0.89	0.89	1	
	Brand D	0.79	0.74	0.75	0.77	0.78	0.70	0.68	0.80	0.91	0.92	0.82	1

The P-value for all the Spearman correlations was less than 0.01 except for the correlation between brand D and brand 6 and 7.

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