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Phenolic substances in food – analytical survey of 11 phenols in Swedish Market Basket samples from 1999, 2005 and 2010

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Phenolic substances in food – analytical survey of 11 phenols in Swedish Market Basket samples from 1999, 2005 and 2010

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Sammanfattning
En tidigare analys av fenolära ämnen i matkorgsprover från 2005 visade att kvantitativa data erhölls för vissa substanser och vissa matriser, medan samtidigt många av fenolerna hade halter under detektionsgränsen (LOD). I den nya, nu presenterade undersökningen analyseras fler fenolära ämnen (11 st), matkorgsprover från 1999, 2005 och 2010 analyseras samtidigt, och analyskänsligheten är i många fall förbättrad. De analyserade ämnena är 4-bromfenol (4-BrP), 4-tert-butylfenol (4-t-BP), 2,6-di-tert-butyl-4-metylfenol (BHT), tert-butyl-4-hydroxyanisol (BHA), 2,4-dibromfenol (2,4-DBP), 4-tert-oktylfenol (4-t-OP), 4-nonylphenol (4-NP), 2,4,6-tribromfenol (TBP), pentaklorfenol (PCP), triclosan (TC) och bisfenol A (BPA).
De undersökta matkorgsproverna kommer från studier där matkassar från vanliga livsmedelskedjor i Sverige tas in för analys, och där de insamlade livsmedlen analyseras gruppvis (ex kött, fisk, mejeriprodukter, spannmål) för en mängd näringsämnen men även toxiska ämnen. Med hjälp av försäljningsstatistik kan konsumtionen av olika livsmedelsgrupper beräknas, och tillsammans med haltdata kan per capita-intaget för dessa ämnen tas fram. Matkorgsundersökningarna från 1999, 2005 och 2010 gjordes enligt samma metod, vilket gör att förändringar i exponering för ämnena över tid kan undersökas.
Halterna av fenolära ämnen i prover från 12 olika livsmedelsgrupperna visar att halterna i flera fall ligger under LOD, men att <LOD-värden för olika fenolära ämnen gäller skilda livsmedelsgrupper. Av de 25 analyserna för varje fenolärt ämne låg 7-25 under LOD för prover från Matkorgen 2010. Vid beräkningar av intag av dessa fenolära ämnen via kosten, med användande av matkorgsmetodik, erhålls per capitaintag från 44 ng/person/dag (2,4-DBP) till 15 500 ng/person/dag (4-NF). För BPA erhålls ett intag på 2 460 ng/person/dag, dvs ca 37 ng/kg kroppsvikt/dag (vid medelvikt 67,2 kg), och detta kan ställas i relation till EFSAs TDI på 5 mikrog/kg kroppsvikt och dag. För övriga fenolära ämnen finns dock inga motsvarande hälsomässiga referensvärden som man kan jämföra mot, och detta hämmar förstås riskbedömningen. Avsaknaden av data som antyder trender I halter över tid kan bero på de få datapunkterna från varje ämne som insamlas per år, trots att de analyserade proverna var blandningar av ett större antal matkassar.

Introduction

Phenolic substances present in food are not commonly analysed, and relatively few results are found in scientific literature. One exception is bisphenol A (BPA), a recently much debated substance that could leak from plastics used in food contact materials and thus may be found in certain food items. Also, some phenols are used as food additives (BHT, BHA – antioxidants) and their presence in food is expected. However, other phenolic substances present in our environment may also end up in food, even if they are not directly used in food production and processing. In these cases, their presence in food could be a result of a contamination along the food distribution pathway, including food packaging materials.

If we have little information on levels of phenolic compounds in our food, even less is known of possible health effects. We know that BPA is suspected be a hormone-disrupting chemical, but effects of other phenols are not much studied. In *in vitro*-studies, the estrogenic potential of halogenated phenolic chemicals was tested, and some of them expressed certain estrogenic activities (Körner et al., 1998; Schultz et al., 2002). Also the thyroid hormone system may be affected by halogenated phenolic compounds, as suggested in several reports (Butt et al., 2011; Kudo et al., 2006; Shimizu et al., 2013; van den Berg et al., 1991), and several brominated flame retardants are indeed believed to exert some of their hormonal effects after metabolism to phenolic transformation products (Brouwer et al., 1998).

Measureable levels of environmental phenolic compounds have been shown to occur in human samples of serum (Meijer et al., 2008; Glynn et al., 2011; Gyllenhammar et al., 2012; Fujii et al., 2014), breast milk (Fujii et al., 2014) and amniotic fluid and urine (Philippat et al., 2013). In some of the studies, dietary exposure of corresponding substances were also estimated (Gyllenhammar et al., 2012; Fujii et al., 2014). The measured compounds are differing between studies, which makes it difficult to compare between studies/countries.

In an earlier study on Market Basket food samples collected in 2005, selected phenolic compounds were analysed in pooled food group samples obtained from four Swedish towns (Ankarberg et al., 2006). The selection of compounds was based on our own toxicological and chemical expertise, with guidance from the present scientific literature. The phenolic compounds analysed at that time were 2,6-di-*tert*-butyl-4-methylphenol (BHT), *tert*-butyl-4-hydroxyanisol (BHA), 4-nonylphenol (4-NP), 4-tert-oktylphenol (4-t-OP), pentachlorophenol (PCP), triclosan (TC) and bisphenol A (BPA). The analyses were performed at IVL, Stockholm, with financial support of the Swedish EPA.

Results from analyses of the 2005 market basket samples showed comparably low levels, and many levels were below the limit of quantification (LOQ), but there were several interesting findings. BPA resulted in clearly measurable levels in certain food groups, primarily those of animal origin (fish and meat: 2.5-30 ng/g fresh wt.), BHA had high levels in certain, but not all, fish samples (up to above 3 000 ng/g fresh wt.), and PCP gave measurable levels in most fish and meat samples. 4-NP resulted in measurable levels I certain vegetable samples (fruit, vegetables, potatoes), whereas the rest of the studied phenolic compounds (BHT, 4-t-OP,TC) only in a few cases had levels above LOQ.

In the present study, we have extended the scope of the earlier reported investigation, and the aim is now to report on food levels of an extended number of phenolic compounds (n=11) and also to compare intake levels of these phenols from 1999, 2005 and 2010 market basket studies, with the ambition to study possible trends. In addition to the compounds studied earlier (BHA, BHT, 4-NP, 4-t-OP, PCP, TC and BPA) the following ones were added to analytical scheme: The brominated phenols 4-bromophenol (4-BrP), 2,4-dibromophenol (2,4-DBP) and 2,4,6-tribromophenol (TBP) (as EU has interest in these compounds; EFSA 2012), and also 4-tert-butylphenol (4-t-BP). Estimated intake levels of the analysed phenols were compared with literature and health reference data, if present. The analyses were also this time performed at IVL, Stockholm.

Materials and Methods

Food samples

The food samples for analyses were taken from the sample bank of Market Basket samples at NFA, comprising samples obtained in 1999, 2005 and 2010. In short, homogenates of each food group (12 groups, ca 50 g each) were produced, and these were pools from all the grocery shops/chains included in the respective project round. In case of the 2010 sampling, two homogenate pools from each food group were produced, defined as standard price and low price alternatives. From certain homogenates, double samples were taken to check matching of analytical result. In all, about 50 food samples were analysed. More information on the Swedish market basket studies are given in separate articles and report (Darnerud et al., 2006; Törnkvist et al., 2011; NFA, 2012).

Analytical details

In the present project, the food samples, comprising different food matrices, were analysed for selected phenolic compounds, listened in Table 1. In this table, limits of quantification and detection are given. Within this analytical task, new LOQ for BHT and BHA was produced. The analyses were performed at IVL, Stockholm.

Final analytical determination was made by GC-MS/MS, in MRM (multi reaction monitoring) mode in case of non-halogenated compounds, and in NCI (negative chemical ionization) mode in case of halogenated compounds.

The following quality criteria were followed for optimal identification and quantification of the analytes: a) Retention time not more than ± 0.05 min from reference standard, b) the ratio of chosen MRM ion signals should be not more than ± 15 % of reference standard, and c) the limit of quantification should be defined as signal-noise relationship equal to or more than 10:1 (LOQ).

To minimize problem with high blank values, all glass material and chemicals (NaSO4, NaCl) that are used in analytical work were heated to +400 ˚C. All other material that cannot be heated was rinsed thoroughly with solvent before usage. Columns and solvent used in extraction and separation steps were controlled before usage.

Analytical blanks were used for background determination, and background levels were subtracted from registered values. Internal standards were used to correct for losses during sample extraction and separation. Repetitiveness was checked by analyzing parallel samples.

Some samples from 1999 were stored in plastic vials. To check the potential migration of the studied phenolic compounds from vials to food, empty plastic vials were extracted with solvents and the resulting extracts were analysed.

Results

A full overview of the analytical result is given in Table 2. The table gives information on the levels (in ng/g fresh wt.) in the 12 different food homogenates from all three samples years, for all the 11 phenolic compounds included in this study. It should be noted that samples between LOD and LOQ are given in red, whereas levels beneath LOD are shown by "<" characters (the LOD and LOQ levels are given in Table 1). Duplicate samples of homogenates of meat (2010 N), fish (1999), and vegetables (2010 N) were included in the analyses.

Results from extraction of empty plastic vials with solvents resulted in low or non-detectable levels of the actual compounds, and migration from these vials were therefore considered not to interfere with the analytical results.

In Table 3, the levels of phenolic compounds in 2010 market basket food groups were presented, expressed as lower, median and upper bound values. Depending on the sensitivity of the specific analysis and the number of samples beneath the limit of detection, LB and UB vales for a specific food group and compound could vary considerably (when analyses are <LOD, LB values will be zero). When all analyses for a specific food group are above LOD, only one value is presented (presented as an MB value in the table). In case of 4-t-OP, all analyses from 2010 are below LOD, and for the other compounds a varying number of results <LOD are found (7-23, of totally 25 analyses).

Data in Table 4 give the estimated per capita intake of the studied phenolic compounds, presented as the contribution per food group and as the total intake by addition of all 12 groups present in the market basket study. The per capita consumption of these food groups, based on statistics from the Swedish Board of Agriculture (see Table 5), was multiplied with the phenolic compound level of the respective group. The resulting data show the population (per capita) average intake of these compounds, and consequently neither the range in intake nor specific consumer groups could be studied with this method. By adding all 12 food groups together, the total intake can be estimated. Of the studied compounds, the estimated total intake was highest in case of 4-NP (15 500 ng/person/day), about three times the intake of the second highest, BHA, and six times the intake of BHT, 4-t-OP and BPA.

In order to give further information on the relative contribution of different food groups to the total intake of a specific compound, the data from Table 4 could be visualized as circle diagrams, as made for BHA, 4-NP, PCP and BPA in Figure 1. By studying these diagrams it is clearly shown that depending on studied compound, different food groups are major food contributors (e.g. BHA – fish;

4-NP – cereals and fats; PCP – cereals; BPA – vegetables). Concerning BPA, apart from the vegetable contribution all other food groups contribute rather equally, indicating that this compound may be a widely distributed in our environment, with the possibility to contaminate many different food groups.

The potential difference in intake levels over time was studied by comparing intake data from the market baskets from 1999, 2005 and 1010, based on MB levels (all samples analysed at the same lab, and at the same time) (Table 6). For visualisation, data for some of the compounds is also presented in Figure 2. Some changes are visible between sampling time points, but obvious trends are difficult to identify. In relative numbers, between 1999 and 2010 per capita intake levels of five compounds decreased to 60-85% of the initial 1999 values, whereas the six other showed increased intake levels (105-225%) during the same period. It could aslo be noted that some compounds (among them BHA, see figure 2) for some unknown reason showed considerably higher intake levels in the 2005 samples compared to both 1999 and 2010 levels (Table 6).

Discussion

In this paper, results on levels of 11 exogenous phenolic compounds in food samples from Swedish Market Basket studies from 1999, 2005 and 2010 are presented, and based on these data estimations of food intake of the compounds have also been performed. One additional aim of the study was to study potential time trends in intake by using the three market basket sampling occasions, but the small data set and the fact that levels in several cases were below LOD gave only limited information as regards differences in intake over time.

As was seen in the data presentation, the analyses of phenols in the different food groups resulted in several cases in levels below LOD (see bottom of Table 3). In this case LB levels were equal to zero and there was a clear difference between LB and UB levels (in one case (4-t-OP), the 2010 levels in food were beneath LOD in all 25 analyses!). Consequently, depending on LB or UB levels the estimations, e.g. of intakes, performed on these data will vary to a certain extent.

Using the market basket approach and the new analytical data on phenolic compounds, estimation of the population mean, per capita, intake for the average Swedish consumer could be performed. For many of the studied compounds estimation of total diet exposure for Swedish consumers was done for the first time. It should however be noted that due to the many analytical results below LOD, these estimations are only approximate.

The estimated intake values could be used in risk evaluation against defined reference values for health effects. However, such reference values are rarely found for this type of phenolic compound, and may, to our knowledge, only be present in case of BPA.

Data on specific phenols

The group of three brominated phenols, 4-Br-P, 2,4-DBP and TBP, is of interest as the included compounds could occur in our environment both as a consequence of human pollution by e.g. PBDEs and similar preparation, but also as products formed from certain marine algae (e.g. Löfstrand et al., 2010). In the Nordic environment, the presence of brominated phenols has been investigated (Schlabach et al., 2011). In a Japanese duplicate diet study, the dietary intake of organohalogen phenols was followed (Fujii et al., 2014). The intake was dominated by TBP, followed by tetrabromo bisphenol A (TBBPA) and 6-OH-BDE47. The intake levels of brominated phenols found in the present study are low when compared to the other analysed phenols (ca 150 ng/person/day, sum of three BPs), but there are no reference intake values to compare against.

In a study on dietary intake of BHT and BHA in Korea, Suh et al. (2005) could find no detectable levels of BHA, whereas the estimated intake of BHT was set to 15.6 ng/kg body wt./day. In a study on Lebanese students (Soubra et a.,l 2007) the estimated mean intake BHT and BHA was considerably higher: 25 and 150 microg/kg body wt., respectively. In both these studies, the estimated mean intakes were lower than the ADI's for BHA and BHT set by EU (0-0.5 and 0-0.05 mg/kg bw, respectively), but a fraction of the Lebanese population will have an intake above ADI. Our own estimated per capita intakes, 40 (BHT) and 75 (BHA) ng/kg bw/day, are well below these ADIs.

In our study 4-NP was found in relatively high levels in some food groups, especially fats (ca 350 ng/g fresh wt.) According to German data, this compound is ubiquitous in food, and found in quantifiable levels in 80% of all food samples. According to the German estimations, the mean intake of 4-NP for the German consumer was 7.5 microg/person/day (Guenther et al., 2002; Raecker et al, 2011), which could be compared with the present Swedish intake estimate of about twice as much, i.e. 15.5 microg/person/day.

PCP is found in comparably high levels in blood in humans, and this has been shown in women by both Swedish and Norwegian data (Meironyté Guvenius et al., 2003; Glynn et al., 2011; Rylander et al., 2012). Regarding food intake, the present intake estimation result in a much higher intake (610 ng/person/day) as compared to Japanese intake estimation based on a double portion method (2-5 ng/person/day) (Fujii et al., 2014). The reason for this large discrepancy is not known, but may be a combination of choice of food packaging materials in the two countries and of procedures for food sampling and preparation.

TC, a compound with antibacterial properties, has found wide-spread use in a variety of consumer products such as toothpastes, deodorants and soaps. In recent years, the use of this compound has been restricted due to discussion of health issues. The presence of TC in plasma and breast milk has been observed in Swedish women (Allmyr et al., 2006) and these levels could be associated to the use of personal care products. In Swedish food the levels of TC are low, and often under LOD, and the calculated low food intake is therefore not very accurate.

BPA is a compound that has been under intense debate during the last years, due to its proposed endocrine disrupting properties and at what exposure this may occur. Therefore, it is of interest to compare the estimated food intake of BPA in this study with intakes results from other studies. In a US study using NHANES data as a base for the calculation (LaKind and Naiman, 2011), the estimated intake for the whole population was 34 ng/kg bw/day, which fits very nicely to our present estimation of 37 ng/kg bw/day. Also, a Japanese estimation resulted in a BPA intake (for high-exposure populations) of 37-64 ng/kg bw/day for men and 43-75 ng/kg bw/day for women, i.e. relatively near both the Swedish and US levels. As a comparison, EFSA has recently recommended a new temporary ADI of 5 microg/kg bw/day.

Changes over time

The estimated intake of phenolic compounds in food were shown to change quite markedly for some compounds between the investigated time points, whereas other compounds were more stable over time (Table 6, Figure 2). In case of observed changes between years it was difficult to note any stable trend, and the 2005 levels seemed to differ the most compared to other time points. The lack of visible time trends in intake could also be a consequence of the few data points sampled per year, in spite of that the analysed homogenates were mixtures from a number of food baskets. To conclude, the present data cannot confirm any distinct (increasing or decreasing) time trend for the studied compounds, but future studies may increase and refine the basis for trend analysis.

Uncertainties in the analyses and estimations

As already mentioned, a number of chemical analyses showed results below LOD, and in four cases (4-BrP, 2,4-DBP, 4-t-OP, TC) 20 or more of a total of 25 analyses, resulted in registered signals below LOD. This fact will increase the uncertainty of presented results, but it should however be noted that these four compounds gave a relatively small intake, in comparison to other phenolic compounds. To check the accuracy of the chemical analyses, three pairs of duplicate samples were analytically tested (meat 2010 N; fish 1999; vegetables 2010 N). In cases when both values showed results above LOD, the duplicate results correlated moderately well (up to 2.5 times the difference between samples, except for one case (Fish, BHA) where difference was above 6 times). In case of the 2005 Market Basket samples it was of interest to compare earlier IVL analysis of phenolic compounds with the new, present analysis performed at the same laboratory. However, as many of the detection limits were changed such a comparison will be of limited value. Nevertheless, when calculated as intake values based on MB results, the agreement (ratio) between results were good in case of 4-NP and PBA (1.2-1.6), moderate in case of BHT, 4-t-OP, and TC (4-5.7), and weak in case of BHA and PCP (8 and 12). It should also be noted that these comparisons could be hampered by changes in food consumption between the two time points. Lastly, the studied compounds are in several cases ubiquitously found in environment and these compounds might contaminate the food samples at the sampling occasion or during further preparation of the sample. However, the relative contribution of different food groups to the estimated total intake, that are very different from compound to compound, speaks against such a general contamination hypothesis.

Comments on possible combinational effects

The above presentation has discussed levels of phenolic compounds one by one and compared intake levels with ADI or similar, when such reference levels are present. However, as we have shown in this report we are exposed to a number of exogenous phenolic compounds and there are of course many more of these compounds in food, and consequently also in our body, than those studied at present. Indeed, a Swedish study on blood donors showed that over 100 phenolic halogenated compounds (PHC) were analytically indicated in plasma, of which two major compounds were 2,4,6-tribromophenol (TBP) and pentachlorophenol (PCP) (Hovander et al., 2002). In case of at least some of the halogenated phenols, mechanistic studies have shown that they can interfere with thyroid hormones by several modes (van den Berg et al., 1991; Kudo et al., 2006; Shimizu et al., 2013), and disturbances in thyroid hormone homeostasis has been shown in animals and suggested in humans as a result of exposure to persistent halogenated aromatic hydrocarbons (Brouwer et al., 1998). Exogenous phenols, among of which some analysed in the present study (e.g. BPA), may disrupt also other hormonal systems such as those related to sex hormones (e.g. Körner et al., 1998). Consequently, we may have to give more interest in this group of compounds found in environment and in food, and learn more about their individual effects, but it is even more important to find out potential health consequences of their combinative effects (e.g. Meulenberg, 2009).

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Table 1. Limits of detection (LOD) and quantification (LOQ) (ng/g fresh weight), specified for different sample types, with abbreviations of analysed phenolic compounds below

LOD/LOQ specification	4-BrP	4-t-BP	BHT	BHA	2,4-DBP	4-t-OP	4-NP	TBP	PCP	TC	BPA
LOD 10 g	0.03	0.2	0.5	0.1	0.03	2	3	0.03	0.1	0.1	1
LOD 5 g	0.07	0.5	1	0.14	0.06	4	7	0.06	0.2	0.2	2
LOD fat samples (1 g)	0.3	2	1	0.7	0.3	20	30	0.3	1	1	10
LOQ 10 g	0.1	0.7	1.5	0.2	0.1	5	10	0.1	0.3	0.4	2
LOQ 5 g	0.2	1.4	3	0.4	0.2	10	20	0.2	0.6	0.8	4
LOQ fat samples (1 g)	1	7	15	2	1	50	100	1	3	4	20
LOD beverages 50 ml	0.007	0.05	0.1	0.01	0.006	0.3	0.7	0.006	0.02	0.02	0.2
LOQ beverages 50 ml	0.02	0.1	0.3	0.04	0.02	1	2	0.02	0.06	0.06	0.6

4-BrP = 4-bromophenol
4-t-BP = 4-tert-butylphenol
BHA = tert-butyl-4-hydroxyanisol
BHT = 2,6-di-tert-butyl-4-metylfenol
2,4-DBP = 2,4-dibromophenol
4-t-OP = 4-tert-octylphenol
4-NP = 4-nonylphenol
TBP = 2,4,6-tribromophenol
PCP = pentachlorophenol
TC = triclosan
BPA = bisphenol A

Table 2. Levels of phenolic compounds in Swedish market basket samples from 1999, 2005 and 2010 (ng/g fresh wt.). Values between LOD and LOQ are shown in red

IVL Id	Sample NFA	Year (3	Vial	4-BrP	4-t-BP	BHT	2/3-BHA	2.4-DBP	4-t-OP	4-NP	TBF	PCP	TC	BPA
2298	To11:13 Slurry 1 Cereales	1999	*plastic*	0.04	1.39	6.75	0.51	<0.03	<2	16.27	0.07	1.16	0.22	<1
2315	To11:28 Slurry 1 Cereales	2005	Glass	0.05	1.51	1.11	0.38	<0.03	<2	18.27	0.10	1.12	0.13	<1
2327	To11:33 N 1 Cereales	2010_N	Glass	0.42	1.29	1.78	<0.07	<0.03	<2	17.90	0.11	0.93	<0.1	<1
2341	To11:33 L 1 Cereales	2010_L	Glass	0.03	1.13	0.56	<0.07	<0.03	<2	29.81	0.10	0.78	<0.1	<1
2299	To11:13 Slurry 2 Pastries	1999	Glass	<0.07	1.34	10.00	5.95	<0.06	<4	7.17	<0.06	0.46	0.24	2.17
2316	To11:28 Slurry 2 Pastries	2005	Glass	<0.07	5.65	1.13	7.94	<0.06	<4	<7	<0.06	0.63	<0.2	<2
2328	To11:33 N 2 Pastries	2010_N	Glass	<0.03	2.74	1.39	24.27	<0.03	<2	<3	0.04	0.37	<0.1	6.58
2342	To11:33 L 2 Pastries	2010_L	Glass	<0.03	4.71	<0.5	0.60	0.03	<2	10.92	0.05	0.42	<0.1	8.23
2303	To11:13 Slurry 5 Dairy pr.	1999	*plastic*	<0.03	<0.2	<0.5	<0.07	<0.03	<2	<3	<0.03	0.11	<0.1	<1
2319	To11:28 Slurry 5 Dairy pr.	2005	Glass	<0.03	<0.2	<0.5	0.23	<0.03	<2	<3	<0.03	2.38	<0.1	1.78
2332	To11:33 N 5 Dairy pr.	2010_N	Glass	<0.03	<0.2	<0.5	<0.07	<0.03	<2	<3	<0.03	0.13	<0.1	<1
2345	To11:33 L 5 Dairy pr.	2010_L	Glass	<0.03	<0.2	1.47	<0.07	<0.03	<2	<3	<0.03	0.31	<0.1	<1
2304	To11:13 Slurry 6 Egg	1999	*plastic*	<0.03	2.93	<0.5	<0.07	<0.03	<2	<3	<0.03	0.27	0.20	1.24
2320	To11:28 Slurry 6 Egg	2005	Glass	<0.03	<0.2	<0.5	<0.07	<0.03	<2	<3	<0.03	0.13	<0.1	<1
2333	To11:33 N 6 Egg	2010_N	Glass	<0.03	<0.2	<0.5	<0.07	<0.03	<2	<3	<0.03	<0.1	<0.1	<1
2346	To11:33 L 6 Egg	2010_L	Glass	<0.03	<0.2	1.05	<0.07	<0.03	<2	<3	<0.03	0.23	<0.1	<1
2309	To11:13 Slurry 11 Sugar, sweets	1999	*plastic*	<0.07	2.92	2.79	3.02	<0.06	<4	8.58	<0.06	0.71	<0.2	<2
2325	To11:28 Slurry 11 Sugar, sweets	2005	Glass	<0.07	<0.5	1.53	3.21	<0.06	<4	<7	<0.06	0.23	<0.2	<2
2339	To11:33 N 11 Sugar, sweets	2010_N	Glass	<0.07	3.45	8.32	0.90	<0.06	<4	13.41	<0.06	<0.2	<0.2	<2
2351	To11:33 L 11 Sugar, sweets	2010_L	Glass	<0.07	4.02	2.96	0.49	<0.06	<4	<7	<0.06	1.64	<0.2	3.03
2300	To11:13 Slurry 3 Meat	1999	Glass	0.05	0.54	<0.5	<0.07	0.19	<2	<3	0.26	0.66	0.14	<1
2317	To11:28 Slurry 3 Meat	2005	Glass	0.06	0.71	<0.5	<0.07	<0.03	<2	4.86	0.28	0.36	<0.1	<1
2329	To11:33 N 3 Meat	2010_N	Glass	<0.03	0.33	<0.5	0.44	<0.03	<2	<3	<0.03	0.25	<0.1	1.22
2330	To11:33 N 3-B Meat (1	2010_N	Glass	<0.03	0.41	<0.5	<0.07	<0.03	<2	4.29	<0.03	0.25	0.10	1.66
2343	To11:33 L 3 Meat	2010_L	Glass	<0.03	<0.2	<0.5	0.17	<0.03	<2	<3	<0.03	0.43	<0.1	<1
2305	To11:13 Slurry 7 Fats	1999	Glass	<0.3	2.30	8.23	50.77	<0.3	<20	31.53	<0.3	1.36	1.15	1.24
2321	To11:28 Slurry 7 Fats	2005	Glass	<0.3	2.20	7.50	91.74	<0.3	<20	345.32	<0.3	1.41	<1	2.10
2334	To11:33 N 7 Fats	2010_N	Glass	<0.3	<2	17.54	0.83	<0.3	<20	51.11	<0.3	<1	<1	<1
2347	To11:33 L 7 Fats	2010_L	Glass	<0.3	<2	25.56	0.95	<0.3	<20	269.15	<0.3	<1	<1	4.86
2301	To11:13 Slurry 4 Fish	1999	*plastic*	0.05	0.45	<0.5	9.58	0.15	<2	<3	<0.03	0.38	<0.1	5.38
2302	To11:13 Slurry 4-B Fish (1	1999	*plastic*	0.13	0.31	<0.5	1.44	0.21	<2	3.82	<0.03	0.26	0.32	3.52
2318	To11:28 Slurry 4 Fish	2005	Glass	0.25	0.29	0.64	22.96	0.24	<2	<3	0.07	0.15	0.12	7.76
2331	To11:33 N 4 Fish	2010_N	Glass	0.77	<0.2	1.49	67.08	<0.03	<2	<3	<0.03	0.20	<0.1	5.00
2344	To11:33 L 4 Fish	2010_L	Glass	0.29	<0.2	0.91	61.21	0.10	<2	4.03	0.20	<0.1	0.12	1.98
2308	To11:13 Slurry 10 Potatoes	1999	*plastic*	<0.03	<0.2	<0.5	<0.07	0.06	<2	<3	<0.03	0.31	<0.1	<1
2324	To11:28 Slurry 10 Potatoes	2005	Glass	0.47	<0.2	1.12	<0.07	<0.03	<2	<3	<0.03	<0.1	<0.1	<1
2338	To11:33 N 10 Potatoes	2010_N	Glass	<0.03	<0.2	0.88	<0.07	<0.03	<2	<3	<0.03	0.20	<0.1	<1
2350	To11:33 L 10 Potatoes	2010_L	Glass	0.07	<0.2	<0.5	<0.07	1.03	<2	<3	0.61	<0.1	<0.1	<1
2306	To11:13 Slurry 8 Vegetables	1999	*plastic*	0.04	0.35	<0.5	0.51	<0.03	<2	7.08	<0.03	0.44	<0.1	11.97
2322	To11:28 Slurry 8 Vegetables	2005	Glass	0.38	<0.2	<0.5	<0.07	<0.03	<2	<3	<0.03	0.41	<0.1	3.64
2335	To11:33 N 8 Vegetables	2010_N	Glass	<0.03	<0.2	<0.5	<0.07	0.10	<2	<3	0.09	0.13	<0.1	3.69
2336	To11:33 N 8-B Vegetables (1	2010_N	Glass	<0.03	<0.2	<0.5	<0.07	<0.03	<2	<3	<0.03	0.21	<0.1	4.34
2348	To11:33 L 8 Vegetables	2010_L	Glass	<0.03	<0.2	0.68	<0.07	<0.03	<2	<3	<0.03	0.39	<0.1	1.63
2307	To11:13 Slurry 9 Fruit	1999	*plastic*	<0.03	<0.2	4.72	0.18	<0.03	<2	<3	<0.03	0.22	<0.1	<1
2323	To11:28 Slurry 9 Fruit	2005	Glass	<0.03	<0.2	0.69	0.66	<0.03	<2	7.31	0.04	0.21	<0.1	<1
2337	To11:33 N 9 Fruit	2010_N	Glass	<0.03	<0.2	0.51	0.59	<0.03	<2	<3	0.05	0.19	<0.1	<1
2349	To11:33 L 9 Fruit	2010_L	Glass	<0.03	<0.2	<0.5	0.76	<0.03	<2	<3	<0.03	0.20	<0.1	<1
2310	To11:13 Slurry 12 Beverages	1999	*plastic*	<0.007	0.16	<0.1	0.49	<0.006	<0.3	2.00	0.01	0.13	<0.02	0.29
2326	To11:28 Slurry 12 Beverages	2005	Glass	<0.007	0.17	<0.1	5.63	<0.006	<0.3	<0.7	0.01	0.13	<0.02	<0.2
2340	To11:33 N 12 Beverages (2	2010_N	Glass	<0.007	0.09	<0.1	2.14	<0.006	<0.3	<0.7	<0.006	<0.02	<0.02	<0.2

1: double sample; 2: only normal price assortment; 3: N=normal price, L=low price

Table 3. Levels of phenolic compounds in 2010 market basket food groups, purchased on the Swedish market (ng/g fresh wt., mean of 2-3 analyses).

Food group		**4-BrP**	**4-t-BP**	**BHT**	**BHA**	**2,4-DBP**	**4-t-OP**	**4-NP**	**TBP**	**PCP**	**TC**	**BPA**
cereals	LB				0	0	0				0	0
	MB	0.22	1.2	1.2	0.035	0.015	1	24	0.10	0.86	0.05	0.50
	UB				0.070	0.030	2				0.10	1.0
pastries	LB	0		0.70		0.017	0	5.5			0	
	MB	0.015	3.7	0.82	12.3	0.024	1	6.2	0,045	0.40	0.05	7.4
	UB	0.030		0.95		0.032	2	7.0			0.10	
dairy pr.	LB	0	0	0.75	0	0	0	0	0		0	0
	MB	0.015	0.10	0.88	0.035	0.015	1	1.5	0.015	0.22	0.05	0.50
	UB	0.030	0.20	1.0	0.070	0.030	2	3.0	0.030		0.10	1.0
egg	LB	0	0	0.50	0	0	0	0	0	0.12	0	0
	MB	0.015	0.10	0.62	0.035	0.015	1	1.5	0.015	0.14	0.05	0.50
	UB	0.030	0.20	0.75	0.070	0.030	2	3.0	0.030	0.16	0.10	1.0
sugar etc	LB	0				0	0	6.5	0	0.80	0	1.5
	MB	0.035	3.7	5.6	0.70	0.030	2	8.2	0.030	0.85	0.10	2.0
	UB	0.070				0.060	4	10	0.060	0.90	0.20	2.5
meats	LB	0	0.23	0	0.20	0	0	1.4	0		0.03	0.97
	MB	0.015	0.26	0.25	0.22	0.015	1	2.4	0.015	0.31	0.06	1.1
	UB	0.030	0.30	0.50	0.23	0.030	2	3.4	0.030		0.10	1.3
fats	LB	0	0			0	0		0	0	0	2.4
	MB	0.15	1	22	0.89	0.015	10	160	0.015	0.50	0.50	2.7
	UB	0.30	2			0.030	20		0.030	1.0	1.0	3.0
fish	LB		0			0.050	0	2.0	0.10	0.10	0.06	
	MB	0.54	0.1	1,2	64	0.055	1	2.8	0.11	0.12	0.08	3.5
	UB		0.2			0.060	2	3.5	0.12	0.15	0.11	
potatoes	LB	0.035	0	0.44	0	0.050	0	0	0.30	0.10	0	0
	MB	0.042	0.1	0.56	0.035	0.055	1	1.5	0.31	0.12	0.050	0.50
	UB	0.050	0.2	0.69	0.070	0.060	2	3.0	0.32	0.15	0.10	1.0
vegetabl.	LB	0	0	0	0	0.050	0	0	0.045		0	
	MB	0.015	0.1	0.25	0.035	0.055	1	1.5	0.052	0.17	0.050	4.0
	UB	0.030	0.2	0.50	0.070	0.060	2	3.0	0.060		0.10	
fruit	LB	0	0	0.26		0	0	0	0.025		0	0
	MB	0.015	0.1	0.37	0.68	0.015	1	1.5	0.032	0.20	0.050	0.50
	UB	0.030	0.2	0.50		0.030	2	3.0	0.040		0.10	1.0
beverages	LB	0		0		0	0	0	0	0	0	0
	MB	0.0035	0,09	0.05	2,1	0.003	0.15	0.35	0.003	0.010	0.010	0.10
	UB	0.007		0.10		0.006	0.30	0.70	0.006	0.020	0.020	0.20
n		25	25	25	25	25	25	25	25	25	25	25
LOD<n<LOQ		4	4	9	3	1	0	3	5	10	2	4
n<LOD		20	15	11	12	21	25	17	17	7	23	15

LB: <LOD=0; MB: <LOD=½LOD; UB: <LOD=LOD. If only MB value, all analyses above LOD (mean of 2-3 analytical values)

Table 4. Estimated per capita intake of phenolic compounds in different food groups and totally, based on 2010 Market Basket data, and MB levels in food (cf. Table 3). Values in ng/person/day (except *, using a mean body weight of 67.2 kg, from 2010 Market Basket study).

	4-BrP	**4-t-BP**	**BHT**	**BHA**	**2,4-DBP**	**4-t-OP**	**4-NP**	**TBP**	**PCP**	**TC**	**BPA**
cereals	50.9	277.5	277.5	8.1	3.5	231.2	5549.6	23.1	198.9	11.6	115.6
pastries	0.8	187.5	41.6	623.4	1.2	50.7	314.2	2.3	20.3	2.5	375.1
dairy pr.	6.4	42.7	375.4	14.9	6.4	426.6	639.9	6.4	93.8	21.3	213.3
eggs	0.3	2.3	14.3	0.8	0.3	23.0	34.5	0.3	3.2	1.2	11.5
sugar etc	4.3	459.2	695.0	86.9	3.7	248.2	1017.7	3.7	105.5	12.4	248.2
meat	3.1	54.1	52.0	45.7	3.1	207.9	499.1	3.1	64.5	12.5	228.7
fats	6.0	39.7	874.0	35.4	0.6	397.3	6356.2	0.6	19.9	19.9	107.3
fish	27.4	5.1	60.8	3243.8	2.8	50.7	141.9	5.6	6.1	4.1	177.4
potatoes	5.3	12.5	70.3	4.4	6.9	125.5	188.2	38.9	15.1	6.3	62.7
vegetabl.	2.9	19.3	48.2	6.8	10.6	192.9	289.3	10.0	32.8	9.6	771.5
fruits	3.6	23.8	87.9	161.5	3.6	237.5	356.3	7.6	47.5	11.9	118.8
beverages	1.2	29.7	16.5	693.3	1.0	49.5	115.5	1.0	3.3	3.3	33.0
TOTAL	112.0	1153.3	2613.4	4925.0	43.7	2241.0	15502.5	102.7	610.8	116.5	2463.1
TOTAL (ng/kg b.w./day)*	1,67	17,2	38,9	73,3	0,651	33,3	231	1,53	9,09	1,73	36,7

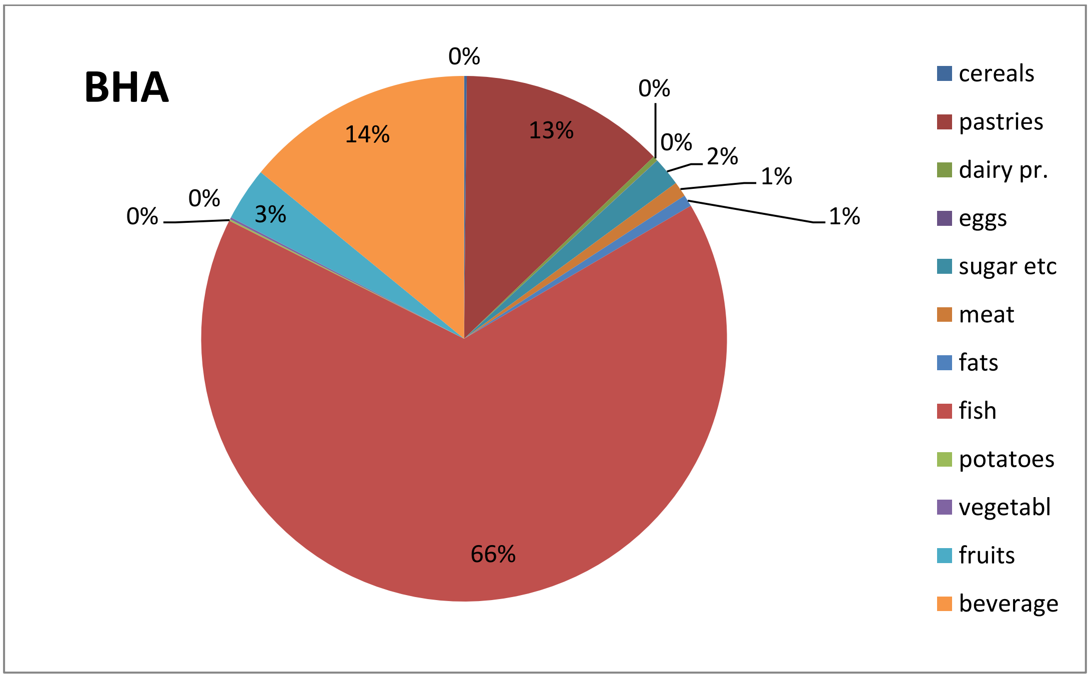
Table 5. The yearly per capita consumption (in kg fresh wt.) according to the 1999, 2005 and 1010 Swedish Market Baskets surveys, presented as 12 food groups that were used as basis for analyses.

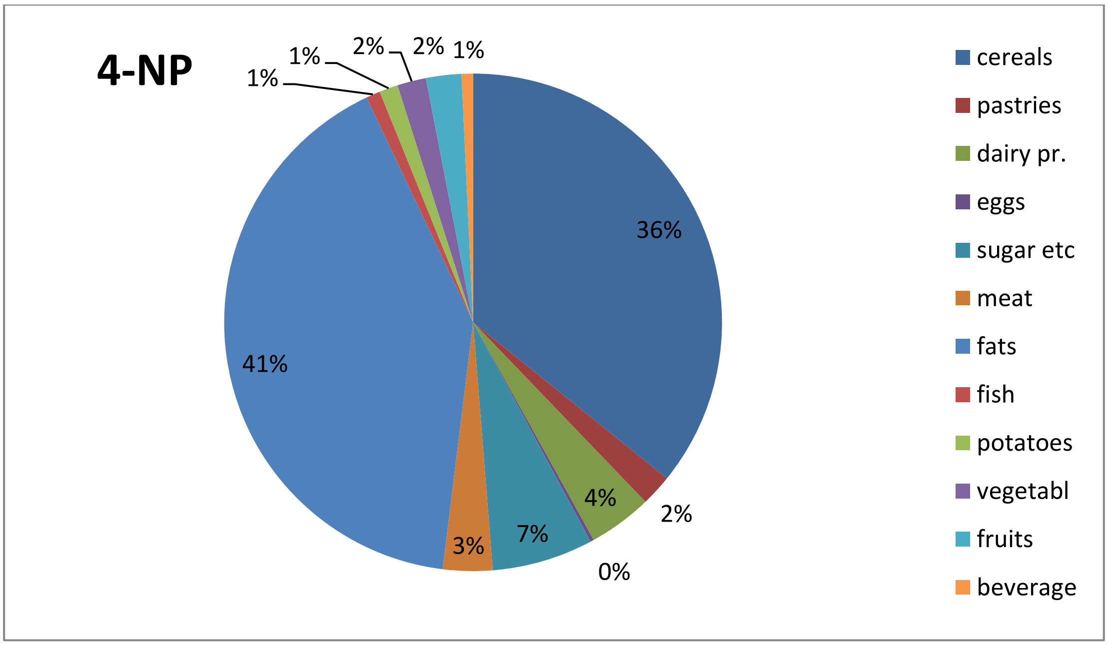
	consumption (kg/person/yr)		
food group	***1999***	***2005***	***2010***
cereals	69,4	91,1	84,4
pastries	13,7	19,1	18,5
dairy pr.	168,5	175,8	155,7
eggs	9,2	8,1	8,4
sugar etc	35,4	37,8	45,3
meat	56,7	74,4	75,9
fats	17,5	14	14,5
fish	13,3	16,7	18,5
potatoes	51,4	44,3	45,8
vegetabl	54,8	63,6	70,4
fruits	64,1	68,3	86,7
beverage	118,8	126,7	120,5

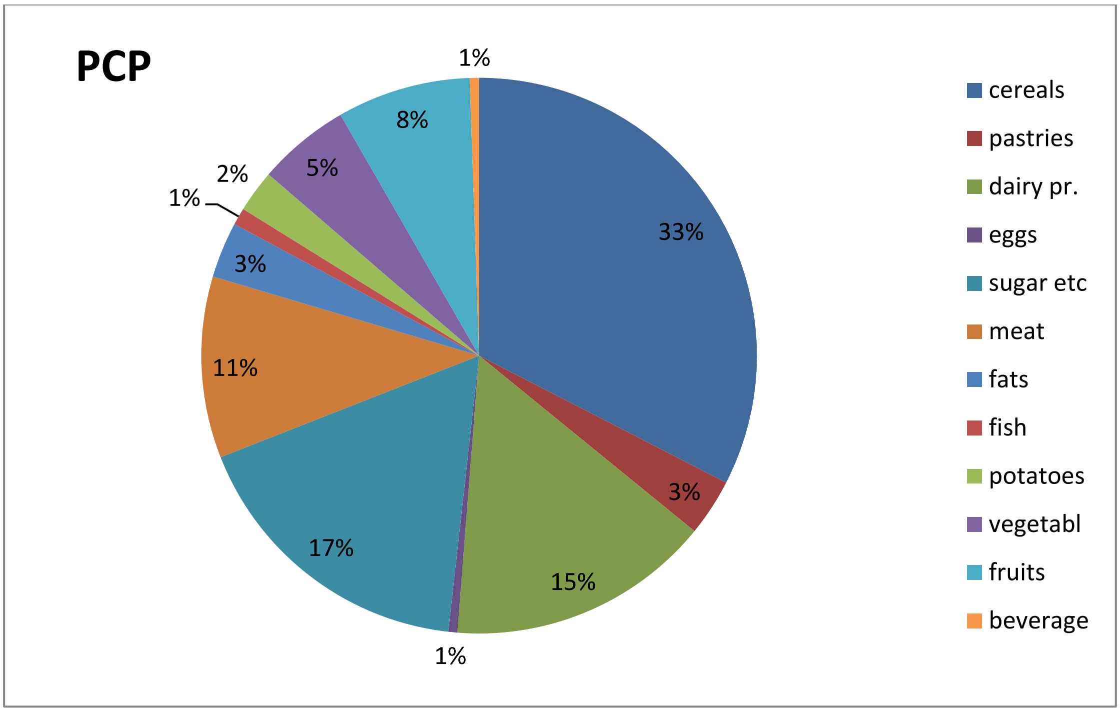
Table 6. Total intake of phenolic compounds from food, based on per capita consumption as registered in 1999, 2005 and 2010 Market Basket studies. Intakes are expressed as ng/person/day, based on MB levels in analysed food group homogenates.

	4-BrP	4-t-BP	BHT	BHA	2,4-DBP	4-t-OP	4-NP	TBP	PCP	TC	BPA
1999	49,7	1060	3398	3557	71,8	2133	7086	83,7	744	196	2887
2005	189	1099	1305	7617	44,1	2233	22025	118	1776	136	2514
2010	112	1153	2613	4925	43,7	2241	15502	103	611	116	2463

Figure 1. Relative contribution (%) from different food groups to the total intake of BHA, 4-NP, PCP and BPA, based on estimated per capita intake values from 2010 market basket samples (cf. Table 4).






PCP
1%
8%
5%
2%
1%
3%
33%
11%
3%
17%
15%
1%
cereals
pastries
dairy pr.
eggs
sugar etc
meat
fats
fish
potatoes
vegetabl
fruits
beverage

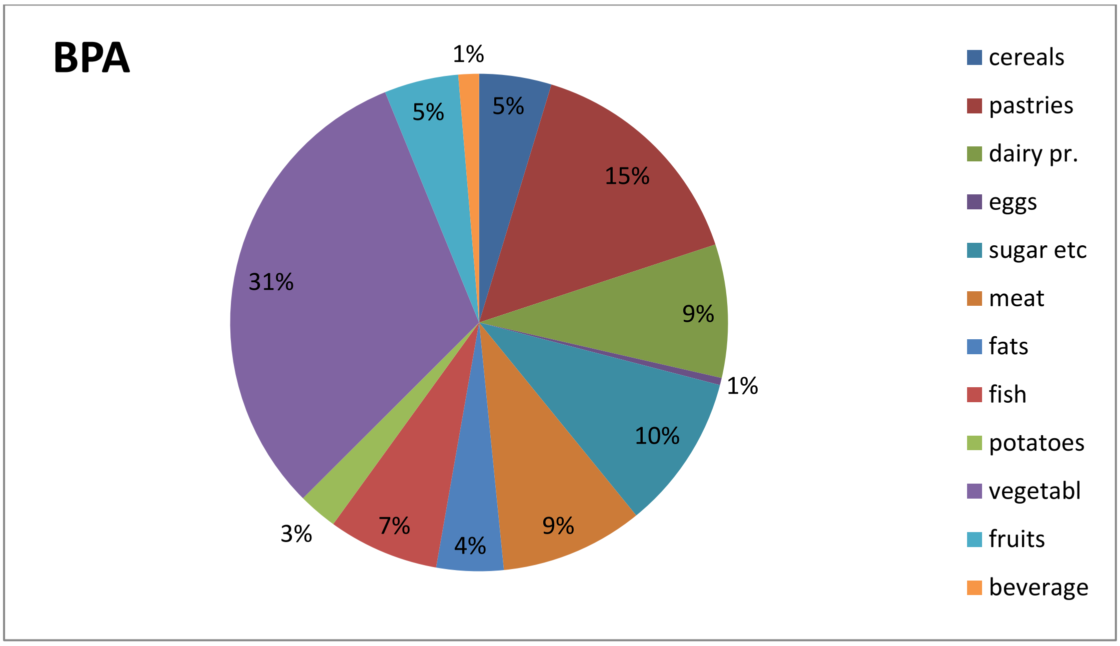

BPA
1%
5%
5%
15%
31%
9%
1%
10%
3%
7%
4%
9%
cereals
pastries
dairy pr.
eggs
sugar etc
meat
fats
fish
potatoes
vegetabl
fruits
beverage

Figure 2. Changes in estimated intake over time, in studied phenols with ≥40% of analysed food levels above LOD (c.f. Table 3). Intakes based on Market Basket samples and per capita consumption data (y-axis: ng/person/day; MB levels) from the years 1999, 2005 and 2010.