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Polycyclic aromatic hydrocarbons (PAHs) in meat products and estimated PAH intake by children and the general population in Estonia

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Abstract

The concentrations of benzo(a)pyrene and 11 other polycyclic aromatic hydrocarbons (PAHs) were analysed from 322 commercial cured meat products and 14 home-grilled meat samples as part of the Estonian food safety monitoring programme during 2001-2005. The maximum acceptable concentration of 5 µg kg⁻¹ for benzo(a)pyrene was exceeded in 3.4% of samples. The highest PAH concentrations were detected in home-grilled pork samples. Using of disposable grilling unit resulted in 1.6 times higher PAH concentrations compared to the traditional wood-burning grill. The average intake of benzo(a)pyrene and sum of 12 PAHs from meat products was estimated for children (age 1-16 years) on the basis of individual food consumption questionnaire and for the general population based on the national food consumption

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3 data. The highest total PAH concentrations detected were $16 \mu\text{g kg}^{-1}$ in smoked meat
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5 and ham, $19 \mu\text{g kg}^{-1}$ in smoked sausage and $6.5 \mu\text{g kg}^{-1}$ in smoked chicken samples.
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8 Since smoking and grilling are prevalent meat cooking methods in Estonia, the impact
9
10 of meat products is assessed to be significant in overall PAH intake.
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15 **Keywords:** polycyclic aromatic hydrocarbons, benzo(a)pyrene, meat products, intake
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Introduction

Polycyclic aromatic hydrocarbons (PAHs) include several potent carcinogenic compounds consisting of two or more fused aromatic rings. PAHs are formed during incomplete combustion processes or in high-temperature pyrolysis of coal, oil and other organic materials. The most important sources of PAHs have been identified as coke ovens in the production of aluminium, iron, and steel; heating in power plants and residences; cooking; motor vehicle traffic; environmental tobacco smoke; and the incineration of waste material (WHO 1998). Over 100 PAHs have been identified in the environment as pollutants occurring as complex mixtures (Mottier et al. 2000).

Many of PAHs have been demonstrated to be carcinogenic for experimental animals and they are also supposed to have a significant impact on the disease burden caused by various cancers in human population (Phillips 1999). PAHs highlighted to be genotoxic and carcinogenic by the Scientific Committee on Food (SCF) are the following: benz(a)anthracene, benzo(b)fluoranthene, benzo(j)fluoranthene, benzo(k)fluoranthene, benzo(g,h,i)perylene, benzo(a)pyrene, chrysene, cyclopenta(c,d)pyrene, dibenz(a,h)anthracene, dibenzo(a,e)pyrene, dibenzo(a,h)pyrene, dibenzo(a,i)pyrene, dibenzo(a,l)pyrene, indeno(1,2,3-cd)pyrene, 5-methylchrysene (European Commission 2005a). Joint Expert Committee on Food Additives (JECFA) has added 16th analyte to the list – 7H-benzo-(c)fluorene (JECFA 2005). European Commission has required further investigation of the levels of benzo(a)pyrene and other PAHs, in particular those highlighted to be carcinogenic by SCF, in food (European Commission 2005a, Wenzl et al. 2006). The International Agency of Research on Cancer (IARC) has categorized 15 PAHs, including benzo(a)pyrene, benz(a)anthracene, benzo(b)fluoranthene, benzo(j)fluoranthene,

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3 benzo(k)fluoranthene, dibenz(a,h)anthracene, dibenzo(a,e)pyrene, dibenzo(a,i)pyrene,
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5 dibenzo(a,l)pyrene dibenzo(a,l)pyrene, indeno(1,2,3-cd)pyrene and 5-methylchrysene,
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8 as reasonably anticipated to be human carcinogens (IARC 1973, 1983, 1987). In
9
10 mammalian cells PAHs undergo metabolic activation to diol epoxides that bind
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12 covalently to cellular macromolecules, including DNA, thereby causing errors in
13
14 DNA replication and mutations that initiate the carcinogenic process (Janoszka et al.
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16 2004).
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22 Diet is the primary source of human exposure to PAHs contributing to more than 90%
23
24 of total exposure to PAHs (WHO 1998, SCF 2002). PAHs may be present as
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26 contaminants in various food items and beverages including drinking water (Chen
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28 2004, King et al. 2004), vegetables, fruits, cereals, oils (Dennis et al.1991, Moret and
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30 Conte 2000, Moret and Conte 2002, Rojo Camargo and Toledo 2003, Tao et al. 2006,
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32 Voutsas and Samara 1998), fish (Šimko 1991, Lodovici et al. 1995, Moret and Conte
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34 2000, Yurchenko and Mölder 2005, Karl and Leinemann 1996, Stołyhwo and
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36 Sikorski 2005) and meat (Kazerouni et al. 2001, Elhassaneen 2004, Šimko 2002,
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38 Mottier 2000, Janoszka 2004). The possible sources of PAHs in food are
39
40 environmental contamination from the atmosphere, soil or water, contamination from
41
42 packaging materials and thermal treatment which is used for the preparation and
43
44 manufacturing of food (Guillen 1994). PAHs are lipophilic and have very low
45
46 aqueous solubility so they accumulate in lipid tissue of plants and animals. The levels
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48 of PAHs found in unprocessed foods reflect the background contamination which
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50 originates from long distance airborne transportation of contaminated particles and
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52 natural emissions.
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3 Food processing procedures, such as smoking, drying and cooking, are generally
4 supposed to be the major sources of PAH contamination of food. Formation of PAHs
5 in meat products is affected by several factors: the methods used for preparation of
6 food (grilling, frying, roasting etc.), temperature and time of cooking, distance from
7 the heat source and drainage of fat (SCF 2002, Kazerouni et al. 2001). Several
8 mechanisms of formation of PAHs have been proposed, such as melted fat that
9 undergoes pyrolysis when dripping onto the heat source and pyrolysis of the meat due
10 to the high temperature (Lijinsky 1991). Because PAH formation occurs on or near
11 the surface of meats, food cooked without being exposed to smoke does not show
12 significant levels of PAH. The PAH concentrations in smoked food depend on several
13 variables in the smoking process, including the type of smoke generator and wood,
14 combustion temperature and degree of smoking (Moret et al. 1997). Lowering the
15 temperature of smoke formation to 300-400 °C combined with the use of filters, the
16 PAH content of smoke can be decreased by about 90% (Sikorski 2005).

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39 Benzo(a)pyrene (BaP) is the most widely known and studied of the PAHs due to its
40 importance as one of the most potent animal carcinogenic PAH, its relatively easy
41 analysis and presence in wide variety of food items. BaP seems to be a good marker
42 for other PAHs in food items, the correlation coefficient between the total PAH and
43 the BaP concentration has been detected to be 0.87 and between the carcinogenic
44 PAHs and BaP 0.98 (Kazerouni et al. 2001). Maximum acceptable concentration of 5
45 $\mu\text{g kg}^{-1}$ has been established for BaP in smoked meats and smoked meat products
46 (European Commission 2005b).
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3 Several methods for analysis have been used for the determination of PAHs in food.
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5 PAHs have been extracted from homogenized food samples using various techniques
6
7 such as liquid-liquid extraction (Rojo Camargo and Toledo 2003) or supercritical
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9 fluid extraction (Järvenpää et al. 1996, Lage Yusty and Daviña 2005). Solid phase
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11 extraction (Nazarkina et al. 2001) and gel permeation chromatography (Yurchenko
12
13 and Mölder 2005) have been used for sample cleanup. The determination of PAHs in
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15 food has been carried out by various analytical methods, including thin layer
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17 chromatography (Kazerouni et al. 2001, Moret and Conte 2000), high performance
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19 liquid chromatography (Chen et al. 1996, Chiu et al. 1997, Rojo Camargo and Toledo
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21 2003, de Boer and Law 2003), gas chromatography with mass-selective detector
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23 (Mottier et al. 2000, Yurchenko and Mölder 2005, Jira 2004) or flame ionization
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25 detector (Moret and Conte 2000).
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34 Intake of PAHs has been estimated in some European countries: in Austria, Italy, The
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36 Netherlands and UK, furthermore the estimated BaP intake is available for Germany,
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38 Sweden and USA (SCF 2002). According to several studies the major dietary
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40 contributors are cereals, oils and vegetables although PAH levels in cereals are often
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42 low (Dennis et al. 1983, de Vos et al. 1990, Tao et al. 2006). Grilled or barbequed
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44 meat caused a significant proportion (21%) of the average total daily BaP intake in
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46 USA according to the data obtained by Kazerouni et al. (2001).
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53 Various food items and products in Estonia have been analysed for BaP content
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55 starting from year 1999 and for PAH content from 2003 in the framework of the
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57 Estonian food safety monitoring programme, initiated in 1998 (Reinik et al. 2001).
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59 The aim of the present study was to determine the levels of PAHs in commercial meat
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3 products and grilled meat prepared by traditional Estonian habits. Furthermore, the
4
5 aim here was to estimate the human exposure to PAHs and BaP through various meat
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7 products. As grilling, smoking and barbequing of meat are widely used in Estonia, the
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9 assessment of dietary risks connected with the intake of PAHs is essential.
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12 13 14 15 **Materials and methods**

16 17 *Meat samples*

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19 Altogether 322 samples from 34 Estonian meat manufacturers were analyzed in
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21 2001 – 2005 for the BaP content. Samples, from which 94% constituted smoked and
22
23 grilled meat products, were collected from the main retail outlets of Tartu, the second
24
25 largest town in Estonia. Samples were divided into 9 product groups (Table II). In
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27 addition to BaP, concentrations of 11 PAHs, included in the European Commission
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29 list (European Commission 2005a), were analysed from 22 industrial and 14 home-
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31 grilled meat products in 2005.
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38 Various types of grilling equipment are used for home-grilling. In present study two
39
40 commonly used grilling methods were used: traditional wood burning grill and small
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42 disposable charcoal grill unit. In the traditional wood-burning grill the distance
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44 between burning coals and meat is approximately 20 cm and the temperature at the
45
46 surface of the meat is between 180-240 °C. In recent years disposable charcoal grills
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48 have gained popularity for their convenience and shorter preparation time. Compared
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50 to traditional grilling the heat source is much closer to the grilled product in these
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52 disposable grills. 14 samples of meat products were grilled using both methods and
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54 analyzed for the content of 12 PAHs.
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Analysis of PAHs

Analyses of the PAHs in meat samples were carried out at Tartu Laboratory of Estonian Health Protection Inspectorate. The gas chromatographic method with mass-selective detector (GC/MS) was used to determine the following PAHs: benz(a)anthracene, sum of benzo(b)-, benzo(j)- and benzo(k)fluoranthene, benzo(g,h,i)perylene, benzo(a)pyrene, chrysene, dibenz(a,h)anthracene, dibenzo(a,e)pyrene, dibenzo(a,l)pyrene, indeno(1,2,3-cd)pyrene and 5-methylchrysene. High performance liquid chromatography (HPLC) with fluorescence detection was used for samples in which only benzo(a)pyrene content was measured.

For sample preparation the method described by Ojaveer and Tanner (1996) with some modifications was used. The sample was homogenized and hydrolyzed in concentrated hydrochloric acid overnight. Dichloromethane/hexane solution 25:75 was added. The solution was filtered through the layer of Na₂SO₄ and NaHCO₃ 1:1. The eluate was evaporated to dryness, ethyl acetate/cyclohexane 50:50 solution was added. Portion of the extract was injected into a gel chromatograph. The fraction containing PAHs was collected and evaporated to dryness. The residue was dissolved in methanol.

For benzo(a)pyrene analysis an Hewlett Packard 1100 HPLC equipment with Agilent 1100 fluorescence detector was used. The limit of determination was 0,3 µg kg⁻¹ and recovery was 80 %. A Hewlett Packard Model 6890 gas chromatograph equipped with mass selective detector and chromatographic column HP-5 (30m x 0.25 mm i.d. silica capillary column with film thickness of 0.25 µm) was used for the analysis of PAHs. The temperature was programmed from 80 to 300 °C. Positive-ion chemical

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3 ionization mass spectrometry was performed with ammonia reagent gas. The method
4 performance parameters are given in Table I.
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10 Both methods are accredited by Estonian Accreditation Centre. This laboratory has
11 successfully participated in international proficiency tests using both methods of
12 analysis. A reference material, smoked fish paste containing 16 PAHs (prepared by
13 Estonian Institute of Chemical Physics and Biophysics), was analysed in each series
14 of analyses, the results for HPLC and GC/MS analysis of benzo(a)pyrene are
15 coincident in accepted range.
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27 [Insert Table I about here]
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31 *Method for intake estimation*

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33 To obtain data about the consumption of cured meat products by children in Estonia a
34 questionnaire developed by the scientists of the Food Hygiene and Control
35 Department of Estonian University of Life Sciences was used. Questionnaires were
36 distributed to schools, kindergartens and family doctor centres and asked to fill in by
37 the children's parents. The main aim was to estimate the daily consumption of meat
38 products by children. Data on the number and age of children in the family,
39 consumption of meat products per child, preference for domestic and imported
40 products and attitude towards food additives were included in the questionnaire. The
41 data from 346 children were received in 2000 – 2003. For the statistical analysis of
42 data the responses were grouped in five groups by age of the children. According to
43 registered data the maximum and average amounts of consumed meat products in
44 grams in various age groups were calculated.
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6 Food consumption data from Estonian Board of Statistics were used for the estimation
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8 of average intake of PAHs and BaP in the general population in Estonia. Furthermore,
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10 an estimation was made for PAH and BaP intake through theoretical consumption of
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12 100 g of each meat product per day. The national food consumption data were
13
14 collected in 2001 – 2004.
15

16 17 18 19 20 **Results and discussion**

21 22 *BaP in commercial meat products*

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24 During the years 2001–2005 the range and average concentrations of BaP in 322
25
26 samples from various industrial and commercially available meat products were
27
28 determined (Table II). BaP contents in analyzed meat products were at range of <0.3
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30 (below the determination limit) and 31.2 $\mu\text{g kg}^{-1}$. In 64% of the analyzed samples BaP
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32 concentrations remained below the limit of determination (<0,3 $\mu\text{g kg}^{-1}$). The
33
34 maximum acceptable concentration of 5 $\mu\text{g kg}^{-1}$ for BaP in smoked meat products
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36 (European Commission 2005b) was exceeded in 11 samples. High PAH concentration
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38 containing products were home-made ham, smoked meat, smoked chicken and
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40 smoked sausages with concentrations of 30.6, 31.2, 15.0 and 20.0 $\mu\text{g kg}^{-1}$,
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42 respectively.
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48 [Insert Table II about here]
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51 In one earlier study concentrations of BaP have been found in the range of 0.03-100
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53 $\mu\text{g kg}^{-1}$ in smoked meat products (Šimko 2002). The highest concentrations were
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55 detected in smoked sausages and smoked ham. In cooked meat products in Poland
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57 0.11-3.93 $\mu\text{g kg}^{-1}$ of BaP have been detected (Janoszka et al. 2004).
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3 *PAHs in commercial smoked meat products*
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6 22 samples of meat products were analyzed for the concentrations of 12 PAHs in
7
8 2005 (Table III). Eleven of those, except benzo(g,h,i)perylene, are categorized by
9
10 IARC as probable or possible carcinogens (IARC 1973, 1983, 1987).
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12 [Insert Table III about here]
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15 Benzo(a)anthracene was detected in 15 samples, BaP in 13 samples.
16
17 Benzo(a)anthracene and chrysene were found in highest concentrations. 5-
18 methylchrysene, dibenzo(a,h)anthracene, dibenzo(a,l)pyrene, dibenzo(a,e)pyrene
19 concentrations remained below the determination limit in all samples. PAHs were not
20 found in 4 samples. There are several factors influencing the final PAH content in the
21 products in smoking and grilling process: type of wood and smoke generator, smoke
22 temperature, duration of smoking or grilling, availability of oxygen and oven
23 dimensions. As most of the cooked sausages undergo only a slight smoking process,
24 BaP and PAH concentrations close to or under the determination limits were
25 observed.
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41 Our results are consistent with the study by Phillips (1999) where the concentration
42 range of total PAHs in smoked meat was between 2.6 and 29.8 $\mu\text{g kg}^{-1}$, and highest
43 concentration of carcinogenic PAHs was 16.0 $\mu\text{g kg}^{-1}$. Reports of previous works
44 (Šimko, 2002) indicate that BaP concentrations are between 1% and 20% of the total
45 carcinogenic PAHs. According to our results BaP comprises in average 6.1 % of the
46 total sum of PAHs in commercial smoked meat products.
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4 *PAHs in home-made grilled meat products*

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6 The contents of PAHs in grilled products are shown in Table IV. Using of disposable
7
8 grill resulted in higher (maximum 1.6 times) concentrations of PAHs compared to
9
10 traditional grilling.

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12 [Insert Table IV about here]

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15 Total amounts of 12 PAHs was higher in grilled pork than in grilled chicken or
16
17 sausages. The difference occurs probably due to variation in fat content. When fatty
18
19 meat is grilled, more fat drops onto the coals and due to the high temperature, PAHs
20
21 are formed. In consequence, smoke carries the PAHs onto the surface of meat
22
23 products. Lower concentrations of PAHs in sausages can be explained by shorter
24
25 grilling time.
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32 Benzo(a)pyrene, benzo(a)anthracene, sum of benzo(b),(k),(j)fluorenes, chrysene and
33
34 benzo(g,h,i)perylene were detected in all samples. The highest concentrations were
35
36 determined for chrysene, sum of benzo(b),(k),(j)fluorene and benzo(a)anthracene,
37
38 maximum values being 6.2, 5.1 and 3.0 $\mu\text{g kg}^{-1}$, respectively. The results in table IV
39
40 indicate a certain relation between the amount of benzo(a)pyrene and total amount of
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42 PAHs. According to our results BaP comprises in general 6.3 % of the total sum of
43
44 the PAHs in home-grilled meat products.
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51 Home-prepared meat products, especially those which were prepared using a
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53 disposable grill, contained higher concentrations of BaP and PAHs compared to
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55 commercial products. The mean concentration of BaP in industrial and disposable-
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57 grill meat was 0.7 $\mu\text{g kg}^{-1}$ and 1.0 $\mu\text{g kg}^{-1}$, respectively. The average levels of PAHs
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3 in home-grilled meat were higher compared to industrial smoked products, mean
4 contents being correspondingly $12.5 \mu\text{g kg}^{-1}$ and $7.5 \mu\text{g kg}^{-1}$.
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10 Elhassaneen (2004) determined 11 PAHs in charcoal-broiled beefburgers. The range
11 of PAHs was between $0.31\text{-}14.95 \mu\text{g kg}^{-1}$ and most frequently pyrene and
12 benzo(a)pyrene were detected. The range of BaP concentration was $0.99\text{-}4.8 \mu\text{g kg}^{-1}$.
13
14 Contents of 16 PAHs in seven different barbequed meat sausages were determined in
15 a Swiss study. The highest concentrations were found for phenanthrene and
16 naphthalene, BaP contents were between “not detected” and $2.81 \mu\text{g kg}^{-1}$ (Mottier et al.
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The highest concentrations of PAHs reported in meat have been detected in food
cooked over open flames. For example, in barbecued meat, the total PAHs were found
to be present at levels up to $164 \mu\text{g kg}^{-1}$; with BaP being present at levels as high as
 $30 \mu\text{g kg}^{-1}$ (Panalaks 1976). The results for the total sum of PAHs are difficult to
compare between the studies published earlier by other authors as there is variation in
analyzed compounds and several non-carcinogenic PAHs have been included in
earlier works.

Intake of BaP and PAHs by children from the consumption of meat products

The questionnaire concerning the daily consumption of meat products children's
parents were asked to complete during one observation week. Data from 346 children
at the age of 1 to 16 years were obtained (Reinik et al. 2005). In the PAH and BaP
intake estimation an individual approach was applied: using the mean concentrations
of BaP (Table II) and PAHs detected in meat products (Table III and IV) and taking

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2
3 the individual data for consumption of cooked sausages, smoked sausages, ham and
4
5 grilled meat products by each respondent child. The intake of BaP by children, for
6
7 the period of 2001-2005 is represented in ng per day in Table V.
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12 [Insert Tables V-VI about here]
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17 The mean BaP intake by children was found to be 14 ng per day, maximum reaching
18
19 to 140 ng per day. Intake per kg of body weight (bw) is higher in younger age groups
20
21 (1-9 years), maximum intake is more than 10-fold compared to the average value of
22
23 0.46 ng per day per kg of bw. The average intake of PAHs from meat products was
24
25 found to be 192 ng per day (Table VI), whereas maximum intake was estimated to be
26
27 up to 1575 ng per day. BaP comprises 7.3% of total PAH intake. Both for BaP and
28
29 PAH the highest intake is obtained from smoked meat products followed by cooked
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31 sausages and grilled products.
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39 Dietary intake of BaP by schoolchildren and toddlers estimated in a UK survey COT
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41 (SCF 2002) was in the range of 1.4-3.8 ng kg⁻¹ bw per day. Similarly to our work the
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43 youngest age groups were found to have the highest exposure for both BaP and PAH
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45 per kg of bw, about 2.4-fold higher intake than for adults (SCF 2002).
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51 *Intake of BaP and total PAHs by Estonian population from the consumption of meat*
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53 *products*
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56 Dietary intake of food contaminants depends on both the nutritional habits of the
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58 examined population group and the concentrations of contaminants in food. Food
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60 consumption data from the Board of Statistics were used in the calculation of average

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3 intake for the general population. Data include the size of the population and average
4
5 amounts of monthly consumed meat products in Estonia in 2001-2004.
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10 The lowest concentrations of BaP were detected in cooked sausages and beef
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12 hamburgers, $0.15 \mu\text{g kg}^{-1}$ as a mean value. However, cooked sausages are the most
13
14 frequently used meat product in general population with the average consumption of
15
16 35 g per day. The mean intake of BaP from consuming cooked sausages was
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18 estimated to be 5.3 ng per day in our study. The highest concentrations of BaP were
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20 determined in smoked and grilled meat products. From before-mentioned product
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22 groups smoked sausages give the highest intake of BaP; 11 ng per day. The average
23
24 consumption of grilled, smoked and other cured meat products is 65 g per day in
25
26 general Estonian population (Table VII) which generates BaP intake of 29 ng per day
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28 ($0.45 \text{ ng kg}^{-1} \text{ bw per day}$). As food consumption database indicates only average
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30 consumption, the real intake values for persons who eat high quantities of meat
31
32 products frequently can be much higher.
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38 [Insert Table VII about here]

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41 According to mean concentrations of PAHs found in present study and taking into
42
43 account average daily consumption and the theoretical consumption of 100 g of meat
44
45 product, average daily PAH intake was estimated. Highest concentrations of PAHs
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47 were detected in grilled meat, concentration of $16 \mu\text{g kg}^{-1}$. Consuming 26 g of smoked
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49 and grilled meat products per day as average in Estonia, causes a 234 ng daily PAH
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51 intake ($3.6 \text{ ng kg}^{-1} \text{ bw per day}$).
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58 As different methods have been used for the estimation of intake of food
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60 contaminants and extensive variation of diets, results obtained in earlier studies differ

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3 to the large extent. In USA and in some European countries the estimated BaP intake
4 from food is reported to be between 0.1-1.6 μg per day (Guillen and Sopelana 2003),
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6 according to surveys conducted in six EU countries the mean or national-averaged
7
8 intake of BaP for an adult person was estimated to be between 0.05 and 0.29 μg per
9
10 day (SCF 2002) and the total intake of 19 PAHs was estimated to be 8.8 μg per day in
11
12 Finland (SCOOP report 2004). Large variation in estimated intake values is probably
13
14 due to the food groups and items considered, differences in analytical methods and to
15
16 statistical analysis of concentration values below the detection and determination
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18 limits. The estimated contribution of meat products to the overall intake of PAHs also
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20 differs between countries from very low for UK to 21 % in USA and 27 % in France
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22 resulting in the second contributing food group after bread and cereals (SCF 2002,
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24 SCOOP report 2004).
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34 As smoking and grilling are prevalent meat preparation methods in Estonia, the
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36 impact of meat products is assumed to be significant in the overall PAH intake. The
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38 mean intake values obtained in this study were lower compared to the estimates by
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40 other countries, however the results showed that for high consumers intake may be
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42 significant and exceed average value up to 10 times.
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50 **Conclusions**

51 The content of benzo(a)pyrene in 322 meat products was determined by an HPLC
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53 method. BaP contents in meat products were in the range of <0.3 and $31.2 \mu\text{g kg}^{-1}$,
54
55 with a mean concentration of $0.8 \mu\text{g kg}^{-1}$. The maximum acceptable concentration of
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57 $5 \mu\text{g kg}^{-1}$ for BaP in smoked meat products was exceeded in 11 samples, mainly
58
59 home-made ham, smoked meat and smoked chicken. Concentrations of 12 PAHs
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3 were determined in 22 commercial smoked and 14 home-grilled meat products. For
4
5 smoked products the highest PAH concentrations were detected to be $19 \mu\text{g kg}^{-1}$,
6
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8 whereas the average concentration was $7.5 \mu\text{g kg}^{-1}$.
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12 The intake of BaP and PAHs by Estonian children from meat products was estimated
13
14 on the basis of the data obtained from the individual food consumption
15
16 questionnaires. The mean BaP intake was found to be 14 ng per day, maximum intake
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18 being more than 10 times higher. The average intake of PAHs by children from meat
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20 products was found to be 192 ng per day, while maximum intake values were
21
22 estimated to be up to 1575 ng per day.
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28 The mean intake by the general population from the consumption of meat products
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30 was found to be 29 ng of BaP per day and 346 ng of total PAH per day. For
31
32 consumption of 100 g of smoked or grilled meat products per day the mean daily
33
34 intake was estimated to be 87 ng for BaP and 942 ng for PAHs. The PAH intake from
35
36 the consumption of meat products per kg of body weight is highest among children in
37
38 a age group of 1-9 years; the mean daily intake exceeding by 1.5 times, and the
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40 maximum by 10-fold the average intake of the whole population ($5.3 \text{ ng kg}^{-1} \text{ bw}$ per
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42 day).
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51 Considering the toxicity of PAHs and high consumption of grilled and smoked meat
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53 products in Estonia, the data reported on the levels of carcinogenic PAHs in food and
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55 information on daily intake by children and the whole population is clearly
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57 significant.
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Table I GC/MS method performance characteristics

PAHs	Retention time (min)	Detection limit ($\mu\text{g kg}^{-1}$)	Determination limit ($\mu\text{g kg}^{-1}$)	Recovery %
Benzo(a)anthracene	12.25	0.2	0.7	101
Σ Benzo(b),(k),(j) fluoranthene	17.09	0.3	0.9	97
Benzo(g,h,i)perylene	25.73	0.2	0.5	87
Benzo(a)pyrene	18.75	0.1	0.3	90
Chrysene	12.40	0.2	0.7	97
Dibenzo(a,h)anthracene	24.83	0.2	0.7	99
Dibenzo(a,e)pyrene	32.57	0.2	0.5	99
Dibenzo(a,l)pyrene	30.74	0.2	0.5	85
Indeno(1,2,3)pyrene	24.57	0.3	1.0	92
5-methylchrysene	14.21	0.2	0.7	98

Table II Benzo(a)pyrene concentrations ($\mu\text{g kg}^{-1}$) in 322 commercial meat products in Estonia 2001-2005.

Product group	No. of samples	Concentration range ($\mu\text{g kg}^{-1}$)	Median concentration ($\mu\text{g kg}^{-1}$)	Average concentration ($\mu\text{g kg}^{-1}$)
Smoked sausage	115	<0.3-20	<0.3	1.1
Ham	116	<0.3-30.6	<0.3	0.8
Smoked meat	20	<0.3-31.2	0.5	0.7
Smoked chicken	29	<0.3-15.0	0.3	1.3
Grilled chicken	6	<0.3-1.0	0.6	0.5
Grilled sausage	10	<0.3-1.2	0.7	0.6
Grilled meat	6	0.7-1.8	1.0	1.1
Cooked sausage	16	<0.3-1.0	<0.3	0.15
Others	4	<0.3	<0.3	
Total no. of samples	322			

Table III PAH concentrations ($\mu\text{g kg}^{-1}$) in 22 commercial smoked meat products in Estonia 2005.

PAH	Ham n=6		Smoked sausage n=13		Smoked chicken n=3	
	range	mean	range	mean	range	mean
Benzo(a)anthracene	<0.7-3.2	1.6	<0.7-5.6	1.6	1.0-2.2	1.7
Σ Benzo(b),(k),(j) fluoranthene	<0.9-3.8	1.3	<0.9-2.4	0.9	<0.9-1.2	0.7
Benzo(g,h,i)perylene	<0.5-0.5	0.3	<0.5-0.7	0.4	<0.5	0.25
Benzo(a)pyrene	<0.3-0.5	0.4	<0.3-1.3	0.4	<0.3-0.8	0.4
Chrysene	<0.7-5.5	2.6	<0.7-7.0	2.0	<0.7-1.3	0.9
Dibenzo(a,h)anthracene	<0.7		<0.7		<0.7	
Dibenzo(a,e)pyrene	<0.5		<0.5		<0.5	
Dibenzo(a,l)pyrene	<0.5		<0.5		<0.5	
Indeno(1,2,3)pyrene	<1.0-1.1	0.6	<1.0-2.4	0.8	<1.0	0.5
5-methylchrysene	<0.7		<0.7		<0.7	
Σ 12 all PAHs		8.0		7.3		5.7

Concentrations below determination limit (LOQ) were counted as LOQ/2 to calculate mean levels of PAHs.

Table IV PAH concentrations ($\mu\text{g kg}^{-1}$) in 14 Estonian home-grilled meat products.

PAHs	Grilled pork				Grilled sausage				Grilled chicken			
	Wood burning grill n=3		Disposable charcoal grill n=3		Wood burning grill n=2		Disposable charcoal grill n=2		Wood burning grill n=2		Disposable charcoal grill n=2	
	range	mean	range	mean	range	mean	range	mean	range	mean	range	mean
Benzo(a)anthracene	1.6-2.5	1.9	3.0	3.0	1.4-2.1	1.8	1.0-1.7	1.4	0.7-1.2	1.0	1.5-1.7	1.6
Σ Benzo(b),(k),(j)-fluoranthene	1.7-3.8	3.0	4.3-5.1	4.6	2.6-2.7	2.6	2.7-3.5	3.1	2.2-2.6	2.4	3.0-3.6	3.3
Benzo(g,h,i)perylene	0.5-1.5	1.1	1.8-2.3	2.1	1.3-1.4	1.4	1.4-1.5	1.4	1.2-1.4	1.3	1.5-1.8	1.7
Benzo(a)pyrene	0.7-1.0	0.8	1.0-1.8	1.4	0.6-0.8	0.7	0.5	0.5	0.3-0.5	0.4	0.8-1.0	0.9
Chrysene	2.4-6.2	3.7	4.7-6.2	5.6	1.5-2.3	1.9	1.5-3.5	2.5	0.9-1.6	1.3	2.2-3.1	2.7
Dibenzo(a,h)-anthracene	<0.7		<0.7		<0.7		<0.7		<0.7		<0.7	
Dibenzo(a,e)pyrene	<0.5		<0.5		<0.5		<0.5		<0.5		<0.5	
Dibenzo(a,l)pyrene	<0.5		<0.5		<0.5		<0.5		<0.5		<0.5	
Indeno(1,2,3)-pyrene	0.5-1.7	0.9	1.7-2.0	1.9	0.5	0.5	0.5	0.5	0.5-1.5	1.0	0.5-1.8	1.2
5-methylchrysene	<0.7		0.4-0.7	0.6	<0.7		0.4	0.4	<0.7		0.4-0.7	0.6
Σ 12all PAHs		13		20		10		11		8.6		13

Concentrations below determination limit (LOQ) were counted as LOQ/2 to calculate mean levels of PAHs.

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Table V. Estimated daily intake of benzo(a)pyrene (ng per day and ng kg⁻¹ body weight per day) from the consumption of meat products by Estonian children.

Age	Daily intake of BaP (ng per day)								Daily intake of BaP (ng kg ⁻¹ bw per day)	
	Cooked sausage		Smoked sausage and ham		Grilled meat products		Total		Mean	Max
	Mean	Max	Mean	Max	Mean	Max	Mean	Max		
1-3	2.7	7.7	2.4	37	1.2	21	6.3	66	0.47	4.7
4-6	4.8	13	4.3	37	1.9	19	11	69	0.55	3.5
7-9	4.8	17	6.2	86	2.2	37	13	140	0.47	5.0
10-12	4.2	20	8.2	76	2.7	37	15	133	0.40	3.5
13-16	6.8	20	13.3	53	5.1	18	25	91	0.39	1.4
Mean	4.7		6.9		2.6		14		0.46	

Table VI. Estimated daily intake of all PAHs (ng per day and ng kg⁻¹ body weight per day) from the consumption of meat products by Estonian children.

Age	Daily intake of PAH (ng per day)								Daily intake of PAH (ng kg ⁻¹ bw per day)	
	Cooked sausage		Smoked sausage and ham		Grilled meat products		Total		Mean	Max
	Mean	Max	Mean	Max	Mean	Max	Mean	Max		
1-3	58	118	18	278	17.5	325	94	721	7.0	53
4-6	102	275	32	278	30	300	164	853	8.2	43
7-9	102	355	47	645	34	575	183	1575	6.5	56
10-12	90	426	62	570	43	575	195	1575	5.1	41
13-16	144	426	98	398	80	288	322	1112	5.0	17
Mean	99		51		41		192		6.4	

Table VII. Mean daily intake of benzo(a)pyrene and PAHs from the consumption of meat products in general population in Estonia calculated on the basis of the food consumption survey from the National Board of Statistics and the theoretical consumption of 100g of each product per day.

Product group	Average consumption (g per day)	Mean BaP content ($\mu\text{g kg}^{-1}$)	Mean $\Sigma 12$ PAHs content ($\mu\text{g kg}^{-1}$)	Mean BaP intake (ng per day)		Mean total PAH intake (ng per day)	
				At average daily consumption	Through daily consumption of 100g	At average daily consumption	Through daily consumption of 100g
Cooked sausage	35	0.15	3.2	5.3	15	112	320
Smoked sausage	10	1.1	7.3	11	110	75	730
Ham	2.0	0.8	8.0	1.6	80	16	800
Smoked meat	6.7	0.7	8.0	4.7	70	54	800
Smoked chicken	1.0	1.3	5.7	1.3	130	5.7	570
Grilled chicken	1.7	0.5	11	0.85	50	18	1100
Grilled sausage	2.0	0.6	10	1.2	60	21	1000
Grilled meat	2.7	1.1	16	3.0	110	44	1600
Others	2.9	0.15	-	0.45	15	-	-
Total	64			29		346	