Investigation into polycyclic aromatic hydrocarbons (PAHs) in banana chips
Report to the Food Standards Agency
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Investigation into polycyclic aromatic hydrocarbons (PAHs) in banana chips

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Opinions and interpretations are outside the scope of UKAS accreditation. The following reported analyses fall within the scope of UKAS accreditation: PAHs
# Glossary of Main Terms

<table>
<thead>
<tr>
<th>Term or Acronym</th>
<th>General Meaning Of Term</th>
</tr>
</thead>
<tbody>
<tr>
<td>BaP</td>
<td>Benzo[a]pyrene</td>
</tr>
<tr>
<td>EFSA</td>
<td>European Food Safety Authority</td>
</tr>
<tr>
<td>FAPAS</td>
<td>Food Analysis Proficiency Assessment Scheme</td>
</tr>
<tr>
<td>JRC-IRMM</td>
<td>Joint Research Centre - Institute for Reference Materials and Measurements</td>
</tr>
<tr>
<td>PAHs</td>
<td>Polycyclic aromatic hydrocarbons</td>
</tr>
<tr>
<td>PAH 4 Sum</td>
<td>Sum of 4 PAHs (beno[a]pyrene, benz[a]anthracene, benzo[b]fluoranthene, chrysene)</td>
</tr>
<tr>
<td>Whole weight</td>
<td>Values based on the sample as received ‘whole’ or wet</td>
</tr>
<tr>
<td>Lower bound</td>
<td>assumes values at less than the limit of detection are zero (e.g.&lt;0.01=0)</td>
</tr>
<tr>
<td>Upper bound</td>
<td>assumes values at less than the limit of detection are equal to the limit of detection (e.g. &lt;0.07=0.07)</td>
</tr>
<tr>
<td>µg/kg</td>
<td>Microgram per kilogram (x 10^-6/ part per billion)</td>
</tr>
<tr>
<td>HRGC-LRMS</td>
<td>High resolution gas chromatography – unit resolution mass spectrometry</td>
</tr>
<tr>
<td>UKAS</td>
<td>United Kingdom Accreditation Service</td>
</tr>
</tbody>
</table>
Executive Summary

There are known to be elevated levels of Polycyclic aromatic hydrocarbons (PAHs) in banana chips which could arise through the drying process or during the cooking stage, which usually involves frying in coconut oil. PAHs constitute a large class of organic compounds with two or more fused aromatic rings and occur in complex mixtures consisting of hundreds of compounds. As contaminants of food they are of concern because some of these compounds have been identified as genotoxic carcinogens, mutagens and teratogens.

106 retail banana chip samples were sourced locally from supermarkets, health food stores, ethnic food suppliers and from internet retailers. These included 88 samples prepared in coconut oil, 4 in palm oil, 4 sundried and 10 that were supplied with no information on product preparation.

The samples were homogenised and analysed using an established, validated analytical procedure for PAHs that is UKAS accredited to the ISO 17025 standard and has been reported before in detail to the FSA. The methodology is based on internal standardisation (\(^{13}\)Carbon) with measurement by GC-MS.

Of the 28 PAH compounds monitored, those with lower molecular weight were found to occur in the majority of the samples. Of the more toxicologically significant and regulated compounds, benzo[a]pyrene concentrations ranged from <0.06 to 3.22 \(\mu\)g/kg of product, and PAH4 SUM concentrations ranged from 0.29 to 54 \(\mu\)g/kg of product. Approximately two thirds of the samples showed relatively low levels with PAH4 SUM concentrations occurring below 5\(\mu\)g/kg. Samples showing the highest PAH4 SUM concentrations (n=11), were all found to have been prepared using coconut oil.
1. Study Background

There are known to be elevated levels of Polycyclic aromatic hydrocarbons (PAHs) in banana chips due to the cooking/drying methods used to prepare them, which usually involve frying in coconut oil.

PAHs are a complex group of chemicals with two or more fused aromatic ring systems. The molecules do not contain heteroatoms and are not substituted. PAHs occur naturally in coal, crude oil and tar deposits, and can be inadvertently produced as by-products of incomplete fossil fuel or biomass combustion. As environmental pollutants, they are of concern because some compounds have been identified as genotoxic carcinogens, mutagens and teratogens. They occur widely in the environment and their environmental transport is governed by their volatility and chemical reactivity. They are also lipophilic with poor aqueous solubility and unless metabolised, tend to occur in the lipid rich tissues of plants and animals.

The PAH content of some food is regulated in the EU based on the conclusions of EFSA, and uses the system of maximum permitted levels for the sum of four substances (PAH4; the sum of benzo(a)pyrene, benz(a)anthracene, benzo(b)fluoranthene and chrysene) whilst maintaining a separate maximum level for benzo(a)pyrene (BaP).

The processing of food – smoking, cooking over a direct heat source (barbecuing), drying etc, are generally recognised as a major source of PAH contamination in food (Lijinsky and Ross 1967, Saint-Aubert et al 1992, Lintas et al 1979, White et al 2008). In the context of the current investigation, the most commonly used process for preparation of banana chips involves frying in coconut, or other highly saturated, oils. The condensation and absorption mechanisms that occur on the surface of the food are likely to lead to the PAHs being incorporated into the food.

The FSA has carried out a number of studies on PAHs over the last few years, and these have usually been targeted towards certain foods such as smoked shellfish, cereals and dried and smoked products which may be expected to show the presence of PAHs due to processing. Data from the current study will allow background concentrations to be established and should help with the risk assessment and discussion of any potential regulation on the PAH content of banana chips.
2. Study Experimental

2.1 Sample Collection and Preparation

A list of samples including a description and FERA sample number is given in Table 1.

106 retail banana chip samples and products containing substantial quantities of banana chips, such as cereals and trail mixes, were sourced locally from supermarkets, health food stores, ethnic food suppliers and also from internet retailers. There were a total of 88 samples prepared in coconut oil, 4 in palm oil, 4 sundried and 10 that were supplied with no information of how they were prepared. During the initial sampling exercise, limited numbers of samples of banana chips prepared in coconut oil were found to be available, so duplicate products with different batch codes or best before dates were obtained where possible, for those found to have both elevated and lower levels of PAH.

On receipt at the laboratory, each sample was given a unique laboratory reference number and the sample details were logged into a database. For cereals and other mixes, components other than banana chips were separated and excluded from the analytical sample. All samples were ground to a small particle size (<1mm), and were then thoroughly homogenised. Approximately 5g aliquots of sample were taken for analysis.

2.2 Contaminants measured – Specific Analytes

The following analytes were determined: Regulated contaminants are highlighted in **bold**.


2.3 Analytical Methodology

The analytical methodology for the PAHs has been published (Rose et al, 2007) and reported before in detail to the FSA (Fernandes et al 2011). The methodology is based on internal standardisation (\(^{13}\)Carbon) with GC-MS measurement. An aliquot of the homogenised sample was
fortified with $^{13}$C-labelled analogues of target compounds and saponified with methanolic potassium hydroxide. The extracted PAH solutions were purified in two stages with a DMF/cyclohexane partition followed by adsorption chromatography on activated silica. A sensitivity standard was added to the purified extracts and these were measured using high resolution gas chromatography with unit resolution mass spectrometry.

2.4 Quality Control

Fera is the UK National Reference Laboratory for PHs in food. The analytical procedure for PAHs is UKAS accredited to the ISO 17025 standard and includes the assessment of method blanks and reference materials, (RM0651, PAHs in palm oil – FAPAS 2012) for compliance with the accreditation criteria. The methodology also meets the criteria required for evaluating data against the maximum permitted limits for benzo[a]pyrene as specified in EU Commission Regulations. Additionally, FERA has also participated in recent international inter-comparison exercises (JRC-IRMM 2012, 2013 FAPAS 2012, 2013) where results reported by the laboratory were in good agreement with consensus data.
3. Results & Discussion

The concentrations of the 28 PAH compounds measured in this study are given in Table 2. The concentrations are reported in µg/kg of banana chip product as obtained or µg/kg of banana chip portion of product where the banana chips were part of a cereal or mix. Data was rounded to two decimal places for all analytes. Measurement uncertainty has been included for each sample. The reporting limits quoted (as “<”) for all analytes was the limit of determination that prevailed in that instance. The limit is calculated dynamically for each compound in each sample, and takes into account instrument signal-to-noise levels, sample weight, analytical recovery and concentrations detected in method blanks.

Of the 28 PAH compounds monitored, those with lower molecular weight were found to occur in the majority of the samples. BaP concentrations for the regulated compounds ranged from <0.06µg/kg to 3.22µg/kg and PAH4 SUM concentrations ranged from 0.29µg/kg to 54.39µg/kg.

Approximately two thirds of the samples showed relatively low levels with PAH4 SUM concentrations occurring below 5µg/kg, and these products encompassed the range of preparation techniques used (sun-drying, palm and coconut oil frying). Sundried products, however, were found to show lower levels of PAH contamination, with only one of these products having a PAH4SUM value above 5µg/kg. On the other hand, the 10 % of samples with the highest concentrations were all found to have been prepared using coconut oil.
4. References


FAPAS Proficiency Test 0651 Report, Environmental Contaminants, PAHs in palm oil, June 2012. (Consensus value data from participating laboratories used for establishing acceptance criteria for use as an in-house reference material.)


### Table 1: Overview of samples

<table>
<thead>
<tr>
<th>OEC Sample No.</th>
<th>FERA LIMs No.</th>
<th>Product</th>
<th>Lot #</th>
<th>BB Date:</th>
<th>Oil used</th>
</tr>
</thead>
<tbody>
<tr>
<td>22120</td>
<td>S13-063208</td>
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<td>S13-048984</td>
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<tr>
<td>21650</td>
<td>S13-048818</td>
<td>Organic Banana Chips Yoghurt White Chocolate</td>
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<td>21668</td>
<td>S13-049139</td>
<td>Sweetened Banana Chips</td>
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<td>21652</td>
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<td>S13-047888</td>
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<td>Lot #</td>
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<td>Oil used</td>
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<td>22252</td>
<td>S14-005438</td>
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<td>21960</td>
<td>S13-049301</td>
<td>Sweetened Dried Banana Chips</td>
<td>319940543A</td>
<td>31/07/2014</td>
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<tr>
<td>22032</td>
<td>S13-058725</td>
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<td>ID: 319940543A</td>
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<tr>
<td>21614</td>
<td>S13-047869</td>
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<td>ID: D 224</td>
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<td>21556</td>
<td>S13-045689</td>
<td>Tropical Sunshine mix 7% Bananas</td>
<td>N/A</td>
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<td>palm oil</td>
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<td>21628</td>
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<tr>
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<tr>
<td>21547</td>
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<td>BBE 10 MAR 2014</td>
<td>Coconut</td>
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<tr>
<td>21549</td>
<td>S13-045667</td>
<td>Plantain Chips (Savoury)</td>
<td>D1 2</td>
<td>31 01 14</td>
<td>palm oil</td>
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<tr>
<td>22268</td>
<td>S14-009648</td>
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<td>21672</td>
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<td>21658</td>
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<td>Dried Banana Chips</td>
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<td>S13-049164</td>
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<td>BBE: Aug 2014</td>
<td>Coconut</td>
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<td>22113</td>
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<td>Crunchy Oat Granola Tropical Fruits</td>
<td>Lot; LD3273</td>
<td>BB: 31/5/14</td>
<td>Coconut</td>
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<tr>
<td>22114</td>
<td>S13-063197</td>
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<td>Lot; LC3274</td>
<td>BB: 31/5/14</td>
<td>Coconut</td>
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<tr>
<td>21653</td>
<td>S13-048821</td>
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<td>ID: 1172943</td>
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<td>Coconut</td>
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<td>Coconut</td>
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<td>OEC Sample No.</td>
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<td>Product</td>
<td>Lot #</td>
<td>BB Date:</td>
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<tr>
<td>----------------</td>
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<tr>
<td>21639</td>
<td>S13-047894</td>
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<td>Muesli Luxury Fruit &amp; Nut</td>
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<td>S13-063214</td>
<td>Muesli 55% added Fruit</td>
<td>Lot 3298</td>
<td>BB: 25/10/14</td>
<td>Coconut</td>
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<td>S13-063215</td>
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<td>BB: 13/11/14</td>
<td>Coconut</td>
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<td>22128</td>
<td>S13-063216</td>
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<td>Lot L3301</td>
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<td>Coconut</td>
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<td>S14-009863</td>
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<td>BB: 09 12 2014</td>
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</tr>
<tr>
<td></td>
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<td>Prod: FRBNC04</td>
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<td>LS3/268/A 08:28</td>
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<td>Banana Chips</td>
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<td>21546</td>
<td>S13-045537</td>
<td>Sun Dried Bananas</td>
<td>13 157 1236 H74754</td>
<td>BBE Mar 2014</td>
<td>sun dried</td>
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<tr>
<td>21553</td>
<td>S13-045671</td>
<td>Banana Chips coated in Chocolate and Yoghurt</td>
<td>13149 5 74328</td>
<td>BBE Mar 2014</td>
<td>Coconut</td>
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Table 2. PAH concentrations - µg/kg whole weight

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<th>Fruit &amp; Fibre ug/kg whole weight</th>
<th>%U</th>
<th>Banana Chips ug/kg whole weight</th>
<th>%U</th>
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PAH 4 Sum Lower µg/kg 1.20 4.51 37.32 30.12
PAH 4 Sum Upper µg/kg 1.20 61 4.51 37.32 3 30.12 3
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| PAH 4 Sum Upper ug/kg | 3.52 | 15 | 3.52 | 23.12 |</p>
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| PAH 4 Sum Upper ug/kg     | 36.25          | 3             | 0.93              | 2.03  | 37 | 15.59 | 4 |</p>
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<p>| PAH 4 Sum Lower ug/kg                           | 2.54         | 3.24         | 1.23         | 54.39        |
| PAH 4 Sum Upper ug/kg                           | 2.54         | 21           | 3.24         | 12           | 1.44         | 67           | 54.39        | 2            |</p>
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<p>| PAH 4 Sum Lower ug/kg           | 42.67        | 18.88                       | 19.69        | 3.99         |
| PAH 4 Sum Upper ug/kg           | 42.67        | 2                           | 18.88        | 4            | 19.69        | 4            | 3.99         | 13           |</p>
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PAH 4 Sum Lower ug/kg  
4.30  18.47  2.66  4.01

PAH 4 Sum Upper ug/kg  
4.30  12  18.47  4  2.66  18  4.01  12
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| PAH 4 Sum Upper ug/kg          | 0.29               | 341  | 3.29 | 1.22 | 50  | 2.06 | 26 |</p>
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</table>

**PAH 4 Sum Lower ug/kg**

| 1.33 | 0.08 | 4.10 | 0.51 |

**PAH 4 Sum Upper ug/kg**

<p>| 1.33 | 50 | 0.25 | 377 | 4.10 | 13 | 0.51 | 129 |</p>
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<td>Yoghurt Banana Chips</td>
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<td>%U</td>
<td>%U</td>
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<p>| PAH 4 Sum Lower ug/kg | 1.31 | 0.93 | 1.25 | 0.86 |
| PAH 4 Sum Upper ug/kg | 1.31 | 40   | 0.99 | 67   | 1.25 | 49   | 1.05 | 86   |</p>
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<td><strong>&lt;0.06 201</strong></td>
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<td>%U</td>
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<p>| PAH 4 Sum Lower ug/kg | 0.71 | 1.90 | 8.72 | 6.71 |
| PAH 4 Sum Upper ug/kg | 0.71 | 80 | 1.90 | 41 | 8.72 | 7 | 6.71 | 9 |</p>
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<tr>
<td>benzo[k]fluoranthene</td>
<td>0.23</td>
<td>39</td>
<td>0.08</td>
</tr>
<tr>
<td>benzo[e]pyrene</td>
<td>0.60</td>
<td>20</td>
<td>0.26</td>
</tr>
<tr>
<td><strong>benzo[a]pyrene</strong></td>
<td><strong>0.47</strong></td>
<td>54</td>
<td><strong>0.17</strong></td>
</tr>
<tr>
<td>indeno[1,2,3-cd]pyrene</td>
<td>0.21</td>
<td>41</td>
<td>0.09</td>
</tr>
<tr>
<td>dibenzo[a]anthracene</td>
<td>&lt;0.05</td>
<td>201</td>
<td>&lt;0.05</td>
</tr>
<tr>
<td>benzo-[g,h,i]perylene</td>
<td>0.26</td>
<td>22</td>
<td>0.13</td>
</tr>
<tr>
<td>anthanthrene</td>
<td>&lt;0.1</td>
<td>201</td>
<td>&lt;0.1</td>
</tr>
<tr>
<td>dibenzo[a,j]pyrene</td>
<td>&lt;0.1</td>
<td>201</td>
<td>&lt;0.1</td>
</tr>
<tr>
<td>dibenzo[a,e]pyrene</td>
<td>&lt;0.1</td>
<td>201</td>
<td>&lt;0.1</td>
</tr>
<tr>
<td>dibenzo[a,i]pyrene</td>
<td>&lt;0.1</td>
<td>201</td>
<td>&lt;0.1</td>
</tr>
<tr>
<td>dibenzo[a,h]pyrene</td>
<td>&lt;0.1</td>
<td>201</td>
<td>&lt;0.1</td>
</tr>
<tr>
<td>coronene</td>
<td>&lt;0.1</td>
<td>201</td>
<td>&lt;0.1</td>
</tr>
</tbody>
</table>

**PAH 4 Sum Lower ug/kg** | 7.23 | 2.81
**PAH 4 Sum Upper ug/kg** | 7.23 | 8 | 2.81 | 27
Table 3. Quality Control Procedures

In order to demonstrate that adequate confidence can be placed in the results obtained, the following requirements were observed.

Each batch of samples analysed included a full reagent blank, the contribution from which was found to be negligible. For further confidence each batch of samples analysed included a reference material (T0651 Palm Oil), for which results were compared with a criteria range derived from the assigned consensus data from multiple laboratory testing. Results for the batch RM fall within the acceptable range.

Quality Control Results

<table>
<thead>
<tr>
<th>Compound</th>
<th>Assigned Value (ug/kg) From Consensus Data</th>
<th>Target Standard deviation σp (ug/kg)</th>
<th>Acceptable Range ug/kg</th>
<th>9988</th>
<th>9991</th>
<th>10154</th>
<th>10129</th>
<th>10201</th>
</tr>
</thead>
<tbody>
<tr>
<td>benz (a) anthracene</td>
<td>2.34</td>
<td>0.515</td>
<td>1.83-2.86</td>
<td>2.04</td>
<td>2.12</td>
<td>2.17</td>
<td>2.15</td>
<td>2.19</td>
</tr>
<tr>
<td>benzo[b]fluoranthene</td>
<td>1.97</td>
<td>0.434</td>
<td>1.54-2.40</td>
<td>1.82</td>
<td>1.65</td>
<td>1.58</td>
<td>1.66</td>
<td>1.69</td>
</tr>
<tr>
<td>benzo[a]pyrene</td>
<td>1.58</td>
<td>0.347</td>
<td>1.23-1.93</td>
<td>1.55</td>
<td>1.40</td>
<td>1.50</td>
<td>1.54</td>
<td>1.48</td>
</tr>
<tr>
<td>indeno[1,2,3-cd]pyrene</td>
<td>1.56</td>
<td>0.344</td>
<td>1.22-1.90</td>
<td>1.48</td>
<td>1.37</td>
<td>1.41</td>
<td>1.44</td>
<td>1.45</td>
</tr>
<tr>
<td>benzo-[g,h,i]perylene</td>
<td>1.57</td>
<td>0.345</td>
<td>1.23-1.92</td>
<td>1.46</td>
<td>1.28</td>
<td>1.30</td>
<td>1.34</td>
<td>1.39</td>
</tr>
<tr>
<td>chrysene</td>
<td>2.86</td>
<td>0.630</td>
<td>2.23-3.49</td>
<td>2.34</td>
<td>2.64</td>
<td>2.45</td>
<td>2.7</td>
<td>2.64</td>
</tr>
<tr>
<td>PAH4 (sum)</td>
<td>8.50</td>
<td>1.870</td>
<td>6.63-10.37</td>
<td>7.75</td>
<td>7.81</td>
<td>7.70</td>
<td>8.05</td>
<td>8.00</td>
</tr>
</tbody>
</table>
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