Polycyclic aromatic hydrocarbons in spices and dried herbs and associated risk for the Belgian population

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ABSTRACT
A total of 120 spices and herbs bought on the Belgian market have been analysed for occurrence of the four EU regulated polycyclic aromatic hydrocarbons (ΣPAH4). Samples were sorted in three groups according to their use: traditional spices, exotic spices, and herbs. Benzo[a]pyrene has been detected in 56% of samples, while ΣPAH4 were found in 96% of samples. ΣPAH4 were evenly distributed among the three groups with a median content of 7.2 for traditional spices, 5.5 for exotic spices and 6.0 μg kg⁻¹ for herbs. According to the EU legislation, three samples were exceeding maximal limits, with a maximum ΣPAH4 content of 164 μg kg⁻¹. The ΣPAH4 dietary intake has been estimated at 1.4 and 27.8 ng day⁻¹ for average and high consumption, respectively. The risk for the consumer was evaluated by using the margin of exposure (MOE). In all cases, MOE were >20 000, indicating a low concern for the population.

Introduction
Spices and herbs are widely used as flavourings, colourants, and aromas for dishes. Besides their organoleptic qualities, they are also used as preservatives and possess some health-beneficial properties such as antioxidant, antimicrobial, hepatoprotective, or anti-inflammatory effects (Kurian 2012). As the global market of spices and herbs has been expanding continuously for two decades and considering their process of production, questions may arise regarding consumer safety (Businesscoot 2021; CBI 2022). Indeed, the presence of pesticide residues, mycotoxins, and heavy metals has already been reported in the literature (Mubeen et al. 2009; Yogendararajah et al. 2014; Reinholds et al. 2016, 2017) and in the Rapid Alert System of Food and Feed (RASFF) of the European Commission with 112 notifications for pesticides residues, 73 for mycotoxins, and 6 for heavy metals for the 2020–2022 period.

Additionally, polycyclic aromatic hydrocarbons (PAHs) have also been found in large quantities in spices and dried herbs in studies over the last 15 years (EFSA 2008). PAHs are a large class of organic compounds composed of two or more fused aromatic rings (Harvey 1991). Food is considered the main source of exposure to PAHs for a non-smoker. The contamination may occur via food processing (e.g. grilling, roasting, drying, or smoking). However, environmental pollution cannot be excluded for plant products such as spice and herbs. Indeed, PAHs, produced in industrial areas or originated from heavy motor traffic, may be bound to fine particles and be adsorbed on the waxy foliar surface via atmosphere deposition (Igneti et al. 1992; Howsam and Jones 1998; Phillips 1999). PAHs can also contaminate the soil and surface water and then be translocated in the plant before the harvest (Scientific Committee on Food 2002).

The primary concern of PAHs is their carcinogenic and mutagenic properties (Phillips 1999). In 2002, the European Scientific Committee on Food assessed the toxicity of many PAHs, thereby identifying 15 PAHs as genotoxic or carcinogenic (Scientific Committee on Food 2002). Afterwards, the CONTAM Panel of the European Food Safety Authority (EFSA) concluded in 2008 that monitoring of 4 PAHs (i.e. benzo[a]anthracene (BaA), chrysene (CHR), benzo[b]fluoranthene (BbF), and benzo[a]pyrene (BaP)) would be suitable for evaluating the overall occurrence of PAHs in food (EFSA 2008). As a result, the European Commission established maximum levels (MLs) for the sum of these four PAHs and the individual occurrence of BaP in different food matrices (European Commission 2011a). Specifically for spices and herbs, the MLs were set at 50.0 μg kg⁻¹ for the sum of the four regulated PAHs...
(ΣPAH4) and 10.0 μg kg⁻¹ for BaP (European Commission 2015). Since the establishment of these MLs, 12 severe alert notifications have been reported by RASFF in the period 2020–2022.

Although the Regulation (EU) No 2015/1933 entered into force 7 years ago, only a few specific methods dedicated explicitly to PAHs in herbs and spices can be found in the literature. One reason may be the analytical challenge posed by the broad and heterogeneous group of spices and herbs. This food group contains many compounds similar to PAHs that are the major obstacle hampering correct quantification (Sztrenfeld et al. 2019). HPLC-UV/FLD analysis, in particular, may lead to overestimating PAH content as the lack of specificity inherent to this technique avoids the distinction between PAHs signals and matrix interferences. The use of GC-MS/MS efficiently solves this problem (Bratinova et al. 2016). However, if the clean-up is insufficient, the matrix may compromise the adequate separation of the PAH isomers. This is the major concern during GC-MS/MS analysis. Nevertheless, some methods were already reported.

Rozentale et al. (2018) developed a method based on extraction with a dichloromethane (DCM)/hexane mixture, followed by gel permeation chromatography and purification on silica. Even though such technique can be time and solvent consuming, it allowed them to analyse a variety of spices and herbs matching the analytical EU criteria (European Commission 2011b). Urban and Lesueur (2017) considered the use of a modern method such as QuEChERS extraction, initially designed for pesticides analysis in food (Payá et al. 2007), followed by a Captiva Enhanced Matrix Removal-lipid clean-up, a sorbent design to remove lipids based on size exclusion and hydrophobic interaction. With this method, time and solvent consumption are drastically reduced. Although the obtained extract was less cleaned, which needed more frequent instrument maintenance, Urban and Lesueur (2017) successfully used it to quantify PAHs in curry powder.

More recently, Sztrenfeld et al. (2019) proposed a GC-MS/MS method capable of facing the wide variety of spices and herbs available on the market. The method is based on extraction with DCM and hexane followed by a specific Solid Phase Extraction (SPE) clean-up, depending on the spices and herbs considered. This modular method allows very low limits of quantifications (LOQs), matching all criteria of the legislation in force (European Commission 2011b). However, the use of DCM is forbidden in some EU countries. Therefore, in 2020 the European reference Laboratory for processing contaminants (EURL-PC) initiated an inter-laboratory validation of an analytical method adapted from Sztrenfeld et al. (2019), replacing the use of DCM with ethyl acetate and a different SPE sorbent (Sina Wilde and Duedahl-Olesen 2021). Even when lesser cleaned extracts are obtained, this method allows the analysis of a wide variety of matrices similar to spices and herbs with acceptable LOQs for the four regulated PAHs.

Nevertheless, the number of studies dealing with PAHs monitoring in spices and herbs are still limited. Zelinkova and Wenzl (2015) have studied PAHs content in plant and herbal food supplements that are of the same nature and following a drying process very similar to spices and herbs. Their main findings illustrated non-systematic ΣPAH4 contamination originating from environmental pollution and processing techniques, mainly due to bad drying practices. Moreover, high ΣPAH4 content was found in some samples, which could remarkably increase the daily exposure to these substances, stressing the need to continue to monitor this kind of food matrices.

A study of Bogdanović et al. (2019), quantifying PAHs in various spices and herbs in a limited number of samples (20), concluded that the PAHs content is much higher in these matrices. A recent study by Rozentale et al. (2018), focusing on six spices and herbs (thyme, pepper, nutmeg, basil, oregano, paprika), confirmed through the analysis of 150 samples the non-systematic ΣPAH4 contamination observed by Zelinkova and Wenzl (2015). Furthermore, they recommended collecting data to properly manage the risks associated with the presence of PAHs in spices and herbs.

Furthermore, as spices and herbs consumption is continuously increasing over the years (Business scoot 2021), a deeper investigation is needed on the occurrence of ΣPAH4 in spices and herbs. In this contribution, an extensive study on the occurrence of the four regulated PAHs (i.e. BaA, Chr, BbF, and BaP) in 120 spices and dried herbs available on the Belgian market is presented. The compliance of samples compared to maximal limits from the Regulation (EU) No 2015/1933 and the potential risk for the Belgian population have been assessed.

Materials and methods

Chemicals and reagents

A mixture of 4 regulated EU PAHs: BaA, CHR, BbF and BaP (10 ng μl⁻¹ in cyclohexane) and a mixture of isotopically-labelled (deuterated) standards (IL-IS): BaA D12, CHR D12, BbF D12, BaP D12 were purchased from Dr. Ehrenstorfer Gmbh (Ausburg, Germany). Hexane (HPLC grade) was from Biosolve ( Valkenwaard, The
Netherlands), while DCM (HiperSolve HPLC™) was supplied by BDH Chemicals Ltd. (Poole, England). Bond Elut SI (silica) cartridges (500 mg, 3 ml) were purchased from Agilent Technologies (Machelen, Belgium). Empty SPE cartridges (6 ml) of Biotage Isolute® (Sopachem, Gent, Belgium) were filled with Florisil (60–100 mesh) from Fluka (Sigma Aldrich, Overijse, Belgium) to prepare in-house SPE cartridges. SupelMIP™ SPE cartridges were supplied by Supelco (Sigma Aldrich, Overijse, Belgium).

Working solutions of PAHs were prepared from the commercial solution at 10 µg ml⁻¹ in cyclohexane, resulting in a concentration of 1 µg ml⁻¹ and 0.1 µg ml⁻¹. Working solutions of IL-IS PAHs were prepared by diluting the commercial solution at 10 µg ml⁻¹ in hexane to obtain a concentration of 1 µg ml⁻¹. All solutions were stored at −20 ºC.

**Sampling strategy**

120 Samples were selected in 2019 based on the representativeness of the Belgian market and the habits of consumers. The choice of brands was based on Euromonitor’s Belgian market share data (Euromonitor Grocery universe 2017). Organic stores were also included to reflect as much as possible Belgians’ shopping habits. The availability of food products on the Belgian market was also considered a parameter to modulate the number of samples per type of spices or herbs.

**Sample homogenisation**

Samples were milled to a fine powder and subsequently homogenised in accordance with Regulation (EU) No 836/2011 (European Commission 2011b) using a IKA M20 mill (IKA®-Werke GmbH & Co. KG, Staufen, Germany), suitable for dry grinding and powder matrices.

**Extraction and purification**

Afterwards, the PAHs were extracted using the procedure described by (Sztternfeld et al. 2019). Briefly, 1.0 g of homogenised sample was weighed and the internal standards were added (IL-IS PAHs). After equilibrating for 20 min, the PAHs were extracted with 20 ml of a mixture of hexane/DCM (3/1, v/v) using an Ultra Turrax® (IKA, Staufen, Germany). After centrifugation of 5 min at 3900 rpm, the supernatant was concentrated to approximately 1 ml and further purified using SPE. Different SPE sorbents were used, depending on the spices or herbs. Traditional spices were purified with a silica SPE cartridge, while herbs and pigmented spices were purified using a florisil sorbent. Purification of complex spices or fatty spices were first purified with silica, followed by a clean-up using SupelMIP™ PAHs SPE, which is specifically designed for the purification of PAHs in oil. After the purification step, the extracts were concentrated under a nitrogen stream to a volume of 200 µl before analysis by GC-MS/MS.

**GC-MS/MS analysis**

GC-MS/MS analyses were performed on an Agilent 7890B GC system coupled with an Agilent 7000C triple quad mass spectrometer (Agilent Technologies, Santa Clara, CA, USA) with an electronic impact source. The chromatographic column was the J&W GC column Select PAH (30 m × 0.25 mm I.D. × 0.15 µm dₜ), and the liner inlet was a single taper (4 mm I.D.) containing wool, also from Agilent. Injections were performed in splitless mode at 300°C with an injection volume of 2 µl. Helium (99.99999%) was used as carrier gas at a flow rate of 1.2 ml min⁻¹. The oven temperature was programmed according to an Agilent application note (Provoost 2010). The initial temperature was set at 70°C (held for 0.4 min) and increased at a rate of 85°C min⁻¹ to reach 180°C, followed by an increase with 3°C min⁻¹ to 230°C (held for 7 min) and up to 280°C (held for 10 min) with a speed of 28°C min⁻¹. Finally, the temperature increased with 14°C min⁻¹ up to 325°C, held for 3 min. The following source parameters were used: temperature of 280°C and electron energy fixed at 70 eV. Data acquisition was performed in MRM mode, and 2 transitions were selected for each PAH. The results were interpreted using the MassHunter software from Agilent. Two criteria were used for the correct identification of the PAHs: (i) retention time difference between the native compound, and its IL-IS should not differ more than 0.1 min; (ii) ion ratio of the two most abundant product ions (i.e. 2 MRM transitions) should be within the range of 30% compared to the closest calibration standard. Finally, relative areas of analyte and corresponding internal standards were used for the quantification.

**Quality assurance and control**

The sensitivity and stability of the GC-MS/MS system have been controlled by injecting a calibration point at LOQ level (0.5 µg kg⁻¹) at the beginning and the end of the run. A signal-to-noise ratio >3 for all individual PAH for both injections ensures there was no sensitivity loss. Additionally, the concentrations of each calibration level were back-calculated using the regression equations of the calibration curve. The deviation should be lower than
±20%, illustrating accurate PAHs quantification. A procedural blank was analysed for each batch of samples to ensure that no PAH contamination occurred, either at the level of the extractions or at the GC-MS/MS system level. Extraction efficiencies were evaluated for each analysis batch by checking the recovery of a control sample fortified at 10.0 μg kg⁻¹. Results were plotted on a control chart to ensure no method deviation occurred. The method has been validated in accordance with criteria laid down in Regulation (EU) No 836/2011 (European Commission 2011b; Szternfeld et al. 2019). Finally, the developed methodology was applied successfully in two proficiency tests, i.e. EURL PC-PT-2016 on smoked pepper and EURL-PC-PT-2019 on herbal food supplements (Bratinova et al. 2016; Duedahl-Olesen 2019). Acceptable z-scores (|z-score|<2) were obtained for all individual regulated PAHs demonstrating that this method is fit-for-purpose.

**Dietary exposure**

The Food Additives Intake Model (FAIM) is a tool for estimating chronic dietary exposure to food additives. In the first version of the FAIM template, summary statistics from the Comprehensive Database to estimate chronic dietary exposure in different population groups and EU countries were included (EFSA 2011). Although the FAIM tool was initially developed for food additives, it contains valuable information regarding the consumption of spices and herbs. Indeed, the consumption records in the FAIM tool are codified according to the food categories as presented in Annexe II, Part D, of Regulation (EC) No 1333/2008 (European Commission 2011c). One of these categories is dedicated explicitly to herbs and spices (i.e. category 12.2.1). The 1st version of this tool contained the Belgian consumption data of 2004 for this specific category for both the mean of the total population (i.e. 0.0017 g kg⁻¹ body weight (bw) day⁻¹) and the high level for consumers only (i.e. 0.03412 g kg⁻¹ bw day⁻¹). Hence, consumption data present in this tool were then combined with the average concentration data for ΣPAH4 obtained in this study to evaluate the exposure of the Belgian population. The values reported as "< LOQ" (0.5 μg kg⁻¹) have been replaced by LOQ/2 (i.e. 0.25 μg kg⁻¹) to perform medium bound calculations. Afterwards, the exposure due to spices and herbs was added to the overall exposure determined by EFSA (2008).

It should be noted that only the average concentration data were considered. No worst-case scenario using the maximal level found for the ΣPAH4 has been assessed. The reason is that consumption data were not detailed for each spices and herbs. Hence if the maximum concentration would be used instead, it would be applied for spices and herbs for which very low concentration have been observed and would lead to abnormally high and unrealistic dietary exposures.

**Risk assessment**

Risk assessment was based on the margin of exposure (MOE) approach as proposed by EFSA for risks associated with genotoxic and carcinogenic compounds in food and feed such as PAHs (EFSA 2005). The MOE is the ratio between a defined point on the dose-response curve for the adverse effect of ΣPAH4 and the exposure to ΣPAH4. This specific point refers to the benchmark dose (BMD) for which a low but measurable response is seen. In this case, 10% incidence above the population control (BMD₁₀). Then a BMD₁₀ lower limit (BMDL₁₀) corresponding to the lower limit of the one-sided 95% confidence interval of the BMD₁₀ has been defined. This lower bound allows considering uncertainty derived from a given study. If the BMDL₁₀ is derived from an animal carcinogenicity study, the EFSA Scientific Committee suggests that a MOE of 10,000 or higher would indicate a low concern for the public health (EFSA 2005). Is is calculated by MOE = \( \frac{BMDL_{10}}{\Sigma PAH4 exposure} \), where BMDL₁₀ is the benchmark dose lower confidence level in mg kg⁻¹ body weight (bw) day⁻¹ and ΣPAH4 exposure is the total amount of PAH4 ingested by the consumer in mg kg⁻¹ bw day⁻¹. In the case of ΣPAH4, the BMDL₁₀ is 0.34 mg kg⁻¹ bw day⁻¹.

**Results and discussion**

**Sampling**

In total, 120 herbs and spices were purchased in 2019 in Belgian supermarkets (75%) and organic stores and groceries (25%). Spices and herbs have been grouped depending on their nature in 3 different categories to facilitate data treatment and risk assessment: (i) traditional spices, (ii) exotic spices, (iii) dried herbs (Table S1, a supplementary table which can be obtained from the corresponding author). Spices commonly or daily used in Belgian dishes have been classified as traditional spices. Other spices primarily used in Asian or North African cooking have been classified as exotic spices. As a result, 27 samples were categorised in the group traditional spices, 38 samples were allocated to the group exotic spices, 55 samples were considered dried herbs (including 9 samples of mixed herbs).

**Occurrence data**

The results of the occurrence data (BaP and ΣPAH4) are presented in Figure 1, while all the individual results of the four PAHs are shown in the supplementary data.
Overall, the ΣPAH4 have been detected in 93% of the samples, while BaP was present in only 56% of samples (Table S1). This can be explained by the heavier 5-ring structure of BaP, which requires a higher temperature and longer processing time to be formed, compared to other PAHs (BaP, Chr) with a 4-ring system (Harvey 1991). Moreover, BaP and the ΣPAH4 were similarly distributed in each group (Figure 1b), despite the relatively high number of statistical outliers in the exotic and dried herbs groups (Figure 1a). The median BaP levels were below the LOQ for each group, while the medians for the ΣPAH4 were 7.2 µg kg⁻¹ in classic spices, 5.5 µg kg⁻¹ in exotic spices and 6.0 µg kg⁻¹ in dried herbs (Figure 1b). No difference in PAHs contamination has been noticed between supermarkets, organic stores, and groceries samples. The highest outlier of the exotic group was the smoked paprika sample with a concentration for the ΣPAH4 of 164 µg kg⁻¹. This high concentration could originate from the extra smoking step performed with oak wood. Similar results were also observed in other studies. Berki et al. (2020) reported a ΣPAH4 content ranging from 115 µg kg⁻¹ to 3476 µg kg⁻¹ for smoked paprika, which was smoked using the same type of wood. Also, Coleto et al. (2021) reported an average concentration for ΣPAH4 of 780 µg kg⁻¹ for Spanish smoked paprika. These values are much higher than this study and exceeded the MLs set in Regulation
(EU) No 2015/1933 for the ΣPAH4 (European Commission 2015). These huge contents seemed to be linked to the traditional smoking process, resulting in direct contact of smoke with the spices.

An overview of the results of the sub-categories with at least 4 samples, is presented in Table 1. The three highest outliers of the category of the dried herbs correspond to 2 samples of laurel and 1 sample of thyme with concentrations of ΣPAH4 of 154 µg kg⁻¹, 58.2 µg kg⁻¹ and 126 µg kg⁻¹, respectively. The higher concentration in laurel can be explained by their large and waxy foliar surfaces on which PAHs can readily be adsorbed (Phillips 1999; Zelinkova and Wenzl 2015). However, it can also originate from the processing method applied for drying. In some regions, in summer, herbs are dried by being exposed to the sun or using a solar dryer. However, a dryer using wood burning is typically applied in winter to overcome the high humidity. As a result, an increase in the PAH content in the final product could be observed (Coleto et al. 2021). Similarly, the higher level of ΣPAH4 observed in thyme could be explained. Unfortunately, information on the drying processes or the harvest period was not available on the labels of the products. Relatively higher ΣPAH4 concentrations have been found in “bouquet garni” compared to other herbs included in the group “dried herbs” (Table S1). Interestingly, the composition of “bouquet garni” can vary significantly, but at least 50% is constituted of laurel and thyme, which can explain the concentration found.

Only limited studies related to PAHs in spices and herbs have been reported. One of the studies, performed by Lee et al. (2018) reported the same PAHs content in black pepper, with an average content of 1.9 and 16.3 µg kg⁻¹ for the BaP and ΣPAH4, respectively. Another study by Rozentale et al. (2018), focussed on samples produced between 2010 and 2015. They reported comparable results for thyme when excluding the outlier at 126 µg kg⁻¹ without being able to identify the origin of such contamination. Regarding pepper, they found a similar mean concentration for the ΣPAH4 content (i.e. 13.2 µg kg⁻¹). Paprika, basil, turmeric, oregano and curry were less contaminated and showed very similar levels with a concentration for ΣPAH4 ranging between 4.3 µg kg⁻¹ and 6.4 µg kg⁻¹ on average. Except for basil for which they found ΣPAH4 content twice higher than this study (11.6 µg kg⁻¹), Rozentale et al. (2018) found similar results for paprika and oregano with respectively 8.0 µg kg⁻¹ and 4.6 µg kg⁻¹. Finally, rosemary and ginger showed the lowest average contamination levels in ΣPAH4 with 1.4 µg kg⁻¹ and 2.9 µg kg⁻¹, respectively. No BaP was detected in rosemary samples. Only one sample contained a level of 0.5 µg kg⁻¹, which was close to the LOQ for ginger (Table 1 and S1). A recent study of Bogdanović et al. (2019) reported the same ΣPAH4 content (1.2 µg kg⁻¹) in rosemary herbs sampled during the 2017–2018 period.

Our results agreed with the general observations made by Rozentale et al. (2018) and Zelinkova and Wenzl (2015). Indeed, although some trends can be noticed regarding concentrations of ΣPAH4 in spices and dried herbs, no unusual contamination has been noticed for specific products. The non-systematic ΣPAH4 contamination observed in this study may originate from environmental pollution or the drying process.

Finally, regarding samples compliance according to Regulation (EU) No 2015/1933, three samples (2.5%) exceeded the ML for PAHs in spices and dried herbs set at 10.0 and 50.0 µg kg⁻¹ for BaP and ΣPAH4, respectively (European Commission 2015). A smoked paprika, which also showed the highest concentration for both BaP (31.0 µg kg⁻¹) and ΣPAH4 (164 µg kg⁻¹), exceeded the ML for both limits, while a laurel exceeded the ML for ΣPAH4 (58.6 µg kg⁻¹) and the ras-el-hanout spices exceeded the ML for BaP (11.0 µg kg⁻¹). Interestingly, the results of this study were very similar to those of Rozentale et al. (2018), who analysed samples produced prior to Regulation (EU) No 2015/1933.

**Table 1.** Min, max and average BaP and ΣPAH4 content for spices and herbs for which the number of samples higher than 4.

| Spices/herbs | BaP (µg kg⁻¹) | ΣPAH4 (µg kg⁻¹) |
|--------------|--------------|-----------------|
|              | n | min - max | average | min - max | average |
| Laurel       | 4 | <LOQ – 34.0 | 13.0     | 8.8 – 154.0 | 73.9     |
| Thyme        | 6 | <LOQ – 26.4 | 6.6      | 6.1 – 126.0 | 39.0     |
| Pepper       | 15 | <LOQ – 4.8  | 1.4      | 1.3 – 40.7  | 12.3     |
| Paprika      | 7  | <LOQ – 2.0  | 1.1      | 3.0 – 9.1   | 6.4      |
| Basil        | 8  | <LOQ – 0.8  | <LOQ     | 1.0 – 9.8   | 5.7      |
| Tumeric      | 5  | <LOQ – 2.0  | 0.72     | 1.5 – 13.6  | 5.0      |
| Origano      | 5  | <LOQ – 1.3  | <LOQ     | 1.2 – 11.5  | 4.7      |
| Curry        | 6  | <LOQ – 1.0  | 0.55     | 1.7 – 7.0   | 4.3      |
| Tarragon     | 5  | <LOQ – 0.6  | <LOQ     | 1.3 – 6.8   | 3.8      |
| Ginger       | 5  | <LOQ – 0.5  | <LOQ     | 1.4 – 6.5   | 2.9      |
| Rosemary     | 4  | <LOQ       | <LOQ     | <LOQ – 1.5  | 1.4      |

**Dietary exposure**

In 2008, EFSA evaluated the dietary exposure to ΣPAH4, but spices and herbs were not considered since EFSA stated that the impact on the overall exposure would be limited. EFSA determined the exposure originating from different food categories, resulting in an overall exposure to ΣPAH4 of 1158 ng day⁻¹ for the mean consumption level (EFSA 2008). However, since the consumption of spices and herbs continuously increases (Businesscoot 2021), the additional exposure to ΣPAH4 also gains importance. Since the FAIM model contained specific consumption values for spices and herbs, these data were combined with the average contamination level of all samples, as ΣPAH4 were evenly
Table 2. Exposure to ΣPAH4 according to different consumption scenarios, associated MOE, and impact of each scenario on MOE.

| Food categories                         | Exposure ΣPAH4 (ng day⁻¹) | MOE   | decrease of MOE (%) |
|-----------------------------------------|---------------------------|-------|---------------------|
| EFSA 2008 (spices & herbs excluded)    | 1158.0                    | 20,553| -                   |
| EFSA 2008 + spices & herbs median consumption | 1159.0                  | 20,540| -0.06               |
| EFSA 2008 + spices & herbs high consumption | 1172.0                  | 20,300| -1.23               |

distributed among the 3 groups of spices and dried herbs (Figure 1). In accordance with the EFSA opinion of 2008, a body weight of 70 kg was applied, and an average-bound scenario was considered. Hence, the ΣPAH4 intake for spices and herbs was estimated at 1.4 ng day⁻¹ for the mean population and 27.8 ng day⁻¹ for the highly exposed population (P97.5). Next, the specific exposure through spices and herbs was combined with the overall exposure determined by EFSA (2008). As a result, the total dietary exposure for ΣPAH4 was 1159 ng day⁻¹ and 1186 ng day⁻¹ for average consumption and high consumption (P97.5), respectively (Table 2).

Risk assessment

Regarding the risk for the Belgian consumer, EFSA suggests that a MOE ≥10 000 would be a low health concern (if based on the BMLD₁₀ from animal study). This MOE only indicates the level of concern and must not be used to quantify the risk (EFSA 2005). The MOE values were calculated for the different scenarios and are presented in Table 2. Since all MOE values are higher than 20,000, it can be assumed that the exposure to PAHs does not pose a health risk for the Belgian population, considering average contaminations of samples. The impact of the spices and herbs consumption and consumption scenario on the MOE is very low when compared with EFSA data from 2008 that excluded this food category (EFSA 2008). Indeed, the contribution of spices and herbs resulted in a MOE decrease of only 0.12% at an average consumption and 2.34% for a high consumption (Table 2). However, the consumption data used for the exposure assessment originate from the National Consumption Survey (FCS) of 2004. A more recent National FCS is available, but these data are not yet included in the FAIM tool, nor are these data easily translated in specific consumption of herbs and spices. However, it can be assumed that the spices and herbs consumption coming from the National FCS of 2004 is underestimated. Indeed, market studies have shown that the consumption of spices and herbs has increased over the years. In France, an increase of 30% was observed from 2009 to 2018. Furthermore, the global spices market is expected to grow at 3–4% per year between 2020 and 2025. Moreover, Belgium is considered the second-largest spices and herbs consumer in Europe (Businessscoot 2021). Nevertheless, even if the consumption is underestimated, it can be assumed that the risk will remain low. Indeed, to consider spices and herbs as a concern for the consumer (MOE ≤10 000), spices and herbs consumption should be at least between 85 (high consumption) and 854 (average consumption) times higher than the consumption level recorded in 2004, which seems highly unrealistic.

Conclusions

An extensive study of spices and herbs available on the Belgian market enabled the characterisation of the impact of PAHs contamination of spices and herbs for the safety of the Belgian population. Occurrence data of 120 samples showed a similar distribution among the three groups studied, i.e. traditional spices, exotic spices and dried herbs, even if high variability for some categories, particularly in laurel and thyme has been noticed. Our findings confirmed observations from Rozentale et al. (2018) and Zelinkova and Wenzl (2015) regarding heterogenous ΣPAH4 contaminations among samples that may originate from environmental pollution or the drying process. For all consumption scenarios studied, calculated MOE values were above 20,000. Therefore it can be concluded that the exposure of the Belgian population to PAHs through spices does not pose a potential health risk.

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