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Coumarin content in cinnamon containing food products on the Danish market

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ABSTRACT

Coumarin is a hepatotoxic natural compound found in different *Cinnamomum* species such as *Cinnamomum cassia*, *Cinnamomum loureiroi*, and *Cinnamomum burmannii*; all commonly referred to as cassia. Cassia contains high amounts of coumarin in contrast to the more expensive and less used *Cinnamomum verum*, referred to as true cinnamon. Today, many commercially available food products are spiced with cassia and consequently contain coumarin. The content of coumarin in specific food categories is regulated in the European Regulation (EC) No 1334/2008. In this study, 74 food samples labeled with cinnamon were analyzed with a validated UPLC-PDA method. The analyzed content of coumarin was compared to the EU limits. The comparison showed that fine bakery ware exceeded the EU limit for coumarin in almost 50% of the cases. One sample exceeded the EU limit for coumarin with more than a factor of three. A possible explanation for this exceedance is that manufacturers of food with cinnamon lack information about the regulatory EU limits for coumarin and how to comply with the EU regulation. As an addendum, we therefore propose a practical guide to the food industry and the national food administrations. The guide describes the theoretical content of cassia that can be added to food products without exceeding the EU limits for coumarin in 99% of lots produced.

1. Introduction

The cinnamon used domestically and industrially in the preparation of food originates primarily from true cinnamon and from cassia cinnamon (simply referred to as cassia). True cinnamon belongs to the species of *Cinnamomum verum* J. S. Presl whereas cassia generally include the species of *Cinnamomum cassia* J. Presl, also known as *Cinnamomum aromaticus* Nees, *Cinnamomum loureiroi* Nees and *Cinnamomum burmannii* Blume (Ravindran, Nirmal-Babu, & Shylaja, 2004; WHO, 1999). Cassia is cheaper and generally more popular in Europe compared to true cinnamon (Blahova & Svobodova, 2012; Lungarini, Aureli, & Coni, 2008). True cinnamon and cassia taste different because of a different chemical composition. For example, cassia contains more of the aromatic compound coumarin (1-benzopyran-2-one) compared to true cinnamon (Miller, Poole, & Pawloski, 1996; Sproll, Ruge, Andlauer, Godelmann, & Lachenmeier, 2008). Coumarin was regarded as a possible genotoxic carcinogen in the 1980th and 1990th and the European Union set a general coumarin limit in food to 2 mg/kg (European Council, 1988; Lake, 1999). In 2004, new scientific data on coumarin showed a non-genotoxic carcinogenic effect, but it also showed that a subgroup of individuals was sensitive to a hepatotoxic effect from coumarin (Abraham, Wöhrlin, Lindtner, Heinemeyer, & Lampen, 2010). These data allowed the European Food Safety Authority (EFSA) to derive a tolerable daily intake (TDI) of 0–0.1 mg coumarin/kg body weight (EFSA, 2004). In 2005, a high content of coumarin in cookies (BfR, 2006) initiated increased authoritarian coumarin control and debate (Abraham et al., 2010; VKM, 2010). This, in combination with a widespread and increased use of cassia in food products, led the European Parliament and Council to evaluate the maximum coumarin limits (European Council, 1988). In 2008, the European Regulation (EC) No 1334/2008 (European Parliament and Council, 2008) was enacted with the following maximum limits for coumarin: 50 mg/kg in traditional and/or seasonal bakery ware containing a reference to cinnamon in the labeling, 20 mg/kg in breakfast cereals including muesli, 15 mg/kg in fine bakery ware, with the exception of traditional and/or seasonal bakery ware containing a reference to cinnamon in the labeling, and 5 mg/kg in desserts. Coumarin should not be added as such to food products.

The content of coumarin varies considerable in cinnamon sticks and in ground cinnamon. The contents of coumarin in cinnamon sticks were reported to be as high as 12,180 mg/kg (He et al., 2005) and 9900 mg/kg (Woehrlin, Fry, Abraham, & Preiss-Weigert, 2010),

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whereas the content in ground cinnamon was reported to range between 1740 and 7670 mg/kg (Woehrlin et al., 2010), 2650–7017 mg/kg (Blahova & Svobodova, 2012), and 5–3094 mg/kg (Lungarini et al., 2008). This shows that the distribution of coumarin in ground cinnamon is less heterogeneous than in cinnamon sticks. The very low contents (<100 mg/kg) of coumarin observed in cinnamon sticks and in ground cinnamon might originate from *Cinnamomum verum* samples and not cassia (Lungarini et al., 2008). The high variation of the coumarin content in cinnamon complicates an addition of cinnamon to food products that complies with the EU limits for coumarin.

The content of coumarin in food products was investigated in Germany in 2006–2007 (Sproll et al., 2008) and compared to the former EU limits described in the Council Directive 88/388/EEC (European Council, 1988). These limits for coumarin were 2 mg/kg with the exception of special caramels and alcoholic beverages with a limit of 10 mg/kg, and chewing gum with a limit of 50 mg/kg (European Council, 1988). The investigation showed that 85% of all cinnamon flavored cookies exceeded the EU limit of 2 mg/kg (Sproll et al., 2008). Another study found 100% of cinnamon cookies ($n = 10$) touching or exceeding the 2 mg/kg limit (Lungarini et al., 2008). In 2012, an inspection program was initiated at the Danish Veterinary and Food Administration (DVFA) to investigate if food products on the Danish market complied with the higher and current EU regulation (European Parliament and Council, 2008). Despite its absence in the EU regulation, we included tea and crisp bread with cinnamon in the investigation to examine its contribution to the coumarin intake.

Quantification of coumarin is primarily performed with chromatographic techniques such as high performance liquid chromatography (HPLC) (He et al., 2005; Lungarini et al., 2008; Maggi, Borfoni, et al., 2011; Martino, Ramaiola, Urbano, Bracco, & Collina, 2006; Sproll et al., 2008; Woehrlin et al., 2010), gas chromatography (GC) (Maggi, Martonfi, et al., 2011; Miller et al., 1996; Miller, Poole, & Chichila, 1995; Ochiai, Sasamoto, Hoffmann, & Okanoya, 2012), and the more recent introduction of ultra performance liquid chromatography (UPLC) (Wang, Avula, Nanayakkara, Zhao, & Khan, 2013). UPLC has shown its advantage over traditional HPLC and GC analysis with respect to the chromatographic separation time. This paper describes an UPLC method and investigates the coumarin content in Danish food products with cinnamon in relation to the EU limits.

As an addendum, we present a practical guide for Food Administrations and food manufacturers. The practical guide describes the theoretical amount of cassia that can be added to different food products without exceeding the EU limits for coumarin (European Parliament and Council, 2008).

2. Materials and methods

2.1. Chemicals and standards

Methanol (99%) and acetonitrile (HPLC grade) were purchased from Fisher Scientific (United Kingdom). Coumarin (≥99%) and 4-methylumbelliferone (≥98%) (MUM) were purchased from Sigma Aldrich (Denmark).

2.2. UPLC instrumentation and chromatographic conditions

The Waters Acquity UPLC H-class (Milford, MA) consisted of a separation module and a PDA detector. The system was equipped with a Waters ACQUITY UPLC HSS T3 Column 2.1 × 100 mm, 1.8 μm. The chromatographic system consisted of eluent A and B. Eluent A contained 5% methanol in demineralized water. Eluent B contained acetonitrile. Eluents were filtered through a 0.45 μm filter. The chromatographic gradient consisted of linear segments: Initial eluent was 75% A followed by 20% A at 5 min, 20% A at 6 min, 75% A at 7 min, and 75% A at 8 min. Flow rate was 0.6 ml/min, column temperature 45 °C and injection volume 5 μL. The column eluate was scanned from 240 to 340 nm at a sampling rate of 80 points/sec. Chromatograms used for quantification were obtained at 278.1 nm. Sample vials were kept in the autosampler at 30 °C and injections were performed every 8th min.

2.3. Method validation

2.3.1. Linearity

Dilution curves consisted of six dilutions of coumarin in water with concentrations of 0.1, 0.5, 1.0, 2.0, 5.0, and 10.0 mg/l (corresponding to 10× higher levels in samples, mg/kg). Dilution curves were freshly prepared and analyzed on four individual days. Dilution curves showed an $r^2 > 0.9998$ excluding 0.0 as a point on the standard curve. The r^2 in combination with a visual evaluation showed a satisfactory linearity.

2.3.2. Precision

Three samples with a natural content of coumarin were named validation sample VS1 (breakfast cereal), VS2 (brown Christmas cookie), and VS3 (cinnamon roll). The validation samples VS1, VS2, and VS3 contained approximately 1, 15, and 50 mg coumarin/kg, respectively, and were treated like normal samples. Two technicians analyzed four freshly prepared replicates of each validation sample on four different days. The content of coumarin in the validation samples was quantified with comparison to a dilution curve made from a different stock solution in each series. The standard deviation of repeatability (RSDr) and intra-laboratory reproducibility (RSD_{IR}) were generated in accordance with the International Organization for Standardization (ISO) 5725 (ISO, 1994). Results showed a precision with RSDr values between 0.8% and 1.6%, and RSD_{IR} with values between 4.7% and 18.0% as shown in Table 1.

2.3.3. Limit of detection (LOD) and limit of quantification (LOQ)

A breakfast cereal (analogously to the VS1 used in the precision study, Section 2.3.2) consisting of rolled oats, dried banana, raisins, and cinnamon was homogenized. On four individual days, four subsamples were freshly prepared and analyzed. LOD was then calculated as 3 × the relative standard deviation (RSD_{IR}) and LOQ was calculated as 6 × RSD_{IR} of the measured concentration. An RSD_{IR} of 18.0% corresponds to a LOD of 0.5 and a LOQ of 1.0 mg coumarin/kg.

2.3.4. Recovery samples

Recovery samples (RS) consisting of two breakfast cereals (RS1 and RS2), plain wheat bread (RS3), and tea (RS4) from the local supermarket were tested negative for their coumarin content (data not shown). Recovery samples were homogenized and spiked to 25 mg coumarin/kg. Recovery sample 1 was analyzed in duplicate

Table 1
Results from the precision study of validation samples VS1, VS2, and VS3.

Validation parameter	Validation sample		
	VS1	VS2	VS3
Number of assays[a]	4	4	4
Mean coumarin content (mg/kg)	1.0	15.4	48.6
RSDr (%)	1.6	1.0	0.8
RSD_{IR} (%)	18.0	7.1	4.7

[a] Each assay included four subsamples. RSD is relative standard deviation, r is repeatability, IR is intra reproducibility.

on four individual days. Two subsamples of RS2, RS3, and RS4 were analyzed once in duplicate. The analyzed RS1 showed a mean result of 25.3 mg/kg (SD 0.76) corresponding to a recovery of 101%. The analyzed RS2 showed a mean result of 21.8 mg/kg (SD 0.27) corresponding to a recovery of 87%. The analyzed RS3 showed a mean result of 22.1 mg/kg (SD 0.49) corresponding to a recovery of 87%. The analyzed RS4 showed a mean result of 26.5 mg/l (SD 0.07) corresponding to a recovery of 104%.

2.4. Statistics and interpretation of results

In addition to coumarin, all dilutions and sample extracts contained 20 mg/l of MUM. The retention times of MUM and coumarin were used to calculate a retention time (RT) ratio that supported the correct identification of coumarin in samples. Evaluation of numerous RT ratios (data not shown) has shown a variation of the RT ratio of a maximum of 0.03 from any sample to any dilution within one series of analysis. An RT ratio of $\leq$0.03 was, therefore, used as a criterion for correct identification of coumarin. In addition, a characteristic UV profile obtained from 240 to 340 nm supported correct identification.

All samples were first analyzed in single determinations. In case the content of coumarin exceeded the EU limit (European Parliament and Council, 2008), samples were re-analyzed in replicates of two (two separate portions of the same homogenate). In this case, the mean ($\bar{x}$) of all three results were reported. The SD from the three results was used to calculate the confidence intervals (CI). The CI (95%) was calculated as: $\bar{x} \pm t(s/\sqrt{n})$, where t is a value from the t distribution, s is the standard deviation, and n is the number of samples. Results were classified as exceeding the EU limits for coumarin if the mean result included the CI exceeds the limits (European Parliament and Council, 2008). In other cases, samples were classified as acceptable. Data were statistically treated (Miller & Miller, 1993) with the Microsoft® Excel 2000 software and evaluated according to standardized criteria (ISO, 1994).

2.5. Food samples

A total of 74 food samples were collected as a part of a governmental inspection program at the DVFA. The program involved samples labeled with cinnamon from local bakery stores, supermarket storage facilities, and directly from importers. Sample collection was distributed equally throughout Denmark, and included Danish and non-Danish (Belgium, Germany, The Lebanese Republic, The Netherlands, Poland, Spain, Sweden) food products labeled with cinnamon. Samples (n) were included from the categories of traditional and/or seasonal bakery ware ($n = 18$), breakfast cereals ($n = 4$), fine bakery ware ($n = 35$), desserts ($n = 3$), crisp bread (2) and tea ($n = 12$). The distinction between the categories of traditional and/or seasonal bakery ware and fine bakery ware is not specified in the current EU regulation (European Parliament and Council, 2008). At the DVFA, bakery ware readily available to consumers all year round and not part of a traditional custom is placed in the category of fine bakery ware, and includes cinnamon bread and cinnamon rolls (buns). Bakery ware that is not normally consumed on a regular basis but consumed as a part of a tradition and/or season includes traditional Christmas cookies, such as the Danish brunkager and pebernødder. The DVFA regard the high maximum limit (50 mg/kg) for traditional and/or seasonal bakery ware justified in the limited consumption of bakery ware in this category. The few commercially available desserts, breakfast cereals and crisp bread that contained cinnamon, limited the number of samples collected in these categories. Samples were collected from December 2012 through February 2013, and subjected to analysis from January through March 2013. Upon arrival at the laboratory, samples were homogenized and frozen at −18 °C until further analysis.

2.6. Analytical procedure

From tea leaves, a concoction was prepared as prescribed on the tea package. Depending on the prescription, 200–1000 ml of 80–100 °C hot water was added to 5.9–20.1 g of homogenized tea ingredients pr. liter and incubated for 3–6 min prior to filtration. A sample of pudding powder was added water and prepared as described on the package label. Similar to the other food products, coumarin was extracted from the filtered tea concoctions and the prepared pudding.

A slight modified extraction procedure of Scotter and Rees (2010) was used. In brief, 5.00 g of a homogenized sample was placed in a centrifugation tube and added 1000 µl of MUM with a concentration of 1000 mg/l in 80% of methanol and 20 ml of 90% methanol prior to mixing with ultra turrex for 30 s. The mixture was shaken for 30 min and centrifuged for 5 min at 3500 RCF. The supernatant was transferred to a 50.0 ml volumetric flask. Five ml of 90% methanol in water was added to the centrifugation tube and 30 s on a whirl mixer re-dissolved the pellet. Another round of centrifugation was performed and the supernatant was added to the volumetric flask. Water was added to the volumetric flasks to a total volume of 50.0 ml. After shaking, an adequate amount of the extracted sample was filtered through a 0.2 µl PVDF filter directly into HPLC vials. The sample content of coumarin was quantified with comparison to a dilution curve consisting of six dilutions of coumarin with concentrations of 0.1, 0.5, 1.0, 2.0, 5.0, and 10.0 mg/l (corresponding to 10 × higher levels in samples, mg/kg). The dilutions were prepared in water from a stock solution of 1000 mg coumarin/l in 80% methanol. In each analytical series, two samples of homogenized rolled oatmeal were spiked to 25 mg/kg prior to extraction and included for quality assurance purposes.

3. Results and discussion

3.1. Food samples

Coumarin was identified at a retention time around 1.58 min. The RT ratio between MUM and coumarin (Fig. 1) was evaluated and varied with no more than 0.03 from any dilution to any sample. In addition, also the UV profiles (Fig. 2) were compared. In all samples, the UV profile of coumarin was consistent with literature data (Maggi, Barboni, et al., 2011) showing a global maximum at 278.1 nm

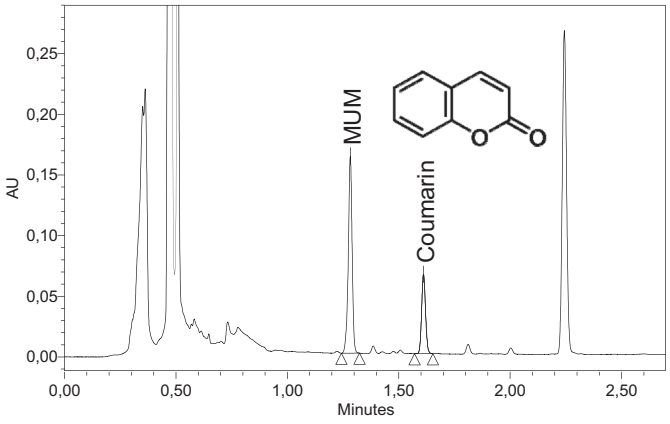

Fig. 1. A chromatogram from a cinnamon roll with a content of about 25 mg coumarin/kg; obtained at a wavelength of 278.1 nm.

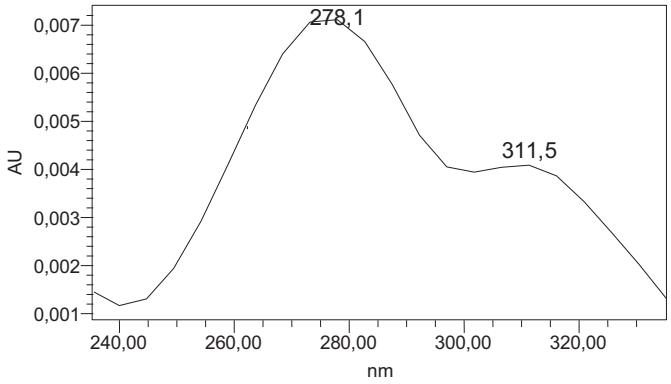

Fig. 2. The UV profile (240–340 nm) obtained from coumarin eluted at 1.58 min (see Fig. 1).

and a local maximum at around 311 nm (the local maximum was present in all samples but a value was not always assigned) (Fig. 2).

A total of 74 food samples were analyzed in a governmental inspection program at the DVFA and compared to the EU limits for coumarin (European Parliament and Council, 2008) (Fig. 3). In the category of traditional and/or seasonal bakery ware, 18 samples were analyzed and showed a content of coumarin ranging between 3.8 and 35.0 mg/kg with a mean coumarin content of 19.5 mg/kg. In the category of breakfast cereals, 4 samples were analyzed and showed a content of coumarin ranging between 0.9 and 10.0 mg/kg with a mean coumarin content of 3.3 mg/kg. In the category of fine bakery ware, 35 samples were analyzed and showed a content of coumarin ranging between 0.4 and 53.4 mg/kg with a mean coumarin content of 16.5 mg/kg. In this category, 17 samples (49%) exceeded the EU limits for coumarin (European Parliament and Council, 2008). One sample contained 53.4 mg coumarin/kg and, thereby, exceeding the EU limit with more than a factor of three. In the category of desserts, 3 samples were analyzed and showed a content of coumarin ranging between 1.0 and 5.1 mg/kg with a mean coumarin content of 3.3 mg/kg. One sample showed a coumarin content (5.1 mg/kg) close to the EU limit (European Parliament and Council, 2008) and was therefore re-analysed. The confidence interval (95%) was 5.1 ± 0.4 mg/kg. The sample did therefore not exceed the EU limit for coumarin; see Section 2.4 for statistics and interpretation of results. It should be stressed that other EU member states could have categorized the food samples differently and, therefore, obtained another degree of compliance.

With the exception of cinnamon rolls (buns) from Sweden, all products that exceeded the EU limit for coumarin were placed in the category of fine bakery ware, were manufactured in Denmark, and collected at Danish bakery stores. Common products in this category were cinnamon rolls (buns) and cinnamon bread. The high level of coumarin in bakery ware is perhaps not an isolated Danish problem; others have also reported high levels of coumarin in bakery ware (eAGRI, 2010; Sproll et al., 2008).

Crisp bread and tea was also subjected to analysis despite the lack of EU limits for coumarin in these food products. Two samples of crisp bread with cinnamon were analyzed and showed a content of coumarin ranging from 16.0 to 23.0 mg/kg with a mean coumarin content of 19.5 mg/kg. Despite this relative high amount of coumarin, crisp bread does probably not contribute significantly to the coumarin intake because of the low weight of crisp bread. Twelve samples of tea (ready to drink) with cinnamon were analyzed and showed a content of coumarin ranging from 0 to 12.0 mg/kg with a mean coumarin content of 2.2 mg/kg. Most of the teas were labeled as chai tea. The coumarin content is less compared to crisp bread, but the normal intake of tea might be larger. In this context, not only the possible exceedance of the TDI may be relevant but also the higher bioavailability of coumarin in the case of a fluid matrix, especially, if the peak level is the critical parameter triggering the toxic effect (what is currently unknown) (Abraham, Pfister, Wöhrlin, & Lampen, 2011). The limited number of samples within tea, breakfast cereals, and desserts disables a valid comparison of their coumarin content. Nonetheless, a daily intake of tea, dessert, and breakfast cereals could be within the approximate same quantity. It could, therefore, be interesting to investigate the coumarin content in a larger number of tea samples to evaluate the need for an EU limit for coumarin in tea.

4. Addendum

4.1. Advisory amounts of cassia in food products

Small bakery stores could have difficulties in keeping up with the relevant EU regulations, and might therefore be unaware of the EU limits for coumarin (European Parliament and Council, 2008). However, even knowledge about the EU limits might not answer the question on how much cinnamon that can be added to bakery ware without exceeding the EU limits for coumarin (European Parliament and Council, 2008). We, therefore, find it appropriate to provide food manufacturers with advisory amounts of cassia that can be added to food products without exceeding the EU limits for coumarin. The following describes how the advisory amounts are calculated.

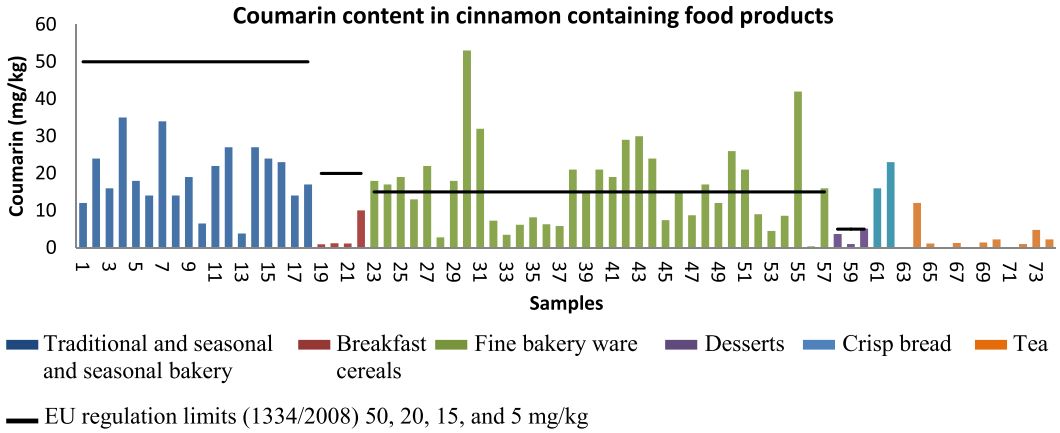

Fig. 3. The content of coumarin in food products compared to the European Regulation (EC) No 1334/2008.

Investigations of the coumarin content in commercially available ground cinnamon samples are reported to vary from 5 to 7670 mg/kg (Blahova & Svobodova, 2012; Lungarini et al., 2008; Woehrlin et al., 2010). However, coumarin contents below 100 mg/kg were considered to originate from *Cinnamomum verum* (Lungarini et al., 2008) and are, therefore, not included in the following calculations. Based on 29 ground cinnamon samples collected in the EU (Blahova & Svobodova, 2012; Lungarini et al., 2008; VKM, 2010), the average content was calculated to be 2793 mg/kg (median, 2636 mg/kg) with a standard deviation of 1333 mg/kg. Assuming that the coumarin content is normally distributed in ground cinnamon samples, 99% of all samples will contain $\mu \pm 2.58\sigma$. μ is an estimate of the true value and σ is the standard deviation. 99% of all samples are, therefore, assumed to be within the range of $2793 \pm (2.58 \times 1333)$ mg/kg with a corresponding maximum content of 6300 mg/kg (rounded up). With this theoretically based maximum content of coumarin in ground cinnamon, we present a practical guide that describes the amount of cassia that can be added to food products without exceeding the EU limits for coumarin in 99% of the cases, see Table 2. The DVFA has published a similar practical guide (Table 2) at their official home page (http://www.foedevarestyrelsen.dk/english/Pages/default.aspx). Furthermore, the Confederation of Danish Industry (DI) approved the practical guide and distributed it among their relevant members.

5. Conclusion

The present UPLC method is fast and suitable for routine testing of coumarin in food products with cinnamon. In this study, the presence of cinnamon in fine bakery ware has resulted in several cases of a coumarin content exceeding the EU limit (European Parliament and Council, 2008). The reason for the high incidence of samples exceeding the EU limit for coumarin in bakery ware is unknown. However, a lack of knowledge about the EU regulation (European Parliament and Council, 2008) could be one explanation. Another explanation could be that food manufacturers of bakery ware are unaware of the cinnamon amounts that can be added without exceeding the EU limits for coumarin. To comply with the EU limits for coumarin, manufacturers of bakery ware with cinnamon are advised to have their cinnamon analyzed for the coumarin content. The coumarin content can then guide the maximum amount of cinnamon that can be added without exceeding the EU limits for coumarin. Another possibility is to inform the food manufacturers of bakery ware about the theoretical amount of cinnamon that can be added to food products without exceeding the EU limits for coumarin. We have made a practical guide that manufacturers of food with cinnamon can use to theoretically comply with the EU regulation (European Parliament and Council, 2008). National food administrations are advised to communicate these possibilities to the food manufacturers of bakery ware in order to limit further exceedances of the EU limits for coumarin. A different strategy is to add true cinnamon to food products instead of cassia; however, this will alter the perception of taste.

Tea is not included in the EU regulation (European Parliament and Council, 2008). However, tea is an important consumable for many people and might contribute to the overall daily consumption of coumarin. In 2004, the European Food Safety Authority (EFSA) recommended a TDI of 0–0.1 mg coumarin/kg body weight (EFSA, 2004). Children represent a sensitive group because of a low body weight and a dietary habit towards sweet food with cinnamon. A child of 30 kg that consumes 250 ml of tea with a content of 12 mg coumarin/l has reached this TDI. In this case, all the TDI of coumarin is allocated to tea, which could indicate that tea might have a problematic high content of coumarin. It could, therefore, be interesting to gather more data on the coumarin content in tea, to consider its inclusion in a future EU regulation.

Table 2
A practical guide that shows the theoretical amount of ground cassia that can be added to food products, without exceeding the EU limits for coumarin (European Parliament and Council, 2008).

Food category	Theoretical maximum amount of cassia (g/kg food)[a]	Corresponding coumarin content (mg/kg) that matches the EU limits (European Parliament and Council, 2008)
Traditional and/or seasonal bakery	7.94	50
Breakfast cereals	3.17	20
Fine bakery ware	2.38	15
Dessert	0.79	5

[a] Calculated from the average coumarin content (2793 mg/kg) in 29 samples of ground cinnamon (Blahova & Svobodova, 2012; VKM, 2010; Lungarini et al., 2008) added 2.58×SD (1333 mg/kg). Statistically, these maximum amounts of cassia will ensure that the coumarin content does not exceed the EU limits (European Parliament and Council, 2008) in 99% of the cases.

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References

Abraham, K., Pfister, M., Wöhrlin, F., & Lampen, A. (2011). Relative bioavailability of coumarin from cinnamon and cinnamon-containing foods compared to isolated coumarin: a four-way crossover study in human volunteers. *Molecular Nutrition & Food Research, 55*, 644–653.

Abraham, K., Wöhrlin, F., Lindtner, O., Heinemeyer, G., & Lampen, A. (2010). Toxicology and risk assessment of coumarin: focus on human data. *Molecular Nutrition & Food Research, 54*, 228–239.

eAGRI. (2010). *The bulletin – Results of monitoring and evaluation of toxic compounds in food chain in agriculture section in 2009*. Praha, Czech Republic: Ministry of Agriculture of the Czech Republic website. Ministry of Agriculture.

BfR. (2006). *High daily intakes of cinnamon: Health risk cannot be ruled out*. Health Assessment No. 044/2006.

Blahova, J., & Svobodova, Z. (2012). Assessment of coumarin levels in ground cinnamon available in the Czech retail market. *Scientific World Journal, 2012*, 263851.

EFSA. (2004). Opinion of the scientific panel on food additives, flavourings, processing aids and materials in contact with food (AFC) related to Coumarin. *The EFSA Journal, 104*, 1–36.

European Council. (1988). Council Directive of 22 June 1988 on the approximation of the laws of the member states relating to flavourings for use in foodstuffs and to source materials for their production. *Official Journal of the European Communities*1–10, 88/388/EEC.

European Parliament and Council. (2008). Regulation (EC) no 1334/2008 of the European Parliament and of the council of 16 December 2008 on flavourings and certain food ingredients with flavouring properties for use in and on foods and amending council regulation (EEC) no 1601/91, regulations (EC) no 2232/96 and (EC) no 110/2008 and directive 2000/13/EC. *Official Journal of the European Communities, L354*, 34–50.

He, Z. D., Qiao, C. F., Han, Q. B., Cheng, C. L., Xu, H. X., Jiang, R. W., et al. (2005). Authentication and quantitative analysis on the chemical profile of cassia bark (cortex cinnamomi) by high-pressure liquid chromatography. *Journal of Agricultural and Food Chemistry, 53*, 2424–2428.

ISO. (1994). *ISO 5725–2. Accuracy (trueness and precision) of measurement methods and results – Part 2: Basic method for the determination of repeatability and reproducibility of a standard measurement method*. International Organization for Standardization.

Lake, B. G. (1999). Coumarin metabolism, toxicity and carcinogenicity: relevance for human risk assessment. *Food and Chemical Toxicology, 37*, 423–453.

Lungarini, S., Aureli, F., & Coni, E. (2008). Coumarin and cinnamaldehyde in cinnamon marketed in Italy: a natural chemical hazard? *Food Additives & Contaminants. Part A: Chemistry, Analysis, Control, Exposure & Risk Assessment, 25*, 1297–1305.

Maggi, F., Barboni, L., Caprioli, G., Papa, F., Ricciutelli, M., Sagratini, G., et al. (2011). HPLC quantification of coumarin in bastard balm (*Melittis melissophyllum* L., Lamiaceae). *Fitoterapia, 82*, 1215–1221.

Maggi, F., Martonfi, P., Conti, F., Cristalli, G., Papa, F., Sagratini, G., et al. (2011). Volatile components of whole and different plant parts of bastard balm (*Melittis melissophyllum* L., Lamiaceae) collected in Central Italy and Slovakia. *Chemistry and Biodiversity, 8*, 2057–2079.

Martino, E., Ramaiola, I., Urbano, M., Bracco, F., & Collina, S. (2006). Microwave-assisted extraction of coumarin and related compounds from Melilotus officinalis (L.) Pallas as an alternative to Soxhlet and ultrasound-assisted extraction. *Journal of Chromatography A, 1125*, 147–151.

Miller, J. C., & Miller, J. N. (1993). *Statistics for analytical chemistry* (3rd ed.). West Sussex, Great Britain: Ellis Horwood Limited.

Miller, K. G., Poole, C. F., & Chichila, T. M. P. (1995). Solvent-assisted supercritical fluid extraction for the isolation of semivolatile flavor compounds from the cinnamons of commerce and their separation by series-coupled column gas chromatography. *Journal of High Resolution Chromatography, 18*, 461–471.

Miller, K. G., Poole, C. F., & Pawloski, T. M. P. (1996). Classification of the botanical origin of cinnamon by solid-phase microextraction and gas chromatography. *Chromatographia, 42*, 639–646.

Ochiai, N., Sasamoto, K., Hoffmann, A., & Okanoya, K. (2012). Full evaporation dynamic headspace and gas chromatography-mass spectrometry for uniform enrichment of odor compounds in aqueous samples. *Journal of Chromatography A, 1240*, 59–68.

Ravindran, P. N., Nirmal-Babu, K., & Shylaja, M. (2004). Introduction. In *Cinnamon and Cassia: The genus Cinnamomum* (pp. 1–13). Boca Raton: CRC Press.

Scotter, M., & Rees, G. (2010). *Development of methods to quantitatively extract biologically active principles from complex foods, flavourings and herbs and spices, to allow their subsequent analysis* (Rep. No. FD 10/01; FERA project No. R6NL). York YO41 1LZ(UK): DEFRA Food and Environment Research Agency.

Sproll, C., Ruge, W., Andlauer, C., Godelmann, R., & Lachenmeier, D. W. (2008). HPLC analysis and safety assessment of coumarin in foods. *Food Chemistry, 109*, 462–469.

VKM. (2010). *Risk assessment of coumarin intake in the Norwegian population – Opinion of the panel on food additives, flavourings, processing aids, materials in contact with food and cosmetics of the Norwegian scientific committee for food safety* (Rep. No. 09/405–2 final). Norway: Norwegian Scientific Committee for Food Safety.

Wang, Y. H., Avula, B., Nanayakkara, N. P. D., Zhao, J., & Khan, I. A. (2013). Cassia cinnamon as a source of coumarin in cinnamon-flavored food and food supplements in the United States. *Journal of Agricultural and Food Chemistry, 61*, 4470–4476.

WHO. (1999). *Cortex Cinnamomi. WHO monographs on selected medicinal plants*. In *WHO monographs* (pp. 95–104).

Woehrlin, F., Fry, H., Abraham, K., & Preiss-Weigert, A. (2010). Quantification of flavoring constituents in cinnamon: high variation of coumarin in cassia bark from the German retail market and in authentic samples from Indonesia. *Journal of Agricultural and Food Chemistry, 58*, 10568–10575.